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An assessment of the role of the North Atlantic as a CO_2 sink

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SUMMARY

A numerical interpolation scheme based upon the lateral diffusive and advective transport of ocean surface waters has been developed to interpolate measurements made in irregular time and space over the oceans. This has been applied to about 2700 surface ocean pCO_2 measurements observed during the period 1972–1992 in the North Atlantic to give the distribution of sea–air pCO_2 difference (ΔpCO_2) over the Atlantic. Although the atmospheric CO_2 concentration has increased by about 28 ppm over this period, the pCO_2 values in the surface waters of subarctic regions have increased little because they are dictated primarily by the properties of underlying deep waters through vertical mixing. Accordingly, ΔpCO_2 values measured north of 50° N have been corrected to the year 1990 using the secular increase of atmospheric CO_2. Because the surface water pCO_2 value in temperate waters tracks the secular increase in atmospheric CO_2 with a time lag of about two years, no correction was applied to the warm water ΔpCO_2 data. It has been assumed that seasonal variations are the same for each year. The net CO_2 flux across the sea surface has been computed over a 4° latitude × 5° longitude grid using the mean monthly ΔpCO_2 values and the gas transfer coefficients estimated using the mean monthly wind speed. It has been found that the areas of the high latitude North Atlantic and the adjacent seas north of 42° N are net CO_2 sinks of 0.2 to 0.5 Gt C a^{-1}. The total sink flux of CO_2 over the temperate North Atlantic areas between 18° N and 42° N is balanced approximately by the source flux over the tropical Atlantic between 18° N and 18° S.

1. INTRODUCTION

The atmospheric CO_2 level has been increasing at a rate about one-half of that which is expected from industrial CO_2 emissions. The respective roles of the oceans and terrestrial biosphere as major sinks for the missing CO_2 have been debated extensively yielding a wide range of estimates (Siegenthaler & Sarmiento 1993). Using simple one-dimensional box-diffusion ocean models calibrated with ^{14}C distribution in the atmosphere and oceans, the annual oceanic uptake of CO_2 has been estimated to be about 2 Gt (Oeschger *et al.* 1975; Broecker & Peng 1982). Similar results were obtained when three-dimensional ocean general circulation models were used (Bacastow & Reimer-Meier 1990; Sarmiento *et al.* 1992). On the basis of distribution and temporal changes of ^{13}C in atmospheric and oceanic CO_2, the annual oceanic uptake of CO_2 has also been estimated to be 2.1 ± 0.8 Gt (Quay *et al.* 1992; Tans *et al.* 1993). Tans *et al.* (1990), however, combined the meridional gradient of atmospheric CO_2 concentration and the net CO_2 uptake flux over the northern oceans, which was computed using the observed sea–air pCO_2 difference and gas transfer coefficient, and estimated the global oceanic CO_2 uptake to be less than 1 Gt a^{-1}.

In this paper, a numerical method for interpolating the pCO_2 measurements made in surface waters, at scattered locations and irregular time intervals over the North Atlantic is presented. This method is based on lateral transport by advection and diffusive mixing of surface waters; developed to obtain an improved estimate for time–space distribution of the sea–air pCO_2 difference (ΔpCO_2) and the net CO_2 flux across the sea surface. Because a limited number of observations are available, the interannual variability is neglected; all data obtained between 1972–1992 are assumed to represent variations within the reference year of 1990 chosen arbitrary for this study. Corrections were made to normalize the measured ΔpCO_2 values to conform to the 1990 atmospheric CO_2 concentration. The resulting monthly distribution of ΔpCO_2, which implicitly includes the effects of ocean circulation and biology, has been combined with the mean monthly wind speed data to estimate the net CO_2 flux over the North Atlantic Ocean in 1990.

2. DATA AND METHODS OF OBSERVATIONS

The principal source of data for this study is the pCO_2 measurements in mixed layer made by the Lamont–Doherty Earth Observatory (LDEO) group (Broecker *et al.* 1979; Takahashi *et al.* 1982; Takahashi *et al.*

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1985; Peng *et al.* 1987; Tans *et al.* 1990; Chipman *et al.* 1993). This has been supplemented by the measurements by Weiss *et al.* (1992), February–April 1982 in the northern North Atlantic and Norwegian–Greenland Seas; by Roos & Gravenhorst (1984) in the North Atlantic; and by Andrie *et al.* (1986) in the equatorial Atlantic. Figure 1 shows the distribution of about 2700 locations where measurements were made during 1972 through 1992. The $p\mathrm{CO_2}$ data used in this paper were obtained by gas–water equilibration methods combined with an infrared analyser or gas chromatograph for CO_2 analyses. These methods are described by Broecker & Takahashi (1966) for continuous underway measurements and by Chipman *et al.* (1993) for measurements with discrete samples. Modified designs for underway measurements have been used by Weiss *et al.* (1992) and other investigators. All the $p\mathrm{CO_2}$ measurements obtained by the LDEO group and by Weiss *et al.* (1992) are calibrated against the manometric CO_2 standards of C. D. Keeling, Scripps Institution of Oceanography. To test the compatibility of data sets obtained by different investigators, the LDEO data obtained during the TTO/NAS expeditions have been compared with those obtained concurrently by Weiss *et al.* (1992). Over a $p\mathrm{CO_2}$ range between 180 µatm and 350 µatm, the Weiss data have been found to be consistent with the LDEO data with a r.m.s.d. of ± 7.5 µatm for 137 pairs of comparison or a standard deviation of the mean of ± 0.7 µatm $(= 7.5/137^{-\frac{1}{2}})$.

Because of serious systematic errors associated with $p\mathrm{CO_2}$ values computed using pH, alkalinity and/or $\mathrm{TCO_2}$ measurements, only the results of direct $p\mathrm{CO_2}$ measurements are used in this paper.

(*a*) *Computation of sea–air $p\mathrm{CO_2}$ difference*

The net flux (F) of CO_2 across the sea surface may be expressed by the relation:

$$F = k \times \Delta p\mathrm{CO_2}, \qquad (1)$$

where k is the CO_2 gas transfer coefficient and $\Delta p\mathrm{CO_2}$ is the difference in the CO_2 partial pressure in surface mixed layer and that in the overlying atmosphere. The gas transfer coefficient is often parameterized as a function of wind speed, for lack of more appropriate physical parameters measurable over the global ocean. The $p\mathrm{CO_2}$ value measured in the bulk water is used to represent that for the ocean surface. Here, the $\Delta p\mathrm{CO_2}$ has been computed by:

$$\Delta p\mathrm{CO_2} = (p\mathrm{CO_2})_{\mathrm{sw}} - (p\mathrm{CO_2})_{\mathrm{air}}, \qquad (2)$$

where $(p\mathrm{CO_2})_{\mathrm{sw}} = (X\mathrm{co_2})_{\mathrm{deq}}[(Pt)_{\mathrm{eq}} - (Pw)_{\mathrm{eq}}]$,

or $(p\mathrm{CO_2})_{\mathrm{sw}} = (X\mathrm{co_2})_{\mathrm{weq}}(Pt)_{\mathrm{eq}}$,

and $(p\mathrm{CO_2})_{\mathrm{air}} = (X\mathrm{co_2})_{\mathrm{dair}}[Pb - (Pw)_{\mathrm{sw}}]$.

The two alternative expressions for $(p\mathrm{CO_2})_{\mathrm{sw}}$ are given for cases when the equilibrated gas samples were analysed with or without water vapor. The subscripts 'sw' and 'air' represent quantities in seawater and air respectively; $X\mathrm{co_2}$, the mole fraction concentration of CO_2 in dry marine air (subscript 'dair') or that in dried and moist equilibrated air (subscripts 'deq' and 'weq') respectively; $(Pt)_{\mathrm{eq}}$, the total gas pressure during equilibration; Pw, the vapor pressure of water at equilibration or *in situ* seawater temperature (subscripts 'eq' and 'sw') respectively; and Pb, the barometric pressure at sea surface. In the concentration range of CO_2, the non-ideal behaviour of CO_2 gas due to CO_2–CO_2 as well as CO_2–N_2–O_2–H_2O molecular interactions is small and partly cancels due to the differencing for $\Delta p\mathrm{CO_2}$. Therefore, CO_2 has been treated as an ideal gas.

The total pressure under which CO_2 in the carrier gas is equilibrated with seawater is needed to define the $p\mathrm{CO_2}$ in seawater. Once it is determined, it does not depend on the barometric pressure since the $p\mathrm{CO_2}$ in seawater is an intrinsic property of the seawater. However, $p\mathrm{CO_2}$ in air varies with barometric pressure even though the mole fraction concentration of CO_2 remains constant. Considering the timescale for CO_2 exchange in surface ocean waters to be about one year, we have chosen a mean barometric pressure (Pb) of 1.000 atm for the computation of $(p\mathrm{CO_2})_{\mathrm{air}}$. Measured pressures of equilibration ($(Pt)_{\mathrm{eq}}$) were used for the computation of $(p\mathrm{CO_2})_{\mathrm{sw}}$, but when $(Pt)_{\mathrm{eq}}$ measurements are not available, it has been assumed to be 1 atm.

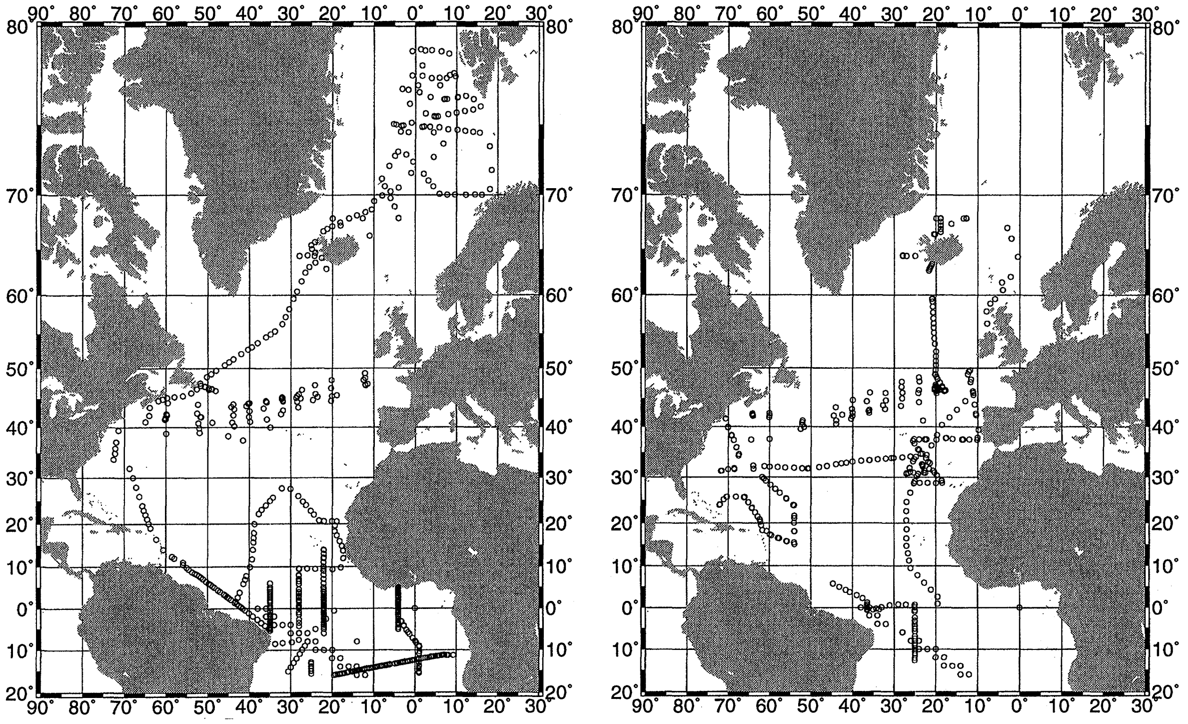
(*b*) *Correction of the observed $\Delta p\mathrm{CO_2}$ to the year 1990*

From 1972 to 1992, the atmospheric CO_2 concentration increased by about 28 ppm, from about 326 ppm to 354 ppm. The magnitude of this increase is significant in comparison with that for $\Delta p\mathrm{CO_2}$, which range between +60 and −120 µatm. Therefore, the secular change in atmospheric CO_2 must be taken into consideration when $\Delta p\mathrm{CO_2}$ data are assembled to represent a single year. We consider two oceanographic provinces separately, the warm water and cold water regime, where vertical mixing is slow and rapid respectively. Measurements of surface water $p\mathrm{CO_2}$ made over the Sargasso Sea in the North Atlantic during the 1957-IGY (Takahashi 1961) 1972–73-GEOSECS and 1981-TTO/NAS programs (Takahashi *et al.* 1982) suggests that $p\mathrm{CO_2}$ in the mixed layer water increased with a similar rate as that in the atmosphere with a time lag of about two years (Takahashi *et al.* 1983). This suggests that, in temperate gyre areas, where the warm upper layers do not mix fast with deep waters across sharp density gradients, $\Delta p\mathrm{CO_2}$ values in the mixed layer are nearly independent of time of measurement. Thus those measured in different years may be treated independent of time without introducing serious systematic errors.

In contrast to the warm water regime, the surface layers in high latitude oceans are known to mix with a large volume of deep waters seasonally. Thus, the $p\mathrm{CO_2}$ in surface waters reflects primarily the characteristics of deep waters, in which anthropogenic effects are diluted to undetectable levels. A comparison between the 1974–79 observations at the Weather Station 'P' in the northeastern subarctic Pacific by Wong & Chan

January - March

April - June



July - September

October - December

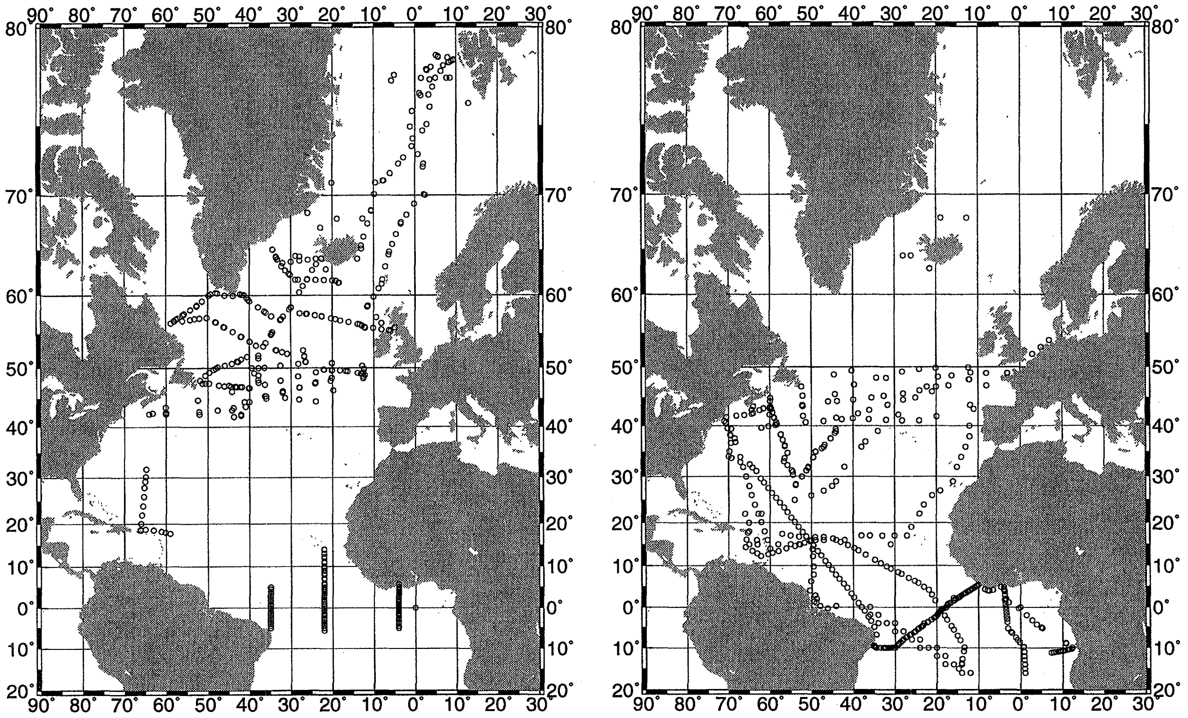


Figure 1. Station locations where pCO_2 measurements were made between 1972 and 1992. At each of about 2700 stations shown, the temperature, salinity, pCO_2 and often the concentrations of total CO_2 and nutrients and alkalinity were measured.

(1991) and the 1985–89 observations by the LDEO group (Takahashi *et al.* 1991, 1993) suggest that the oceanic pCO_2 had not changed measurably over the 10 year period during a time when atmospheric pCO_2 increased steadily. Accordingly, ΔpCO_2 should have changed as the atmospheric CO_2 concentration increased with time. Because relevant observations are not yet available in the North Atlantic and its adjacent seas, we consider two cases. First, those areas north of the North Atlantic Drift may be assumed to be similar to the subarctic North Pacific. Therefore, we corrected $\Delta p\mathrm{CO}_2$ values measured north of 50° N by subtracting the atmospheric $p\mathrm{CO}_2$ increased between the date of measurements and January 1 1990. The $\Delta p\mathrm{CO}_2$ values thus corrected have been used to compute those in the (a) lines in table 1. Second, because high latitude Atlantic surface waters may have taken up some atmospheric CO_2 during their recirculation through the North Atlantic and adjacent seas, the oceanic $p\mathrm{CO}_2$ values in the areas north of 50° N may be assumed to have increased with time with one half the atmospheric CO_2 increase rate. The $\Delta p\mathrm{CO}_2$ values thus corrected have been used to compute those in the (b) lines in table 1. Because seasonal variation in atmospheric CO_2 concentration is already represented in the observed $\Delta p\mathrm{CO}_2$ value by the concurrent measurements of atmospheric and seawater $p\mathrm{CO}_2$, only the effect of the secular increase is considered for the correction. A three year running mean of the atmospheric CO_2 data obtained at Mauna Loa by Keeling & Whorf (1991) is used to obtain the secular atmospheric trend. It gives the 1990 Mauna Loa value of 353.95 ppm and is corrected additionally for a mean meridional difference of 0.7 ppm between 20° N and 50° N.

3. COMPUTATIONAL METHOD

A computational method is needed to calculate the distribution of properties over the North Atlantic Ocean using the observations made at locations in time and space dictated by ship tracks without mathematical regularity. For the purpose of interpolating the observations made in varying space and time, a finite-difference algorithm involving lateral diffusion and advection of surface waters has been developed.

(*a*) *Mathematical framework*

The concentration of properties throughout the oceans is governed by eddy diffusion, advective transport, internal sources and sinks and exchange with the surroundings at sea floors, ocean margins and the sea–air interface. For the purposes of this analysis, which is to provide a uniform and consistent interpolation process for surface mixed-layer properties, internal and boundary sources and sinks are considered negligible. Contributions of CO_2 due to the sea–air exchange and biological activity are included in the sampled observational data and need not be included as an explicit source or sink term in the interpolation model. This same rationale may be applied to separate surface mixed-layer concentrations from waters below. Whatever vertical transport exists, the results of such mixing are embedded in the observed data and need not be modelled separately.

Under these assumptions, the short-term behaviour of surface mixed-layer waters may be approximated using a two dimensional model consisting of diffusive transport and surface water advection. The predicted concentrations are continuous in space and time with respect to observation. The transport equation in two dimensions for a scalar quantity, S, may be written as:

$$\mathrm{d}s/\mathrm{d}t = K\,\nabla^2 S - (\partial S/\partial x\ Vx + \partial S/\partial y\ Vy), \qquad (3)$$

where $\nabla^2 S = \partial^2 S/\partial x^2 + \partial^2 S/\partial y^2$, and K = eddy diffusivity.

In light of the sparseness of the observational data over large areas of the ocean (see figure 1), this equation will be discretized onto a 5 ° longitude by 4 ° latitude spatial grid. Interannual variations are assumed negligible for the purposes of this model. Observational data collected over a twenty year period are mapped onto a single, virtual calendar year. The computations preserve continuity of solutions across the December/January border. The solution is considered fully converged when the transport equation is satisfied over the entire computational domain. The land–sea interface is considered to be an impenetrable boundary

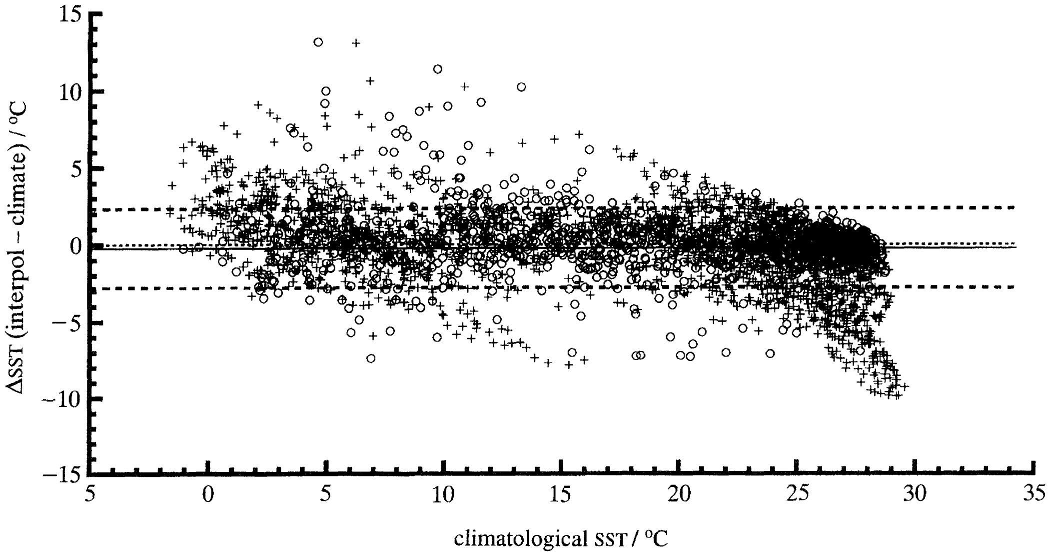


Figure 2. Comparison between the climatological sea surface temperature data (Shea *et al.* 1992) and those computed using the scheme of this paper with the measurements located in figure 1. The solid line denotes the mean value (-0.24 ± 2.55, $n = 3376$). The 'o' symbols indicate the observed SST values and the '+' symbols indicate the SST values estimated for node points without observations.

where the gradient of the scalar field is zero across the normal surface.

The time increment must be chosen to retain numerical stability under both diffusion and advection and to provide physical relevance. A time increment, $dt = 1$ day is chosen. Because the system stability is governed by advective terms, $V_{max} \ll dx/dt$ should be satisfied, where dx is the internode distance and V_{max} is the maximum advective speed. For example, 5° longitude increments at 60° N latitude gives $dx = 277800$ m, $V_{max} \ll dx/dt = 3.2 \text{ m s}^{-1}$ or 6 knots; and at 30° N, $V_{max} \ll 5.6 \text{ m s}^{-1}$ or 11 knots. Because ocean current velocities are generally slower than these values, a one day time step is adequate for convergence.

The finite-difference algorithm uses central differences in space and averages a forward-centered time step about $(t-1)$ with a backward-centred time step about $(t+1)$ to create a recursion relation symmetric in both space and time: $\partial S/\partial t = (S^t - S^{t-1})/\Delta t$ and $\partial S/\partial t = (S^{t+1} - S^t)/\Delta t$. Spatial derivatives are second-order accurate central differences:

$$\partial S/\partial x = (S^x + 1 - S^{x-1})/2\Delta x$$

and

$$\partial^2 S/\mathrm{x}^2 = (S^x + 1 - 2S^x + S^{x-1})/(\Delta x)^2.$$

The symmetry in time of these equations is required so that the influence of the observed data will propagate at the same (computational) speed both forwards and backwards in time. For the computed values to be continuous in space and time, we use the scheme which utilize data from past, present and future as well as northern, southern, eastern and western neighbours. This is an improvement over a widely used Crank-Nicholson scheme, in which only the present and past data are used.

For the surface water advective transport over the Atlantic, the annual mean field (4° × 5°) which has been computed by Inez Fung of the University of Victoria, Canada, on the basis of the monthly wind field of Esbensen & Kushnir (1981) has been used. The advective field is assumed to be unchanged with time. Lateral diffusivity of upper layers of the ocean has been estimated to be between 1000 and 3000 $\text{m}^2 \text{ s}^{-1}$ (Thiele *et al.* 1986; Jenkins 1991). Throughout this study, a constant eddy diffusivity of 1000 $\text{m}^2 \text{ s}^{-1}$ is used. About 1000 iterations were needed for satisfactory solution of the transport equation, equation (3).

(*b*) *Resampling of the observed data*

The observational data must be resampled onto the computational grid (4° latitude × 5° longitude × 365 days). A weighted average, inversely proportional in both space and time, is used to transform data onto this grid. A datum ± 4° latitude, ± 5° longitude, ± 1 day from the sampling point is utilized in the computations. This procedure requires each data point be resampled onto eight computational nodes (its two spatial neighbours in latitude and longitude and its temporal neighbours past and present days). This liberal resampling algorithm is useful to give sparse data more representation, and aids in convergence.

(*c*) *Comparison with the climatological* SST *data*

To evaluate the validity of the interpolation scheme described above, the SST values which have been computed using the measurements made with $p\text{CO}_2$ at about 2700 stations in the Atlantic north of 18° S (see figure 1) have been compared with the climatological field of SST compiled by Shea *et al.* (1992).

In figure 2, the difference between interpolated SST values for each month at 282 node points (a total of 3376) and the climatological mean values are shown. The 'o' symbols signify nodes where measurements were made and the '+' symbols indicate nodes where there are no available observations. The interpolated SST values are shown to be consistent with the climatological data with a mean difference of −0.24 °C with a standard deviation of ±2.55 °C or a standard deviation of mean of ±0.05 °C ($= 2.55/3376^{\frac{1}{2}}$). It also shows that '+' symbols located at temperatures between 25 and 29 ° C deviate up to −10 ° C. These node points are located in the Gulf of Mexico, Caribbean Sea and Gulf of Guinea, where no measurements were made. Our scheme, which has been designed for interpolation, is not suited for predicting the maximum temperature in this region. There are number of points which deviate upward by as much as +13 °C and −7 °C in a SST range between 4–20 °C. These points represent the areas of the Gulf Stream and its extension, where lateral temperature gradients are large. Therefore, pixels which straddle steep gradient areas yield erroneous values. If a finer grid is used, this problem may be eliminated.

4. DISTRIBUTION OF THE CO_2 SINK/SOURCE OVER THE NORTH ATLANTIC

The mean monthly distribution of sea–air $p\text{CO}_2$ difference over the North Atlantic representing the year 1990 is presented in figure 3. On the basis of the mean monthly wind speed data (Esbensen & Kushnir 1981), two sets of air–sea CO_2 gas transfer coefficient, k in equation (1), are computed using the formulations of Tans *et al.* (1990) and Liss & Merlivat (1986), each representing the upper and lower limits respectively. The former has been estimated on the basis of the distribution of ^{14}C in the atmosphere and oceans, and the latter on the basis of wind tunnel experiments and lakes using non-reactive gases such as SF_6. The monthly CO_2 flux across the sea surface has been computed for each pixel area using equation (1), in which mean monthly $\Delta p\text{CO}_2$ and mean monthly wind speed were used (see figure 4). Although high frequency measurements for both of these quantities are desirable for a more accurate assessment of the CO_2 flux, such measurements are not available over a desired range in space and time. The effect on $\Delta p\text{CO}_2$ of the difference between the bulk water and surface skin temperatures is not considered in this paper.

Table 1. *Mean monthly ΔpCO_2 values (μatm) for the year 1990 in 12°-wide zonal belts.*

((a) indicates that the $\Delta p\mathrm{CO}_2$ observations located north of 50° N have been corrected to 1990 assuming no interannual increase in surface water $p\mathrm{CO}_2$; (b) indicates that the $\Delta p\mathrm{CO}_2$ observations located north of 50° N have been corrected to 1990 assuming that surface water $p\mathrm{CO}_2$ increased at a rate of 50 % of the atmospheric increase.)

		recording period												
latitude		Jan	Feb	Mar	Apl	May	Jun	Jly	Aug	Sep	Oct	Nov	Dec	annual
66N–78N	(a)	−69.4	−59.4	−50.1	−48.8	−51.2	−61.9	−70.6	−76.8	−76.0	−67.2	−71.6	−70.8	−62.9
	(b)	−62.0	−52.7	−43.6	−42.5	−45.5	−55.9	−64.7	−70.0	−69.8	−60.9	−64.4	−63.6	−57.1
54N–66N	(a)	−37.9	−31.8	−30.9	−34.0	−43.6	−51.6	−56.0	−63.7	−60.5	−51.2	−45.4	−42.7	−45.7
	(b)	−33.3	−27.1	−25.8	−29.0	−39.1	−46.8	−50.0	−57.3	−54.4	−45.6	−40.6	−38.3	−40.5
42N–54N	(a)	−18.8	−17.5	−22.3	−29.3	−43.4	−40.5	−26.1	−10.6	−12.8	−21.4	−25.7	−25.5	−24.5
	(b)	−18.3	−16.6	−21.3	−28.5	−42.6	−39.4	−24.3	−9.7	−11.7	−20.5	−25.2	−25.2	−23.6
30N–42N	(a)	−23.1	−28.2	−33.2	−37.5	−28.4	−16.4	−9.9	−3.3	−5.2	−13.5	−17.8	−22.3	−19.9
	(b)	−23.0	−28.2	−33.2	−37.4	−28.4	−16.3	−9.8	−3.1	−5.1	−13.4	−17.7	−22.3	−19.8
18N–30N		−13.2	−14.0	−18.2	−19.7	−17.5	−14.2	−12.1	−8.7	−3.8	−4.4	−8.1	11.3	−12.1
6N–18N		−5,5	1.9	−0.3	−4.6	−4.5	−2.8	0.7	6.0	5.9	2.0	−5.6	−6.7	−1.1
6S–6N		23.1	36.1	31.0	23.0	18.5	17.8	18.3	21.0	18.0	18.6	19.8	19.8	22.1
6S–18S		27.8	33.1	29.7	28.8	27.4	27.2	27.4	26.5	20.8	12.3	15.0	25.0	25.1
average	(a)	−6.9	−3.0	−6.3	−10.3	−12.0	−10.2	−7.9	−5.0	−5.7	−8.3	−9.7	−9.1	−8.6
	(b)	−6.2	−2.3	−5.6	−9.6	−11.4	−9,5	−6.9	−4.0	−4.7	−7.4	−8.9	−8.4	−7.8

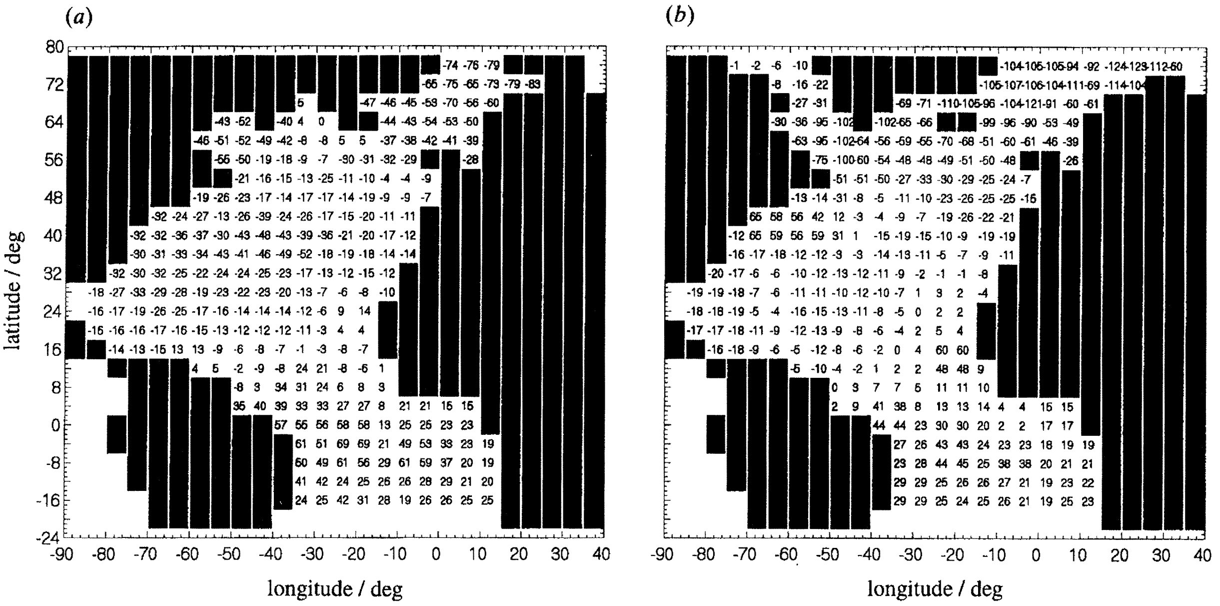


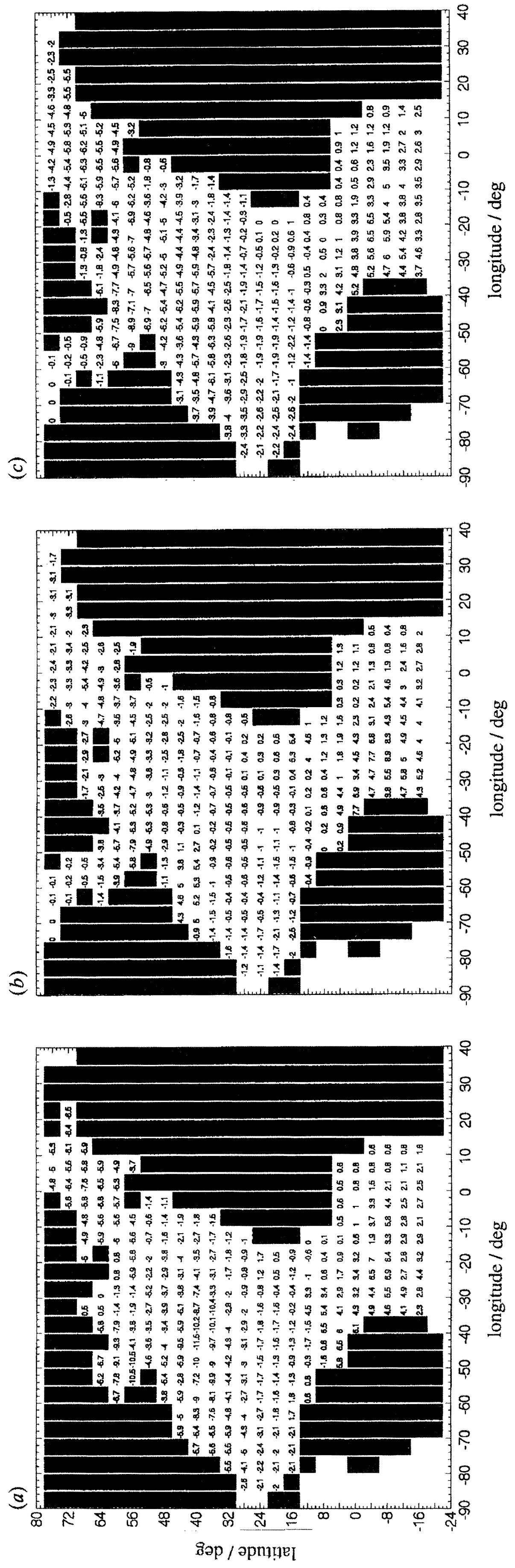
Figure 3. Distribution of sea–air $p\mathrm{CO}_2$ difference (μatm) during: (*a*) February 1990; and (*b*) August 1990.

(*a*) *Distribution of ΔpCO_2*

Table 1 shows the area-weighted mean $\Delta p\mathrm{CO}_2$ values in each 12° zonal band computed using the mean monthly pixel values. The positive $\Delta p\mathrm{CO}_2$ values indicate that the seawater is a source for atmospheric CO_2, whereas the negative values indicate that the seawater is a sink for atmospheric CO_2. The lines marked (a) and (b) represent the values computed on the basis of the two assumptions used for correcting $\Delta p\mathrm{CO}_2$ values as explained in §2*b*. The assumption (a) yields $\Delta p\mathrm{CO}_2$ values approximately 10 % more negative.

Figure 3*a*, *b* shows respectively the distribution of mean monthly $\Delta p\mathrm{CO}_2$ computed for February and August 1990, using the assumption (a). During the winter (figure 3*a*), the North Atlantic areas north of about 18° N are moderate to strong sinks having $\Delta p\mathrm{CO}_2$ values more positive than −60 μatm, whereas the tropical Atlantic areas between 18° N and 18° S are generally moderate sources having $\Delta p\mathrm{CO}_2$ values less than about 70 μatm. There is an unique area in the vicinity of Iceland (represented by seven pixel areas between 60° N and 68° N) where the sea is a CO_2 source during the winter. This is attributed to the effect of upwelling of deep water rich in CO_2 and represents ventilation of deep waters of the North Atlantic.

During August, the high latitude areas north of about 50 ° N turn to strong sinks with $\Delta p\mathrm{CO}_2$ values as low as −120 μatm. This is attributed to the photosynthetic utilization of CO_2 in the strongly stratified surface mixed layer (Takahashi *et al.* 1993). The mid-latitude areas between 50° N and 12° N become much weaker CO_2 sinks with $\Delta p\mathrm{CO}_2$ values generally more positive than −20 μatm due mainly to the effect of warming during the summer. The seasonal $\Delta p\mathrm{CO}_2$



changes in the temperate oceans are caused mainly by seasonal changes in temperature and hence are higher during the summer and lower during the winter; those in high latitude areas are approximately six months out of phase indicating a weaker sink during cold seasons and a stronger sink during warm seasons.

(*b*) *Net air–sea CO_2 flux*

Figure 4*a*, *b* shows the distribution of net monthly CO_2 flux (10^{11} gm C month^{-1}) for each pixel area during February 1990 and August 1990 respectively computed using the $\Delta p\mathrm{CO}_2$ values presented in figure 4*a*, *b*. The effect of wind speed on the CO_2 gas transfer coefficient by Tans *et al.* (1990) is used: k (moles $\mathrm{CO_2\ m^{-2}\ a^{-1}\ \mu atm^{-1}}$) $= 0.016[W\ (\mathrm{m\ s^{-1}}) - 3]$, where W is the wind speed and $k = 0$ if $W < 3\ \mathrm{m\ s^{-1}}$. Pixel areas which are covered with ice more than 50 % of their areas for each month are assumed to have no CO_2 transfer across the entire pixel area.

During February, the area between 34° N and 42 ° N in the central North Atlantic between 20° W and 40° W is the most intense CO_2 sink. This is a combined effect of high wind speed and low $\Delta p\mathrm{CO}_2$ values caused by cooling of the warm Gulf Stream water. The equatorial zone between 2° N and 10° S is the most intense CO_2 source. During August, the most intense CO_2 sink areas are located north of about 50° N where the photosynthetic utilization of CO_2 is fuelled by nutrients brought up to the photic zone during the previous winter. The mid-latitude areas are weak sources or sinks with the exceptions of the coastal upwelling areas. The equatorial areas remain unchanged. Zonal distribution of the monthly net CO_2 flux is summarized in table 2.

The annual net CO_2 flux values summarized in table 2 show that the high latitude areas north of 42° N are CO_2 sinks throughout a year with a net flux of about -0.45 Gt C a^{-1} if the gas transfer coefficients of Tans *et al.* (1990) are used (or about -0.2 Gt C a^{-1} if Liss & Merlivat (1986) coefficients are used). The sink flux of -0.2 Gt C a^{-1} over the temperate North Atlantic between 18° N and 42° N roughly cancels the source flux over the tropical Atlantic between 18° N and 18° S. Tans *et al.* (1990) reported an annual net sink flux of -0.23 Gt C a^{-1} for the Atlantic areas north of 50° N and -0.30 Gt a^{-1} for the temperate Atlantic areas between 15° N and 50° N. Although the estimate obtained by the present study for the temperate Atlantic is similar, the sink flux for the high latitude areas is about 1.5 times as large as that of Tans *et al.* (1990). This difference is primarily a result of the

Figure 4. Distribution of net sea–air CO_2 flux in 4° (latitude) × 5° (longitude) pixel areas during: (*a*) February 1990; (*b*) August 1990; and (*c*) the year 1990. The monthly mean fluxes in (*a*) and (*b*) are given in the unit of 10^{11} gm C month^{-1}, and the annual fluxes in (*c*) are given in 10^{12} gm C a^{-1}. The wind speed dependence on the CO_2 gas transfer coefficient based on carbon–14 distribution in the atmosphere and oceans (Tans *et al.* 1990) and mean monthly wind speed compiled by Esbensen & Kushnir (1981) have been used.

Table 2 *Mean monthly net CO_2 flux for* 12°-*wide zonal belts computed using the CO_2 gas transfer coefficients of Tans et al.* (*1990*) *and Liss & Merlivat* (*1986*).

((a) indicates that the $\Delta p\mathrm{CO}_2$ observations north of 50° N have been corrected to 1990 assuming no interannual increase in surface water $p\mathrm{CO}_2$ values; (b) indicates that the $\Delta p\mathrm{CO}_2$ observations located north of 50° N have been corrected to 1990 assuming that surface water $p\mathrm{CO}_2$ increased at a rate of 50 % of the atmospheric increase. The monthly CO_2 fluxes are given in 10^{12} gm C month^{-1} and the annual fluxes are given in 10^{13} gm C a^{-1}.)

		recording period												
latitude		Jan	Feb	Mar	Apl	May	Jun	Jly	Aug	Sep	Oct	Nov	Dec	annual
Tans *et al.* gas transfer coefficients														
66N–78N	(a)	−11.2	−9.1	−6.6	−5.4	−4.5	−6.0	−6.7	−7.9	−13.4	−16.2	−16.3	−11.7	−11.5
	(b)	−10.0	−8.1	−5.7	−4.7	−4.0	−5.4	−6.1	−7.2	−12.3	−14.7	−14.7	−10.5	−10.3
54N–66N	(a)	−21.8	−17.5	−14.6	−12.4	−13.0	−13.1	−11.9	−15.0	−20.8	−22.6	−22.9	−23.1	−20.9
	(b)	−19.1	−14.8	−12.1	−10.6	−11.6	−11.8	−10.5	−13.4	−18.6	−20.1	−20.4	−20.7	−18.4
42N–54N	(a)	−14.6	−12.8	−13.9	−15.1	−18.2	−14.1	−8.7	−4.5	−6.4	−12.8	−16.8	−18.5	−15.6
	(b)	−14.2	−12.1	−13.2	−14.6	−17.8	−13.7	−8.1	−4.2	−5.9	−12.2	−16.5	−18.2	−15.1
30N–42N	(a)	−19.0	−23.8	−25.7	−23.2	−13.3	−6.1	−2.7	−0.5	−1.7	−7.2	−12.1	−17.2	−15.3
	(b)	−19.0	−23.7	−25.7	−23.2	−13.3	−6.1	−2.7	−0.4	−1.6	−7.2	−12.0	−17.2	−15.2
18N–30N		−7.9	−7.9	−9.7	−9.3	−7.0	−5.3	−4.9	−3.1	−1.4	−2.0	−4.4	−6.4	−6.9
6N–18N		−2.3	0.9	−0.2	−2.1	−2.3	−2.1	−1.0	1.4	1.4	0.5	−2.0	−2.7	−1.0
6S–6N		6.5	8.9	7.2	5.3	5.1	5.9	6.6	8.5	7.0	6.7	7.1	6.9	8.2
6S–18S		8.3	9.4	8.9	9.7	10.1	11.0	11.6	11.8	8.3	4.5	5.0	7.3	10.6
net flux	(a)	−62.2	−51.9	−54.7	−52.5	−43.0	−29.9	−17.7	−9.2	−27.0	−49.1	−62.3	−65.4	−52.5
	(b)	−57.9	−47.5	−50.6	−49.5	−40.8	−27.6	−15.1	−6.5	−23.1	−44.5	−57.8	−61.6	−48.3
Liss & Merlivat gas transfer coefficients														
66N–78N	(a)	−5.5	−4.5	−3.2	−2.6	−2.1	−2.7	−3.0	−3.4	−6.3	−7.8	−8.0	−5.6	−5.5
	(b)	−4.9	−4.0	−2.8	−2.2	−1.8	−2.5	−2.7	−3.1	−5.7	−7.1	−7.2	−5.1	−4.9
54N–66N	(a)	−10.7	−8.6	−7.1	−6.0	−6.1	−6.0	−5.3	−6.8	−9.9	−11.0	−11.2	−11.4	−10.0
	(b)	−9.4	−7.3	−5.9	−5.1	−5.5	−5.5	−4.7	−6.1	−8.9	−9.8	−10.0	−10.2	−8.8
42N–54N	(a)	−7.2	−6.3	−6.8	−7.3	−8.6	−6.5	−4.0	−2.1	−3.1	−6.2	−8.3	−9.1	−7.6
	(b)	−7.0	−6.0	−6.5	−7.1	−8.4	−6.3	−3.7	−2.0	−2.8	−5.9	−8.1	−9.0	−7.3
30N–42N	(a)	−9.3	−11.6	−12.4	−11.0	−6.1	−2.7	−1.1	−0.2	−0.7	−3.4	−5.8	−8.3	−7.3
	(b)	−9.2	−11.6	−12.4	−11.0	−6.1	−2.7	−1.1	−0.1	−0.7	−3.4	−5.8	−8.3	−7.2
18N–30N		−3.7	−3.7	−4.5	−4.2	−3.1	−2.3	−2.2	−1.3	−0.6	−0.9	−2.0	−3.0	−3.1
6N–18N		−1.1	0.4	−0.1	−1.0	−1.1	−1.0	−0.5	0.6	0.6	0.2	−0.9	−1.2	−0.5
6S–6N		2.8	3.7	2.9	2.1	2.2	2.6	3.0	3.9	3.2	3.0	3.2	3.0	3.6
6S–18S		3.6	4.1	3.9	4.4	4.6	5.0	5.4	5.5	3.8	2.1	2.3	3.2	4.8
net flux	(a)	−31.1	−26.4	−27.3	−25.5	−20.3	−13.7	−7.8	−3.9	−13.0	−24.0	−30.6	−32.4	−25.6
	(b)	−29.0	−24.2	−25.3	−24.0	−19.2	−12.7	−6.7	−2.7	−11.2	−21.8	−28.4	−30.5	−23.6

corrections applied to the $\Delta p\mathrm{CO}_2$ values to normalize them to the year 1990. Because such corrections were not used by Tans *et al.* (1990), their $\Delta p\mathrm{CO}_2$ values in high latitude areas were smaller in absolute magnitude representing a time period of 1972–1982. Additionally, the present study reflects our improved understanding of the seasonal variability of $p\mathrm{CO}_2$ in the high latitude North Atlantic as summarized by Takahashi *et al.* (1993).

Because the high latitude CO_2 sink areas coincide with the water mass formation areas of the North Atlantic Deep Water complex, this annual flux, which corresponds to about 5 to 10 % of the annual industrial CO_2 emissions, may be directly transported into the deep interior of the Atlantic Ocean via thermohaline circulation.

(*c*) *Estimation of errors*

Although the errors associated with the estimation of $\Delta p\mathrm{CO}_2$ cannot be made directly, the differences between the climatological SST values and those estimated by means of the present computational schemes may be taken as a measure of errors in estimated $\Delta p\mathrm{CO}_2$ values. As presented in figure 2, the mean annual difference between the climatological and estimated SST values is -0.24 ± 0.05 °C. As the effect of temperature on $p\mathrm{CO}_2$ in seawater is about 15 µatm C^{-1} ($= 0.0423$ C$^{-1} \times 350$ µatm), the mean difference in the SST values corresponds to -3.6 ± 0.8 µatm for $p\mathrm{CO}_2$. If this is taken to indicate the magnitude of errors in the mean annual $\Delta p\mathrm{CO}_2$ for the entire study area, our estimate of -8.6 µatm (see table 1) should have a systematic error of about 40 %. However, the mean annual $\Delta p\mathrm{CO}_2$ value for high latitude areas north of 54° N is about -50 µatm and that between 30° N and 54° N is about -20 µatm, and hence the errors may be about 7 % and 18 % respectively. The net CO_2 fluxes computed should be subject to errors corresponding to these percentage errors.

5. CONCLUSIONS

Net CO_2 fluxes across the sea surface during the year 1990 over the North and tropical Atlantic have been estimated based upon the sea–air pCO_2 difference and gas transfer coefficients. The annual sink flux over the temperate North Atlantic between 42 ° N and 18° N is nearly balanced with the source flux over the tropical Atlantic between 18° N and 18° S. The high latitude North Atlantic areas north of 42° N are an important sink for atmospheric CO_2 having a net annual CO_2 flux of 0.2 to 0.5 Gt C a^{-1}. These CO_2 sink areas coincide with the water mass formation areas of the North Atlantic Deep Water complex, and hence this annual flux, corresponding to about 5 to 10% of the annual industrial CO_2 emissions, may be directly transported into the deep interior of the Atlantic Ocean via thermohaline circulation.

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Discussion

R. G. Williams (*Oceanography Laboratories, University of Liverpool, U.K.*). I support the attempt to seasonally sort the $p\mathrm{CO}_2$ data. However, I am concerned about the lack of source/sink term in the advection and diffusion equation. For example, a water parcel in the mixed layer circuiting the subtropical gyre changes its temperature and carbon content principally through the air–sea fluxes of heat and CO_2. If the advection–diffusion scheme is applied without these source terms, then properties either will be made unrealistically uniform along streamlines with low diffusion or, perhaps more plausibly, will be smeared out horizontally with high diffusion.

T. Takahashi. The computational scheme presented has been developed strictly for the purpose of interpolation observations in space and time, but not for modelling. An addition of sink and source terms for CO_2, as suggested by Dr Williams, would make the interpolated $p\mathrm{CO}_2$ values influenced by those additional terms, which are complicated and may only be parameterized using a number of poorly understood expressions.