

Simulation and assimilation of global ocean $p\text{CO}_2$ and air–sea CO_2 fluxes using ship observations of surface ocean $p\text{CO}_2$ in a simplified biogeochemical offline model

By VINU VALSALA* and SHAMIL MAKSYUTOV, *National Institute for Environmental Studies (NIES), 16-2 Onogawa, Tsukuba 305-8506, Ibaraki, Japan*

(Manuscript received 31 December 2009; in final form 6 July 2010)

ABSTRACT

We used an offline tracer transport model, driven by reanalysis ocean currents and coupled to a simple biogeochemical model, to synthesize the surface ocean $p\text{CO}_2$ and air–sea CO_2 flux of the global ocean from 1996 to 2004, using a variational assimilation method. This oceanic CO_2 flux analysis system was developed at the National Institute for Environmental Studies (NIES), Japan, as part of a project that provides prior fluxes for atmospheric inversions using CO_2 measurements made from an on-board instrument attached to the Greenhouse gas Observing SATellite (GOSAT). Nearly 250 000 $p\text{CO}_2$ observations from the database of Takahashi et al. (2007) have been assimilated into the model with a strong constraint provide by ship-track observations while maintaining a weak constraint of 20% on global averages of monthly mean $p\text{CO}_2$ in regions where observations are limited. The synthesized global air–sea CO_2 flux shows a net sink of 1.48 PgC yr^{-1} . The Southern Ocean air–sea CO_2 flux is a sink of 0.41 PgC yr^{-1} . The interannual variability of synthesized CO_2 flux from the El Niño region suggests a weaker source (by an amplitude of 0.4 PgC yr^{-1}) during the El Niño events in 1997/1998 and 2003/2004. The assimilated air–sea CO_2 flux shows remarkable correlations with the CO_2 fluxes obtained from atmospheric inversions on interannual time-scales.

1. Introduction

Ongoing anthropogenic emissions have caused an increase in the atmospheric concentrations of carbon dioxide (CO_2) from a pre-industrial value of 270 ppm to the present-day value of 380 ppm (Keeling et al., 1982). However, the present-day concentration of anthropogenic CO_2 in the atmosphere represents only half of the net emissions caused by human activities: the other half is absorbed by the terrestrial biosphere and the ocean, thereby moderating the net increase in atmospheric CO_2 concentrations. Uncertainties exist in any quantitative measure of the uptake of atmospheric CO_2 by the land biosphere and the ocean ecosystem, regardless of the calculation method. These uncertainties arise mainly because of the practical difficulties involved in direct estimates of fluxes between the atmosphere and the ocean or land surface. Moreover, there are few observational data with which to quantify the net exchange of CO_2 . Here, we present a strategy of estimating the exchange of CO_2 between the ocean and the atmosphere. We adapt the usual strategy of biogeochemical cycle modelling in an offline ocean circulation model, with

the important addition of constraining the $p\text{CO}_2$ of the modelled surface ocean using observations in a variational assimilation system.

Previous air–sea CO_2 flux estimates, as calculated from a large number of surface ocean $p\text{CO}_2$ observations, suggest that the ocean absorbs nearly 1.5 petagrams of carbon (PgC) each year (Takahashi et al., 2009). This annual mean uptake varies interannually depending on climate mode anomalies such as El Niño (during El Niño years, the uptake is generally 0.5 PgC more than the annual mean), the Pacific Decadal Oscillation, and the North Atlantic Oscillation (Feely et al., 1999; Le Quere et al., 2000; Obata and Kitamura, 2003; Keeling et al., 2004; McKinley et al., 2004; Patra et al., 2005; Wetzel et al., 2005; Lenton and Matear, 2007; Le Quere et al., 2007; Thomas et al., 2008).

The most common strategies employed in estimating air–sea CO_2 fluxes are as follows: (a) In the ‘top-down’ approach, sources and sinks are estimated from observations of atmospheric CO_2 concentrations and Greens functions (i.e. the transport of CO_2 concentrations) derived from transport models. This method is known as inverse modelling of sources and sinks (Gurney et al., 2004; Patra et al., 2005; Jacobson et al., 2007; Gruber et al., 2009). However, the efficiency of this method relies on the total number of observations from land-based tower

*Corresponding author.

e-mail: vinu.valsala@nies.go.jp

DOI: 10.1111/j.1600-0889.2010.00495.x

stations and the regional distribution of the stations (Patra and Maksyutov, 2002). Because of the limited number of observation data, as well as the biases inherent in transport models, the inverse method involves large uncertainties. The direct assimilation of CO_2 observations into transport models also offers optimized estimates of CO_2 fluxes (Baker et al., 2006; Yumimoto and Uno, 2006). (b) The second method is a 'bottom-up' approach, in which process models of oceanic biogeochemical cycles are incorporated into a general circulation model, yielding estimates of CO_2 gas exchange. This approach involves uncertainty related to our limited knowledge of the key processes that govern the carbon cycle of the terrestrial biosphere and the ocean ecosystem. Previous studies have examined the assimilation of various types of data into biogeochemical models (Tjiputra et al., 2007). (c) It is also possible to measure the isotopic ratios of CO_2 in the atmosphere and thereby perform indirect measurements of CO_2 exchange among the terrestrial biosphere, ocean, and atmosphere (Keeling et al., 2001). (d) Mean global air-sea CO_2 fluxes can also be estimated from direct observations of oceanic partial pressure of CO_2 ($p\text{CO}_2$) and gas exchange coefficients (Takahashi et al., 2009). However, the sparse coverage of $p\text{CO}_2$ data requires large-scale interpolation to perform analyses in the global domain. Gruber et al. (2009) compared global estimates of air-sea CO_2 fluxes obtained using these various methods, revealing a general consistency, although with regional-scale inconsistencies.

Here, we present a bottom-up approach to estimating air-sea CO_2 fluxes, with an important modification compared with existing approaches: that is simulated surface ocean $p\text{CO}_2$ is constrained with available ship observations between 1996 and 2004. This approach provides model-predicted, interannually varying, error-minimized estimates of global air-sea CO_2 flux. This oceanic CO_2 flux analysis system was developed at the National Institute for Environmental Studies (NIES), Japan, as part of a project designed to provide prior fluxes for atmospheric inversions using CO_2 observations performed by an on-board instrument attached to the Greenhouse gas Observing SATellite (GOSAT).

Global oceanic $p\text{CO}_2$ observations have been carried out by several research programs. The World Ocean Circulation Experiments (WOCE), Joint Global Ocean Flux Study (JGOFS) and Voluntary Ship of Opportunity (VOP) observation programs, among others, are some of the major programs to collect surface ocean carbon data. Takahashi et al. (2009) combined all these $p\text{CO}_2$ observations into monthly climatological maps on a global scale. However, the aggregation of multi-year $p\text{CO}_2$ observations into monthly climatology results in the loss of valuable information regarding the interannual variability of $p\text{CO}_2$ and air-sea CO_2 fluxes. In contrast, by using multi-year individual observations of $p\text{CO}_2$ as a constraint on a process model, it would be possible to resolve valuable information regarding the interannual variability of air-sea CO_2 fluxes, at least on the regional scale, for which frequent observations of $p\text{CO}_2$ are available.

In this study, we used multi-year $p\text{CO}_2$ data to constrain the model $p\text{CO}_2$. The results demonstrate the potential benefits of using $p\text{CO}_2$ observations as constraints when minimizing model biases and errors.

To minimize the model error via the assimilation of observed $p\text{CO}_2$, we derived a simple biogeochemical model from McKinley et al. (2004). In this model, the net export production in the euphotic zone is estimated from the relation between phosphorus, light availability and primary production, and is then scaled by a regional mask to represent other processes that limit the primary production, such as Fe availability and grazing efficiency. The net export production is then converted to equivalent dissolved inorganic carbon (DIC) consumption within the euphotic zone, using a Redfield ratio. Below the euphotic zone, re-mineralization returns DIC that is exported to the deep ocean. We adopt this simplified ecosystem model for two reasons: (1) our aim is to utilize observations of surface ocean $p\text{CO}_2$ as a main constraint on modelled $p\text{CO}_2$ values, and (2) the construction of an adjoint is simpler if the ecosystem is limited to one component and the adjoint can be limited to one control variable (i.e. DIC).

The remainder of the paper is organized as follows. Section 2 describes the physical and biogeochemical models, and the method of constraining modelled $p\text{CO}_2$. The results are presented in Section 3 and a comparison of our assimilated model output with data from other sources are given in Section 4. The results are discussed in Section 5.

2. Physical, biogeochemical models and experiments

2.1. Physical model

We used a simple biogeochemical model coupled to an ocean tracer transport model driven by reanalysis ocean data. The transport model is the Offline ocean Tracer Transport Model (OTTM), as documented in Valsala et al. (2008). In this model, two-dimensional circulation, temperature, salinity and other diagnostic physical parameters are taken from ocean reanalysis data and used to evolve a tracer. The tracer of interest is DIC, which is a collection of carbonate (CO_3^{2-}), bicarbonate (HCO_3^{-}) and dissolved CO_2 gas. We used the reanalysis products provided by the Geophysical Fluid Dynamics Laboratory (GFDL), NOAA, Princeton, USA, as offline physical circulation. This study uses the data available between 1975 and 2004.

OTTM has self-operating diagnostic mode vertical mixing and subgrid-scale process parameterization schemes, because the coefficients for these processes are not readily available from the reanalysis data; instead, they were estimated within the offline model. Vertical mixing is represented as a combination of the K-Profile Parameterization (KPP; Large et al., 1994) and a background vertical diffusion suggested by Bryan and Lewis (1979). Horizontal diffusion is a combination of

flow-dependent diffusion, in which the coefficients are proportional to the stress and strain experienced in the local fluid volume (Griffies and Hallberg, 2000), and advection fluxes due to eddy-induced transport (Gent and McWilliams, 1990).

The data for horizontal advection were taken from the offline data archive, whereas vertical velocities were calculated based on the principles of mass conservation. The free-surface kinematic boundary condition was determined from the sea surface height. OTTM transport was extensively tested in a previous study by simulating chlorofluorocarbon (see Valsala et al., 2008); this earlier study also contains details of the model design and validation. The exaggerated vertical mixing found in OTTM within the tropics, as reported by Valsala et al. (2008), was resolved by further tuning of the model in this study.

2.2. Chemical model

The chemical compartment of the biogeochemical model consists of a single tracer (DIC). $p\text{CO}_2$ was treated as being in near-equilibrium with the atmosphere at the sea surface via air–sea gas exchange. Within the ocean, DIC consists of CO_3^{2-} , HCO_3^- , and dissolved gaseous CO_2 , although they are transported as a single tracer (DIC). This approach corresponds to the solubility pump model described in Ocean Carbon Cycle Inter Comparison Project-II (OCMIP-II; Orr et al., 1999, available at <http://www.ipsl.jussieu.fr/OCMIP/>).

Air–sea gas exchange depends on piston velocity K_w and the difference in $p\text{CO}_2$ between the surface ocean and the ambient atmosphere above. Because the focus of this study is the contemporary air–sea CO_2 flux, a constant atmospheric $p\text{CO}_2$ of 368 μatm (as observed in 2000) was used. Alkalinity [taken as total alkalinity (TA)] was not transported but was estimated from the reanalysis salinity inputs with a constant conversion factor of 2310 $\mu\text{eq kg}^{-1}$ as, $TA = 2310S/S_g$, in which S is the local surface salinity and S_g is the annual mean surface salinity, integrated globally. This relation is taken from the OCMIP-II protocol (Orr et al., 1999).

The air–sea flux in the chemical model was formulated as $\phi_{\text{GASEX}} = K_w(p\text{CO}_{2\text{OCEAN}} - p\text{CO}_{2\text{AIR}})$. Model $p\text{CO}_2$ was calculated using the routines provided in OCMIP-II (Orr et al., 1999). The term K_w is the piston velocity at which gaseous CO_2 enters or leaves the surface according to wind speed and CO_2 solubility, as formulated in Wanninkhof (1992). We used a gas exchange proportionality constant of $a = 0.337$ taken from OCMIP-II (Orr et al., 1999) when calculating K_w . Surface wind speed for the calculation of K_w was the sum of 10-day averages of 6-hourly squared wind speed (u_{10}^2) and 6-hourly wind speed variance ($\sigma_{u_{10}}^2$). Here, u_{10} indicates wind speed at 10 m height. An index for polar ice-caps was used to partially mask the air–sea gas exchange during sea-ice conditions. This was achieved by compiling a spatial map of the ice index, with values vary between 0.2 and 1, as taken from the OCMIP-II data set (Orr et al., 1999).

2.3. Biological model

The ecosystem model employed here is derived from McKinley et al. (2004). In the present model, net export production in the euphotic zone was calculated from climatological maps of phosphate (P) and light availability (I). An important difference between the present biological model and that of McKinley et al. (2004) is that we used monthly climatological phosphate to determine export production, whereas McKinley et al. transported phosphate and oxygen as two additional prognostic tracers. Given that McKinley et al. (2004) restored the biogeochemical tracers below 1200 m depth to observed climatology, our use of climatological phosphate means that the two studies show similar features below this depth.

A scaling factor was given as a regional mask (α), which implicitly represented the Fe limitation, grazing efficiency and recycling. Export production in the euphotic zone (0–140 m) is formulated as

$$B(z) = -\alpha(x, y) \left(\frac{I_{(x,y,z,12\text{-month})}}{I_{(x,y,z,12\text{-month})} + I_0} \right) \times \left(\frac{P_{(x,y,z,12\text{-month})}}{P_{(x,y,z,12\text{-month})} + P_0} \right), \quad (1)$$

where α represents other controlling factors of export rates. The value of the maximum export rate, α , encapsulates all the processes leading to the export of DIC which were not explicitly represented by phosphate and light limitation (McKinley et al., 2004). In this case, the value of α should be consistent with the models circulation and climatological nutrient fields. The global ocean was divided into 14 regions and the value of α in each region was defined based on the assumption that a given model flow field will produce an annual mean phosphorus distribution consistent with climatological observations. The oceanic regions for the α mask were taken from McKinley et al. (2004), whereas the individual regional values of α were tuned for the circulation used in this study. Because we do not transport phosphate in the present model, we achieved an optimal α mask by tuning the individual regional values of α to obtain model $p\text{CO}_2$ values similar to the climatology of Takahashi et al. (2009).

The half-saturation values for phosphate (P_0) and light (I_0) were set to 0.01 $\mu\text{mol kg}^{-1}$ and 30 W m^{-2} , respectively, which are the same values as those used in McKinley et al. (2004). Fluxes of sinking particles [$F(z)$] were parameterized as in Dutkiewicz et al. (2001). The net export production was converted to an equivalent DIC consumption in the euphotic zone, based on the Redfield ratio, $R_{\text{C:P}} = 117:1$ (McKinley et al., 2004). Within the euphotic zone, $Sb(z) = B(z) + F(z)$; below the euphotic zone, $Sb = F(z)$.

The use of climatological $P_{(x,y,z,12\text{-month})}$ may have a detrimental effect on the predicted $p\text{CO}_2$ because of the absence of interannual variability of export production in our model. One may interpret that those areas in the ocean where our simplified parameterization would influence the predicted $p\text{CO}_2$ are those

areas with P values similar to or less than the half-saturation coefficient P_0 . In these regions, export production depends on P/P_0 . Climatological phosphate is already in balance between the supply of P from the bottom layer and deposition of P by removal from organic matter. We compared the air–sea CO_2 fluxes from our model (non-assimilated run) with those reported by McKinley et al. (2004), revealing that they are well correlated over a seasonal cycle as well as regarding interannual variability [see also Fig. 3.12 in Maksyutov et al. (2010) for a comparison of the seasonal cycle]. Except for equatorial regions and to some extent the North Atlantic, we obtained a statistically significant correlation between the air–sea CO_2 fluxes of the present study and of McKinley et al. (2004).

In addition to air–sea gas exchange, the surface dilution of DIC by rainfall (P) and its concentration by evaporation (E) were included in the model (Φ_{FWEX}). Although the models surface boundary condition is kinematic, we have given an additional estimate of surface dilution to compensate for salinity errors (and corresponding alkalinity errors in the model) in the reanalysis ocean data (Whit Anderson, 2008, personal communication). In this case, to eliminate the net addition of DIC due to virtual fluxes, we subtracted the global mean of $P-E$ before computing the virtual fluxes. This protocol is taken from OCMIP-II. The data for virtual dilution were derived from the same reanalysis data. Therefore, the total (i.e. physical, chemical and biological) components of the model have the following form:

$$\frac{D[\text{DIC}]}{Dt} = R_{\text{C:P}} \times Sb(z) + \phi_{\text{GASEX}} + \phi_{\text{FWEX}}, \quad (2)$$

where $\frac{D[\text{DIC}]}{Dt}$ represents the total changes due to advection, mixing, diffusion and eddy-induced transport.

2.4. Data and model setup

We used the ocean reanalysis data prepared by GFDL as the physical driver of OTTM. These data were produced by Modular Ocean Model-4 (MOM4) forced with Coordinated Ocean Research Experiments (CORE) data sets (Large and Yeager, 2008) and with an assimilation of *in situ* temperature profiles from the National Oceanographic Data Center (NODC) archives using a 3D variational scheme (see <http://www.gfdl.noaa.gov/ocean-data-assimilation>). Details of the physical part of the ocean model can be found in Delworth et al. (2006) and Gnanadesikan et al. (2006).

The physical parameters borrowed from the reanalysis data are velocity (u, v), temperature, salinity, mixed layer depth, evaporation and precipitation rates, surface heat flux, surface wind stress (to derive vertical mixing) and sea surface height. The reanalysis data have a zonal resolution of 1° with a total of 360 grid points. Meridional resolution is 1° at higher latitudes and 0.8° in the tropics, with a total of 200 grid points. The data have 50 vertical levels with a 10 m increment in the upper 225 m and stretched vertical intervals below this layer. The study domain

extends from 81.5°S to 85.5°N . The reanalysis data are available for each month between 1960 and 2004. Physical variables at monthly time scales were interpolated into the models time-step of 2 h.

The physical and biogeochemical model was run for the mean state of DIC (pre-run) using a 12-month mean circulation and other physical parameters derived for the 10 yr between 1975 and 1984. We chose this period for deriving the mean circulation and other physical parameters for the pre-run because our interannual DIC simulation started from 1980. Thus, 12-month mean circulations derived between 1975 and 1984 are expected to yield a reasonable mean state of DIC.

The pre-run of the model was initialized with annual mean values of DIC derived from the GLODAP data set (Key et al., 2004). For the pre-run, surface wind speed and its variance for the air–sea gas exchange calculation were kept as climatological values. We took these climatological data from the OCMIP-II forcing database (available at <http://www.ipsl.jussieu.fr/OCMIP/>). The data were derived from a 5-year average of SSMI monthly squared wind speed and 30-day variance of the instantaneous wind speed from the corresponding monthly period (see also README.satdat provided by OCMIP-II, available at <http://www.ipsl.jussieu.fr/OCMIP/>).

Surface wind-speed data for the air–sea gas exchange calculations in the interannual run (Section 2.2) were taken from ERA-40 (Uppala et al., 2004). We note that the physical circulation used in this study is produced by forcing with optimal surface fluxes derived from CORE, which takes most of its atmospheric data from NCEP/NCAR. However, in CORE data, the NCEP winds are modified with QSCAT satellite winds to eliminate biases (Large and Yeager, 2008). Similarly, other components of the surface fluxes in the CORE data are corrected with available satellite measurements. Therefore, CORE is an optimal surface flux combined from various sources, and the offline circulation used in our study does not restrict us from using ERA-40 winds for the air–sea CO_2 flux calculation.

The pre-run was carried out for 20 years using monthly mean circulation. The monthly mean wind for the surface gas exchange calculations was repeated at each year of the pre-run. The surface DIC concentration and air–sea CO_2 flux of the model reached a quasi-equilibrium state at the end of the first 10 years of the pre-run and thereafter showed a minimum departure from the mean state. The global integral of CO_2 flux from the model was 1.3 PgC yr^{-1} in the first year of the spin-up and 2.25 PgC yr^{-1} at the end of 9 year. Therefore, the estimated drift in the first 9 years of spin-up is approximately 0.1 PgC yr^{-1} . After 16 years, the model CO_2 flux reached 2.35 PgC yr^{-1} . Therefore, the drift between 10 and 16 years is $0.014 \text{ PgC yr}^{-1}$, which represents 14% of the drift during the first 10 years of spin-up. From 16 years onward, the model CO_2 flux remained close to 2.35 PgC yr^{-1} . Although the surface CO_2 flux reached a quasi-equilibrium state after a 20-year spin-up, deeper adjustments (below 2000 m) continued because of the slow communication between surface and

subsurface ocean via deep convection. Moreover, re-mineralized DIC below 2000 m depth communicated with the surface on a time-scale longer than 20 years. However, such deep and slow adjustments are unlikely to influence the present results because of the short period of our analysis and the component of assimilation involved.

The average of the last 5 years of the pre-run was taken as the restart condition for the interannual simulation. The simulation was started with the restart condition and continued through the monthly fields for the year 1980. The simulation was then continued from 1980 to 1995 using real-time data. The model results deviated from the restart condition as soon as the data for 1980 were introduced. A near-steady-state was reached by the time the model had passed through the fields for the year 1980. The restart condition for January 1996 was used for the experimental runs.

We analysed the pre-run and interannual-run $p\text{CO}_2$ and air-sea CO_2 fluxes, and calculated the correlations with the observations reported by Takahashi et al. (2007) and Takahashi et al. (2009). Model performance was reasonable on seasonal to interannual time scales. The pre-run and interannual-run results are basically similar to those of McKinley et al. (2004).

2.5. Constraining the model $p\text{CO}_2$ using observations

The use of a simple biogeochemical model, in which biological and chemical cycles of CO_2 are related to a single tracer (DIC), simplified the construction of adjoint equations for the assimilation. These adjoints are used to constrain model $p\text{CO}_2$ to the corresponding observations. A variational assimilation method was used to minimize the cost function. We employed a descendant algorithm technique similar to that described by Ikeda and Sasai (2000). The assimilation involves the following steps.

(a) The forward model is run through an assimilation window of 2 months, starting from the initial condition. A 2-month window was chosen because the surface $p\text{CO}_2$ shows rapid equilibration with the atmosphere. Therefore, the adjoint has relatively negligible magnitude to correct the initial condition in a longer backward integration. The surface ocean $p\text{CO}_2$, DIC and other variables are saved along the ship observation tracks. The corresponding global-scale variables are saved at monthly intervals.

(b) The observation-model misfit values are run backward in a time-space domain in the adjoint simulation, which starts from the end point of day-60 and ends at day-1. We used a simple adjoint of physical circulation by reversing the direction of the offline currents and integrated the model backward in time while the direction of mixing and diffusion remained unchanged. This adjoint method is similar to that discussed previously by Fukumori et al. (2004), Hourdin and Talagrand (2002) and Hourdin et al. (2002). The adjoint model contains only one control variable: DIC. The surface ocean $p\text{CO}_2$ observations are con-

verted into equivalent CO_2 concentrations using model sea surface temperature (SST), sea surface salinity (SSS) and alkalinity, all of which are reanalysis offline input data. The cost function was chosen to be the squared difference between the data and modelled $p\text{CO}_2$, as follows:

$$J = w_1 \Sigma (p\text{CO}_2 - p\text{CO}_{2\text{ST}})^2 + w_2 \Sigma (p\text{CO}_2 - p\text{CO}_{2\text{CL}})^2, \quad (3)$$

where $p\text{CO}_{2\text{ST}}$ denotes the ship-track $p\text{CO}_2$ data and $p\text{CO}_{2\text{CL}}$ denotes the climatology of Takahashi et al. (2009). The weighting w_1 was kept equal to 1.

In addition to the along-track $p\text{CO}_2$ data, we used the monthly mean surface ocean $p\text{CO}_2$ derived from Takahashi et al. (2009) as an additional constraint. However, the weighting for this mean constraint (w_2) was retained only up to 20% of the weighting of the along-ship-track data assimilation. Moreover, the weighting for this mean constraint was spatially varied according to the inverse variance (σ^2) of the model interannual CO_2 flux variability, which we inferred from our forward model simulation for 1980–1995. This spatially varying weighting helped to constrain the model to the mean $p\text{CO}_2$ only in regions where the interannual variance was minimized. Thus, regions with higher interannual variability in $p\text{CO}_2$ (e.g., the eastern equatorial Pacific) were constrained only when ship-track data were available. Therefore, w_2 in the above equation is assumed the form $\min(0.2, \sigma_{\min}/\sigma)$ in which $\sigma_{\min} = 0.02$ ($\text{mole m}^{-2} \text{yr}^{-1}$)². A map of w_2 is shown in Fig. 5 (bottom panel).

The adjoint equations are deduced from

$$-\frac{\lambda_n - \lambda_{n-1}}{\Delta t} + w_1 \frac{\delta_{nn}}{\Delta t} f_g(p\text{CO}_2 - p\text{CO}_{2\text{ST}}) + w_2 \frac{\delta_{nn}}{\Delta t} f_g(p\text{CO}_2 - p\text{CO}_{2\text{CL}}) + \frac{\partial F_n}{\partial C} = 0, \quad (4)$$

where the adjoint operator λ at n th time-step is given as

$$\lambda_n = W_f \left(\frac{C_{n+1} - C_n}{\Delta t} - F_n \right), \quad (5)$$

where C_n is the control variable at the n th time step, W_f is a weighting coefficient, and F_n is forcing, including CO_2 gas exchange, virtual fluxes and biological export of DIC. f_g is a conversion factor for converting $p\text{CO}_2$ to DIC using SST, SSS and alkalinity (Weiss, 1974). The last term in eq. (4) represents the derivative of forcing with respect to the control variable, as follows:

$$\frac{\partial F_n}{\partial C} = \lambda_n K_w K \left[\frac{(2C - A)(3A - 2C)}{(A - C)^2} \right] + \lambda_n B(z) + \lambda_n (E - P), \quad (6)$$

where A represents alkalinity, K is a CO_2 solubility coefficient (Weiss, 1974) and $B(z)$ is derived from eq. (1). The first term on the right side of eq. (6) represents the derivative of the $p\text{CO}_2$ formulation with respect to DIC.

(c) The cost function is minimized using a descendant algorithm in which at each iteration, the initial condition is

corrected by the gradient of the cost function (Bennett, 2002). The weighted correction to the forcing in the assimilation run incorporates corrections to DIC induced as a combination of corrections to the biology and other surface fluxes such as fresh water and salt errors. However, these corrections are not separable because we use only one control variable (eqs 3–6). The iteration is repeated until the along-track model-to-observation misfit of $p\text{CO}_2$ is minimized below a threshold, defined as the iteration at which the cost function falls below 10% of the initial value.

The model was restarted from January 1996 and $p\text{CO}_2$ was synthesised for a monthly period with an iteration window of 2 months. The whole procedure was repeated every 2 months within the assimilation period.

3. Simulation and synthesis of $p\text{CO}_2$

3.1. $p\text{CO}_2$ data

The number of surface $p\text{CO}_2$ ship observations has expanded considerably since 1995. In Takahashi et al. (2007), compilation of global ocean $p\text{CO}_2$ database, approximately 3×10^5 quality-controlled observations were used. This study made use of Takahashi et al. (2007) data (Version 1.0) as ship-track $p\text{CO}_2$ data (i.e. $p\text{CO}_{2ST}$ in eq. 3). In addition to the research vessels participating in $p\text{CO}_2$ measurements, Ship-Of-Opportunity measurements made by cargo liners have provided a wealth of information about the surface ocean $p\text{CO}_2$, especially in regions such as the North Pacific (Zeng et al., 2002) and North Atlantic. Sampling density in the Southern Ocean has been improved in Version 1.0 $p\text{CO}_2$ data (Takahashi et al., 2007) compared to the older versions. Figure 1 summarizes the $p\text{CO}_2$ observation points used in this study, comprising 236 777 observations between

1996 and 2004. Although not all data in Takahashi et al. (2007) database are sampled along ship tracks (some are from station-data sampled on research cruises), we use the term ‘ship-track’ to represent individual $p\text{CO}_2$ observations taken from Takahashi et al. (2007) database.

The $p\text{CO}_{2CL}$ in eq. (3) is taken from Takahashi et al. (2009). Here we note that, there are following inconsistencies exist (inherited from the Takahashi et al., 2007) in the $p\text{CO}_2$ data that used in this study. (1) Different corrections were applied to 13 981 observations from the southern Indian Ocean lines collected between 1998 and 2000 (file name “OISO” in the LDEO database), which were subjected to a repeated (twice) SST correction in the Version 1.0 of Takahashi et al. (2007). The average error caused by this discrepancy is $-9.27 \pm 3.43 \mu\text{atm}$. (2) In all OISO data files, only CO_2 fugacity ($f\text{CO}_2$) values are reported without $p\text{CO}_2$ values or CO_2 concentrations in dry equilibrated gas. The approximate formula for the conversion of $f\text{CO}_2$ to $p\text{CO}_2$ yields $p\text{CO}_2$ values greater than $f\text{CO}_2$ by about $1.3 \mu\text{atm}$. These two uncertainties were common for both ship-track data and the climatology mean used here. (3) There is a slight change in $p\text{CO}_2$ interpolation near the meridian, which generates an error of less than $10 \mu\text{atm}$ in several grid points around 0°E in Takahashi et al. (2009). For this study, we derived the climatological mean constraint from Takahashi et al. (2009) and obtained ship-track data from Takahashi et al. (2007). The magnitude of data errors is minor compared with the residual error in the assimilation.

To enable post-assimilation comparisons, we combined Takahashi et al. (2007) $p\text{CO}_2$ data into $1^\circ \times 1^\circ$ bins. This approach leaves gaps in regions for which no observations are available, especially in the southern sub-tropical regions of all the major oceans and in the Southern Ocean. The North Atlantic and North Pacific have the largest number of observations. We

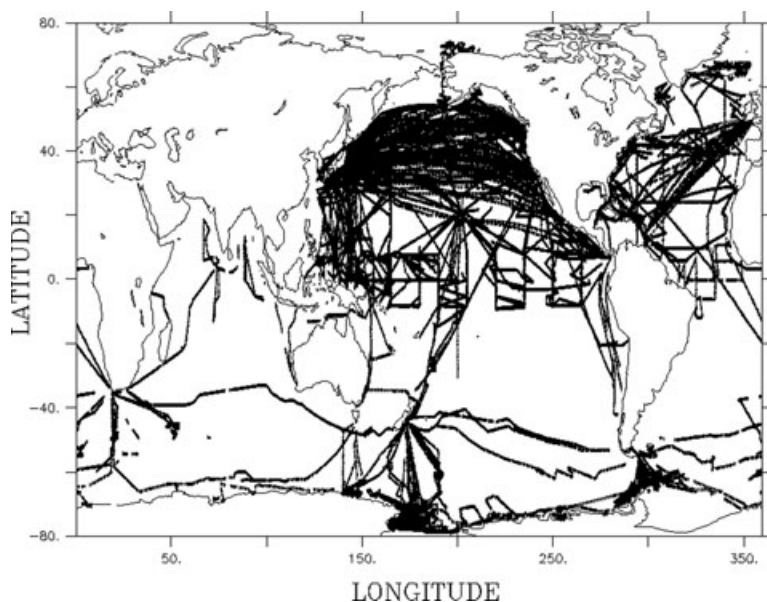


Fig. 1. Total number of ship observations and tracks used in the assimilation of surface ocean $p\text{CO}_2$ in the model. A total of 236 777 observations are used from Takahashi et al. (2007) database.

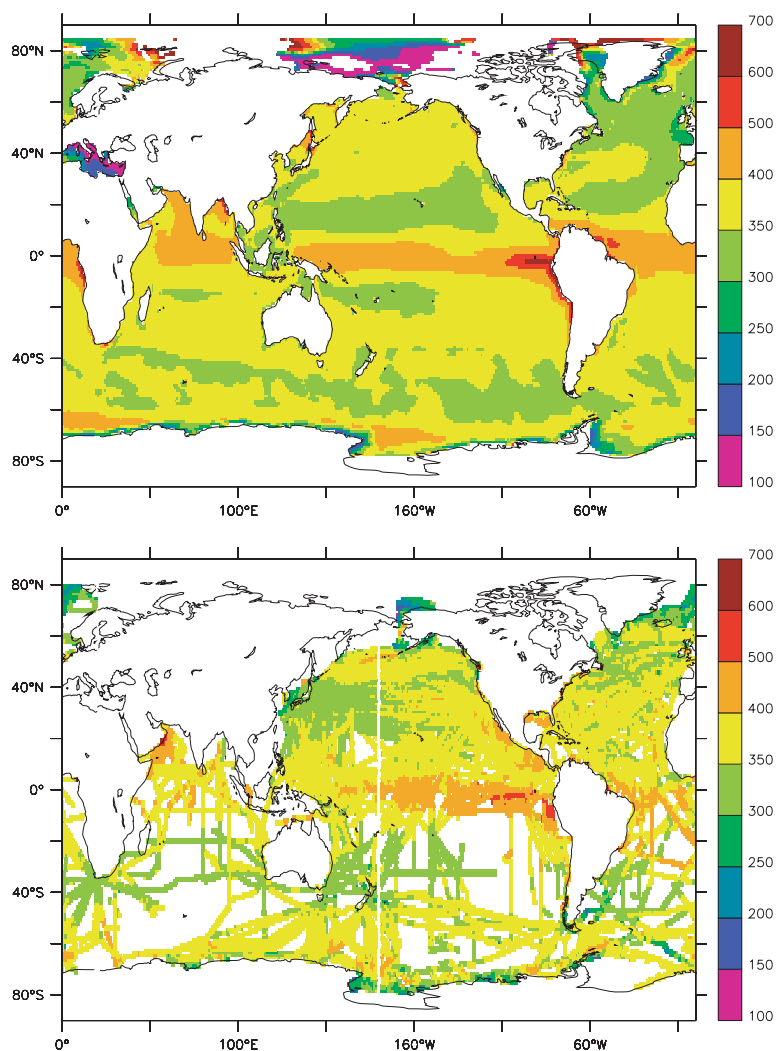


Fig. 2. (Top panel) 1980–1995 mean model simulated surface ocean $p\text{CO}_2$ in the free run and (bottom) observations from Takahashi et al. (2007) regridded into $1^\circ \times 1^\circ$ bins. The free run refers to the non-assimilated interannual simulation from 1980 to 1995. Units are in μatm .

compared this $1^\circ \times 1^\circ$ $p\text{CO}_2$ data set with our model and synthesized $p\text{CO}_2$ results to assess errors and biases.

3.2. Model $p\text{CO}_2$ error

The free-run of the model produced patterns of global mean surface ocean $p\text{CO}_2$ that are consistent with observations (Fig. 2). Here, the free-run refers to interannual model simulations without any constraint on surface ocean $p\text{CO}_2$. Figure 2 shows the annual mean $p\text{CO}_2$ of the model for the period 1980–1995. The free-run performs reasonably well in representing the elevated $p\text{CO}_2$ in the tropical Pacific and depressed $p\text{CO}_2$ in the Southern Ocean and North Atlantic. However, the modelled $p\text{CO}_2$ contains regional-scale errors in magnitude and in phase. For example, $p\text{CO}_2$ in the tropical Atlantic is overestimated in the free-run compared with the observations of Takahashi et al. (2007) (Fig. 2, bottom panel). The North Pacific has a limited springtime bloom related reduced $p\text{CO}_2$ in the free-

run compared with observations. In the Indian Ocean, the Arabian Sea is poorly represented and biased toward a reduced $p\text{CO}_2$.

Figure 3 shows the misfit between the annual mean $p\text{CO}_2$ of the free-run and the observations of Takahashi et al. (2007). Here, errors are calculated as the difference between the annual mean $p\text{CO}_2$ of the free-run and the corresponding mean from the observations of Takahashi et al. (2007). Large errors are found in the tropical Atlantic, North Atlantic, Northwest Pacific and Indian Ocean (Fig. 3, top panel). In the southern subtropics, however, model bias cannot be assessed because of a lack of observed data.

The annual mean error does not capture the seasonal drift in model $p\text{CO}_2$ from observations. To show the total error arising from seasonal biases in the model, we calculated the sum of the absolute misfit of free-run $p\text{CO}_2$ during each month from January to December (Fig. 3, bottom panel), compared with the observations of Takahashi et al. (2007).

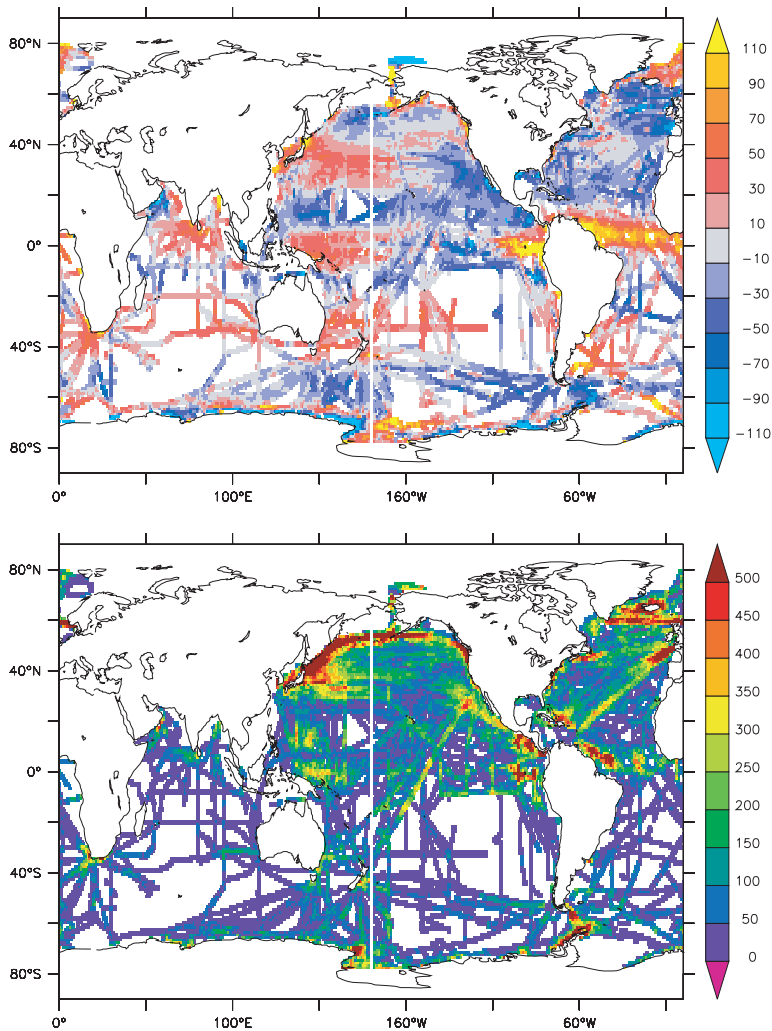


Fig. 3. (Top panel) Annual mean error of model $p\text{CO}_2$ compared to the observations of Takahashi et al. (2007). (Bottom panel) The sum of monthly error calculated as $(\sum_{l=1}^{12} [p\text{CO}_{2\text{mod}} - p\text{CO}_{2\text{obs}}])$. Units are in μatm .

The cumulated seasonal error in the model $p\text{CO}_2$ is large in the subarctic North Pacific and North Atlantic, which is not apparent in the annual mean error. The seasonal bias is also strong in the tropical Atlantic and eastern tropical Pacific, contributing to the annual mean error. The largest error in the seasonal cycle is observed in the North Pacific.

The lack of a one-to-one correspondence between regional-scale observed and simulated surface ocean $p\text{CO}_2$ may reflect the limited and simplified representation of the ecosystem model, which is inefficient in resolving processes such as spring bloom and variability in the euphotic zone. This limitation is reflected in errors in the northeastern Pacific and Arabian Sea, and may explain the errors and biases in the model. Given that our ecosystem model has only one component, the seasonal cycle of biology is not well represented. Moreover, the α values used to obtain export production are too coarse to represent the complete features of the regional ecosystem. For example, Fig. 4 compares the $p\text{CO}_2$ seasonal cycle of our free-run and that of Takahashi

et al. (2009) (black and green lines, respectively), averaged over each of the 14 α regions used in the export production calculation (see also Section 2.3). The subdivisions are the same as those used in table 1 of McKinley et al. (2004). In the present free-run, the tropical to mid-latitude oceans have a reasonable seasonal cycle of $p\text{CO}_2$, although biases are apparent. For example, the seasonal biases in the equatorial Atlantic and Indian Ocean are about 25 μatm . Good agreement between the free-run and the observed $p\text{CO}_2$ seasonal cycle is found for the subtropical South Pacific, subtropical South Atlantic, eastern equatorial Pacific, western equatorial Pacific, subtropical North Pacific and subtropical North Atlantic. However, the model performance is poor in high-latitude oceans (e.g. subpolar North Pacific and subpolar North Atlantic). The free-run seasonal cycle of $p\text{CO}_2$ of the Southern Ocean is in phase with observations, although with relatively weak amplitudes.

In addition to the limitations of the ecosystem model, which explain some of the discrepancies between the model and

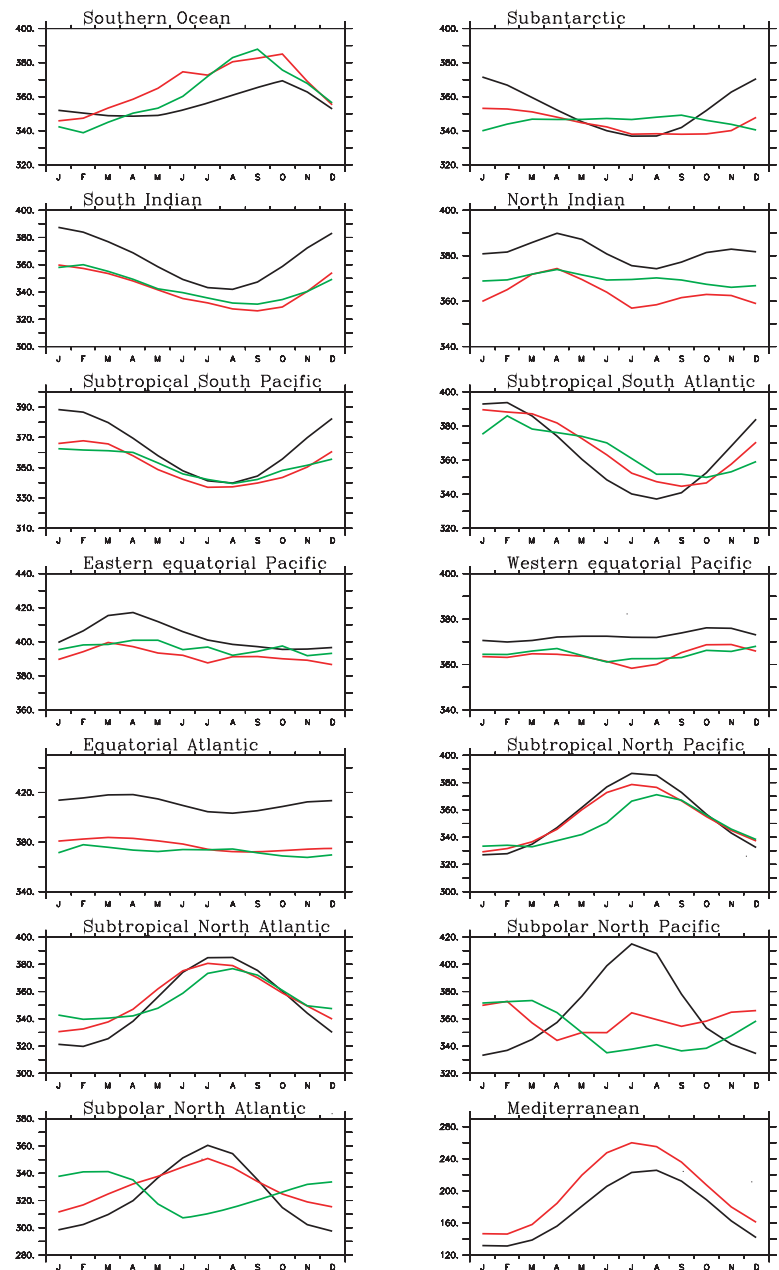


Fig. 4. Seasonal cycle of $p\text{CO}_2$ from 14 α -regions of the biological model are shown. Three lines are shown and they are from the model (black line), Takahashi et al. (2009) (green-line) and the assimilation (red-line). Units are in μatm .

observations, other sources of errors are likely to be associated with biases in surface salt and fresh water fluxes. A more important source of error, especially at high latitudes, arises from alkalinity. The simple linear relation between alkalinity and salinity does not incorporate regional differences in the alkalinity–salinity relation, as noted by Lee et al. (2006). A comparison of the present alkalinity maps with the map provided by Lee et al. (2006), for the 14 regions described earlier, reveals discrepancies in the seasonal cycle of alkalinity at high latitudes. Note that the present model used the alkalinity–salinity relation from OCMIP-II.

3.3. Constraining the surface $p\text{CO}_2$

The upper panel in Fig. 5 shows the interannual variance of air–sea CO_2 flux as resolved in the 16-year period of the free-run. The inverse of this map is normalized to vary between zero and 0.2, and is used for the weighting coefficients (w_2) as a constraint on monthly mean $p\text{CO}_2$ (Fig. 5, bottom panel; see also Section 2.5). This approach guarantees the following conditions: (1) the model biases in the mean seasonal cycle are reduced because the $p\text{CO}_2$ is constrained to a mean cycle, and (2) interannual variability in modelled $p\text{CO}_2$ is relatively weakly influenced by

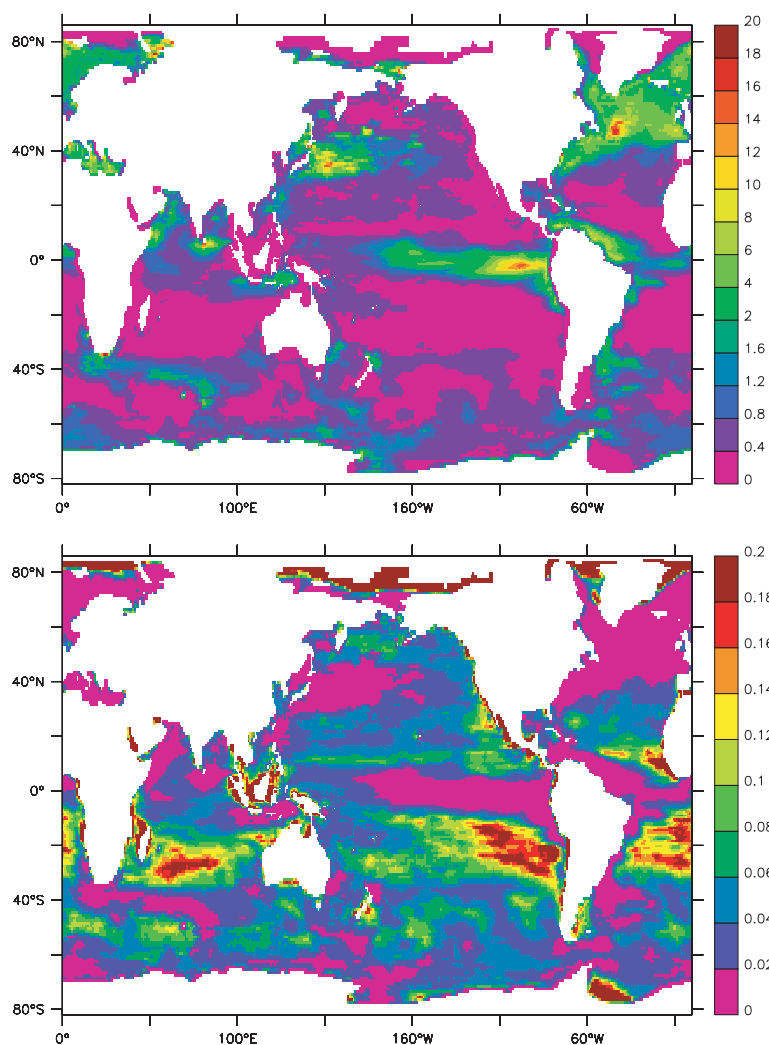


Fig. 5. (Top panel) 1980–1995 interannual variance of air–sea CO₂ flux. Variance is calculated after subtracting the monthly climatology. Units are in $(\text{mole m}^{-2} \text{ yr}^{-1})^2$. (Bottom panel) The weight w_2 used for the climatological constraint (non-dimensional).

this mean constraint because of the inverse variance weighting map employed in this study.

We verified the influence of the climatological constraint on the fit of modelled data to observations along ship tracks. The assimilation was run by switching off the climatological data constraint. The results are analysed for regions with the greatest coverage of data. In the case of no climatological constraint, the cost function along the ship track is reduced by 10% compared with the case of a climatological constraint. The remaining error in the assimilated $p\text{CO}_2$ values due to the climatological constraint is less than $10 \mu\text{atm}$.

The assimilation is run from 1996 to 2004, yielding monthly $p\text{CO}_2$ and air–sea CO₂ flux. The only assimilation in the system is performed for $p\text{CO}_2$. Air–sea fluxes are calculated from the assimilated $p\text{CO}_2$ and gas exchange coefficients. We refer to this calculated air–sea gas exchange as ‘assimilated air–sea CO₂ flux’.

Figure 6 summarizes the assimilated $p\text{CO}_2$ annual mean calculated between 1996 and 2004, as well as the observation data

reported by Takahashi et al. (2007). The assimilation resulted in reduced amplitude errors in annual mean $p\text{CO}_2$. The regional variability of $p\text{CO}_2$ is better represented in the assimilation compared with the free-run (cf. Fig. 1). The elevated amplitudes of $p\text{CO}_2$ in the eastern tropical Pacific are regulated in the assimilation compared with the free-run. The errors in the northern Pacific and Atlantic Ocean are improved in the assimilation, as is the regional $p\text{CO}_2$ distribution in the Southern Ocean.

Figure 7 summarizes the total error remaining in the assimilation compared with the observations reported by Takahashi et al. (2007). Compared with the free-run, an improvement is apparent in the seasonal $p\text{CO}_2$ of the subarctic North Pacific (cf. Fig. 3). This part of the ocean has a dense coverage of $p\text{CO}_2$ sampling contributed by Ship-Of-Opportunity measurements in which the instruments are attached to commercial vessels operating between Japan and North America, and among other Asia-Pacific countries. This wealth of information is readily assimilated into the model, resulting in a marked reduction in errors. Thus, the assimilation performed well in constraining the

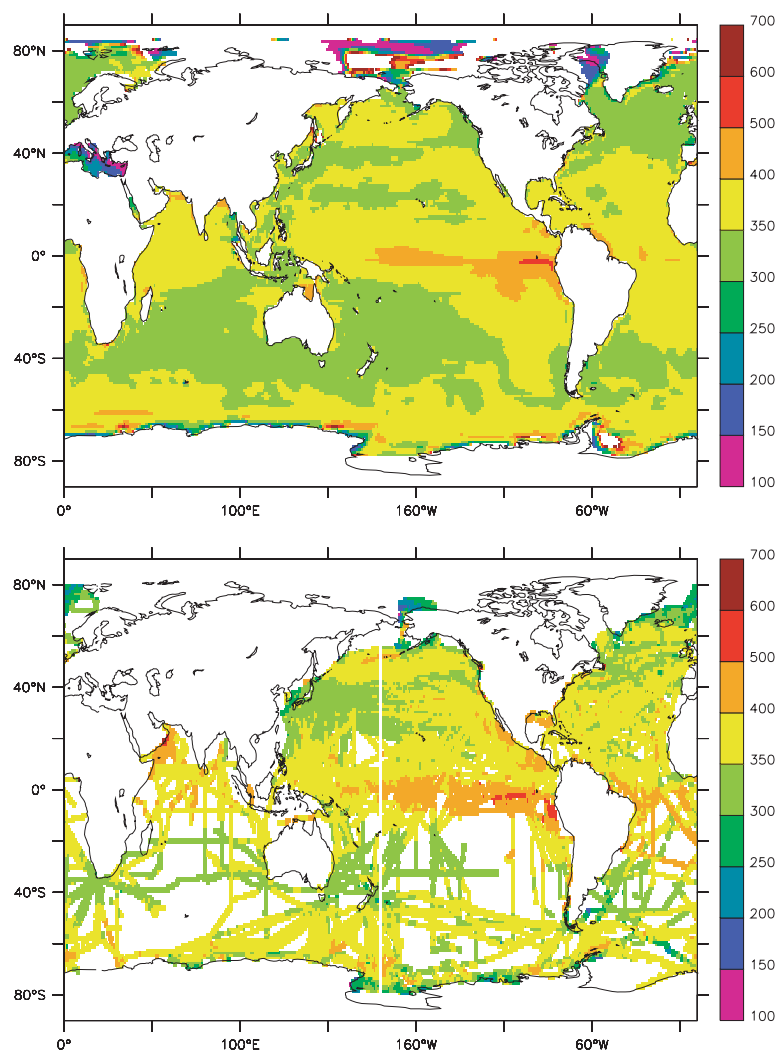


Fig. 6. (Top panel) 1996–2004 mean assimilated surface ocean $p\text{CO}_2$ and (bottom panel) observations from Takahashi et al. (2007) regridded into $1^\circ \times 1^\circ$ bins. Units are in μatm .

model, especially in regions where large amounts of observation data are available.

A comparison of Figs 3 and 7 shows that error reduction in the assimilation is concentrated in the tropical Atlantic, northern Pacific, northern Atlantic and equatorial Pacific. In the eastern equatorial Pacific, however, observations are limited and do not cover a long enough period to resolve interannual variability related to ENSO. Moreover, in this region our model employed only a minimum constraint on the mean $p\text{CO}_2$, depending on the inverse variance map used for weighting. Thus, the amplitude mismatch remains in this region, compared with the data in Takahashi et al. (2007), reflecting interannual signals rather than the residual error of the model.

Figure 4 also compares assimilated $p\text{CO}_2$ as average values derived from the 14 regions used in the biological export calculation (Section 3.2). The assimilation contributed to correcting the phase of the seasonal cycle and resulted in reduced bias; remarkable improvements are seen at high latitudes. For example, the assimilation corrected the seasonal cycle of the sub-polar

North Pacific, because of the large amount of data incorporated from this region. However, the seasonal cycle of the sub-polar North Atlantic was only corrected to a small degree. The LDEO ship-track database (Version 1.0; Takahashi et al., 2007) has not incorporated all the available $p\text{CO}_2$ observations from the North Atlantic.

Figure 8 summarizes the percentage reduction of error in annual mean and seasonal $p\text{CO}_2$ in the assimilation, with respect to the total error in the free-run. The error reductions are calculated as the difference between the free-run error (i.e. Fig. 3) and residual error (i.e. Fig. 7), expressed as a ratio relative to the free-run error. A positive value indicates that the assimilation has improved the model $p\text{CO}_2$ values, with respect to the observed data provided by Takahashi et al. (2007). Sixty percent of the annual mean model biases are eliminated in the assimilation. Forty to sixty percent of the cumulative seasonal errors are also minimized at a regional scale. Marked reductions in error are seen in the tropical Atlantic and North Pacific. In the Indian Ocean, the errors are reduced in both the seasonal and annual

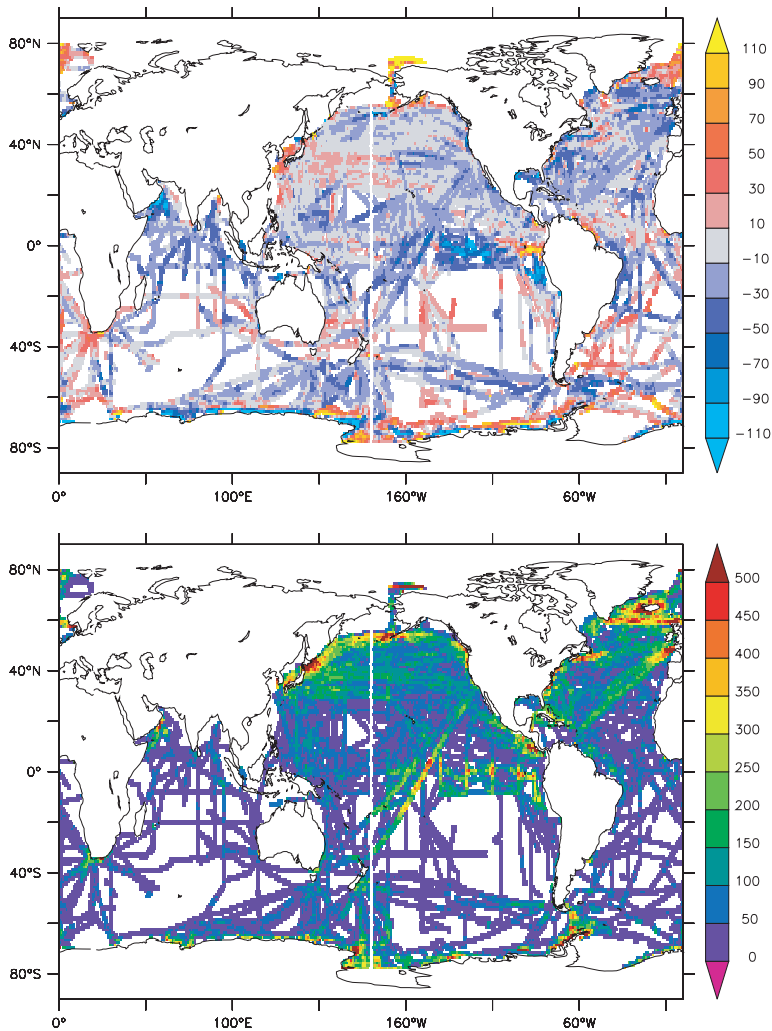


Fig. 7. Same as Fig. 3, but for assimilated run.

mean $p\text{CO}_2$. However, the density of observation tracks within the Indian Ocean is less than that in other oceans.

There are few locations for which the assimilation results in an increase in model $p\text{CO}_2$ error (e.g., regions of the eastern equatorial Pacific). However, large interannual variations in $p\text{CO}_2$ remain in such regions (Fig. 7).

3.4. Conservation of DIC in the assimilation

Here, we consider the following question: To what degree does the adjoint disturb the total balance of globally integrated DIC in the model? The cost function is designed based on the surface ocean $p\text{CO}_2$ mismatch, whereas DIC is adjusted to minimize the cost function. In this case, it is instructive to assess whether the adjoint introduced any DIC imbalance in the model. The only permissible change in DIC within the model is at the surface, via air–sea gas exchange. Although we disturbed the total DIC in the ocean via adjoints, we optimized the near-surface $p\text{CO}_2$ according to observations, which probably resulted in air–sea

fluxes of the desired directions and magnitude. This approach serves the purpose of our assimilation system, which is designed to optimally estimate the net air–sea CO_2 fluxes. The imbalance of DIC produced in the ocean by the adjoints can be considered as corrections to biases in model circulation. In fact, the initial condition taken from GLODAP contains biases, and the model seeks to attain its own equilibrium via the given biogeochemical cycle and transport. In this case, the adjoints compensate for the mismatch in surface ocean DIC (at least at distances for which the adjoints can reach in a 2-month iteration) that originates from the mismatch between modelled and observed $p\text{CO}_2$.

To quantify the model DIC budget and its variations, we calculated the global average DIC concentration (the weighted average in the x , y and z directions) from our assimilation and compared it with area-integrated global air–sea CO_2 fluxes (Fig. 9). The global DIC concentration varies in proportion to the air–sea CO_2 flux. When the air–sea flux is toward the ocean, the DIC concentration increases, and vice versa. This relation is equally apparent in the seasonal cycle (top panel in Fig. 9) and in

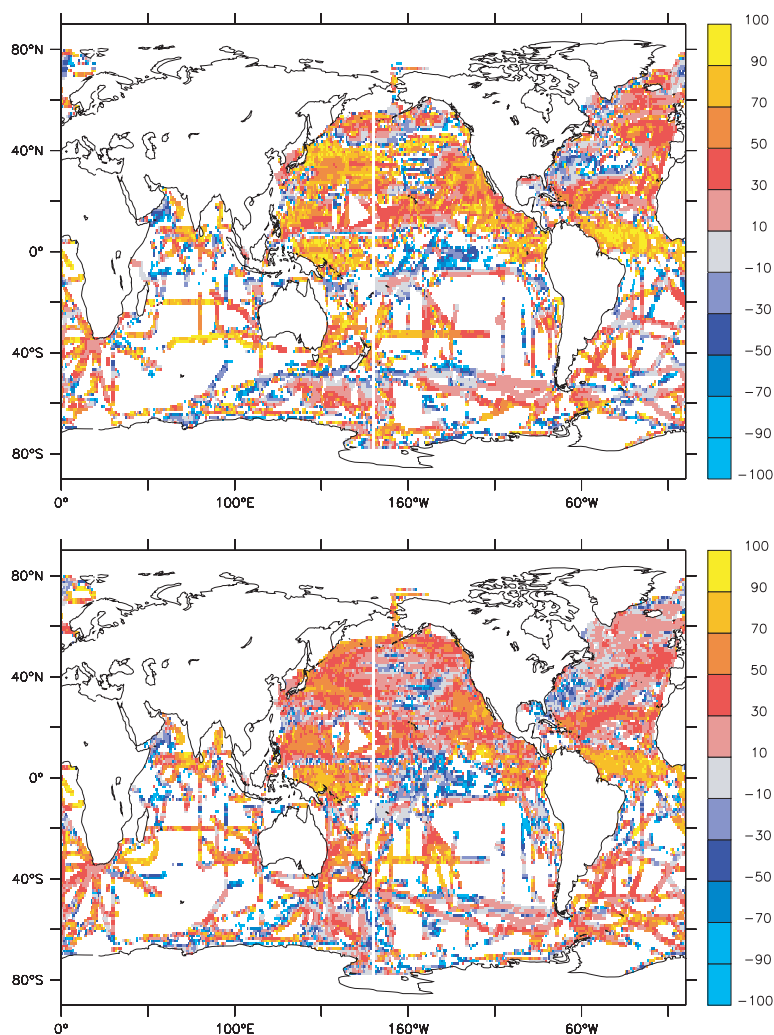


Fig. 8. (Top panel) Percentage error reduction calculated as $(\epsilon_f - \epsilon_a)/\epsilon_f$ for the annual mean $p\text{CO}_2$, where $\epsilon = (p\text{CO}_{2\text{mod}} - p\text{CO}_{2\text{obs}})$ and ϵ_f and ϵ_a corresponds to the error in the free and assimilated run, respectively. (Bottom panel) Same as the top panel but for seasonal cycle in which $\epsilon = (\sum_{i=1}^{12} \text{abs}[p\text{CO}_{2\text{mod}} - p\text{CO}_{2\text{obs}}])$.

interannual variability (bottom panel). The interannual variability of total DIC shows a positive trend from 1998, which can be interpreted as being proportional to increasing CO₂ in the atmosphere and a subsequent increase in oceanic DIC. Although the atmospheric CO₂ was fixed at a constant value in the model, the component of data assimilation could include such trends in the modelled oceanic DIC.

3.5. Assimilated air–sea CO₂ flux

The reduction in biases and errors in the synthesized $p\text{CO}_2$ would enhance the quality of air–sea CO₂ fluxes retrieved from the model. The assimilated CO₂ flux shows an annual mean sink of 1.48 PgC yr^{−1} (1996–2004). The corresponding value from Takahashi et al. (2009) is an annual mean sink of 1.41 PgC yr^{−1}. Thus, the assimilated estimate is closer to the estimate based on observations of Takahashi et al. (2009).

Except for regions of marked interannual variability, the seasonal cycle of the assimilated air–sea CO₂ flux shows a strong

correlation with observed data (Fig. 10). The majority of areas in tropical to high-latitude oceans shows a seasonal correlation above 0.8 (above the 99% level of significance). Biases remain in the assimilation for the equatorial Pacific and Arabian Sea, compared with observations. The Indian Ocean has the poorest coverage of $p\text{CO}_2$ measurements; consequently, the assimilation did not yield any considerable improvements in this region. There are limited observations in the equatorial Pacific with which to calculate the interannual signal; thus, the poor correlation between the seasonal cycle in this region and the observations of Takahashi et al. (2009) does not reflect the models inability; in fact, it may have originated from interannual variability.

The global integral of seasonal CO₂ flux in the assimilation has a minimum sink during June, compared with August and September in the observations presented by Takahashi et al. (2009). The Southern Ocean shows a major phase mismatch in the assimilation, especially during the austral summer. The model performs poorly in representing the sink observed during the austral summer, which may reflect the limitations of the

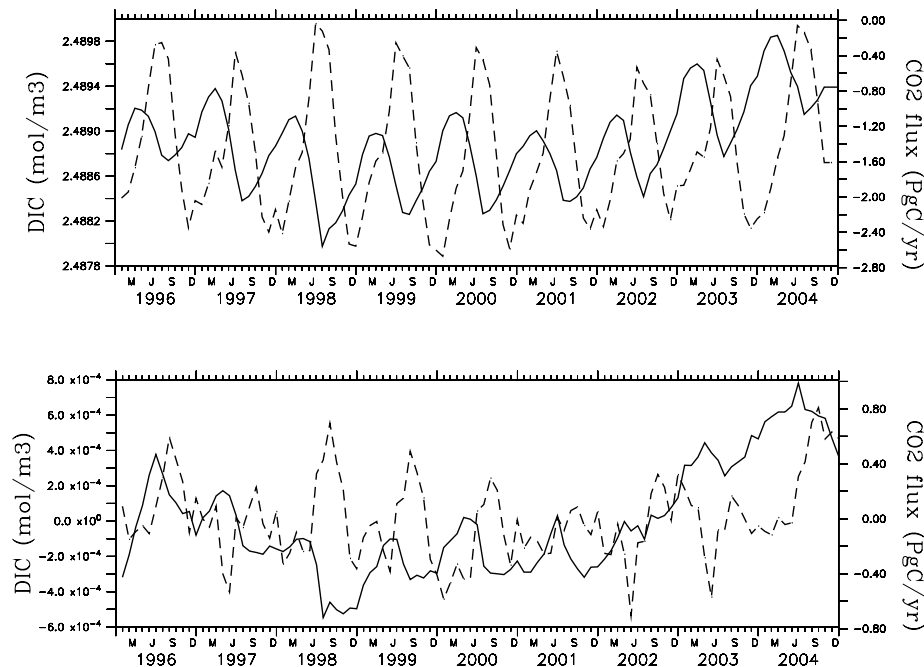


Fig. 9. (Top panel) Globally averaged (weighted average along x -, y - and z -directions) DIC concentrations (full line) and globally integrated air-sea CO_2 fluxes (dash line). Both are from the assimilation. (Bottom panel) Same as top, but for seasonal cycle removed.

ecosystem model regarding the biological cycle. However, the annual mean of the assimilation shows a net sink, as found in the observation data. The mean sink in the Southern Ocean (south of 40°S) in the assimilation is $-0.41 \text{ PgC yr}^{-1}$; the equivalent figure in Takahashi et al. (2009) is $-0.45 \text{ PgC yr}^{-1}$.

Figure 11 (top panel) shows the annual mean CO_2 flux from the assimilation for 1996–2004. The regional patterns of this flux are consistent with the results shown in Takahashi et al. (2009), as a result of two factors: (1) the $p\text{CO}_2$ track observations of Takahashi et al. (2007) are fully utilized in the model to synthesize the surface ocean $p\text{CO}_2$, and (2) the monthly mean map provided by Takahashi et al. (2009) $p\text{CO}_2$ is used as a weak constraint for regions with limited $p\text{CO}_2$ observations. However, the mean constraint was given a weighting of only 20% of the track assimilation, and it is further masked by the inverse of the interannual variability of modelled air-sea CO_2 flux

3.6. Interannual variability of assimilated CO_2 flux

Area-integrated CO_2 fluxes from the global ocean and the Niño region show that the assimilation has weakened the strength of the interannual variability compared with the atmospheric inversions of Patra et al. (2005) and other forward model simulations (Le Quere et al., 2000; Obata and Kitamura, 2003; McKinley et al., 2004). This result may reflect the weak constraint of the model on the monthly mean maps of $p\text{CO}_2$, which could suppress the interannual signals. In addition, the amplitudes of interannual signals in the assimilation are smaller than those in

the present free-runs. The observation data reported by Feely et al. (2002) and the inverse modelling of Patra et al. (2005) and several OGCMs (Le Quere et al., 2000; Obata and Kitamura, 2003; McKinley et al., 2004) show stronger interannual variability than that obtained in this study. However, the magnitude of the interannual signal in the OGCM results is only half that in the atmospheric inversions.

The Niño region shows an interannual anomaly with a magnitude of 0.4 PgC yr^{-1} during the 1997–1998 and 2003–2004 El Niño events. This is smaller in magnitude than the inversions reported by Patra et al. (2005). The years 2002–2004 were marked by weak El Niño conditions in the tropical Pacific, and the CO_2 flux anomalies for these years are negative. Thus, the assimilated CO_2 flux anomalies from the eastern tropical Pacific are consistent with observations, as well as with our general consensus regarding the evolution of CO_2 flux during El Niño years.

4. Comparison with other data sources

Here, we compare our assimilated air-sea CO_2 flux with other data sources. The following sources are used for the intercomparison: the atmospheric inversions reported by Rödenbeck et al. (2003) (Version s96_v3.1) and Patra et al. (2005), and the biogeochemical general circulation model fluxes of Le Quere et al. (2005). The seasonal cycle of global air-sea CO_2 fluxes is compared among these four products, as well as the seasonal cycle

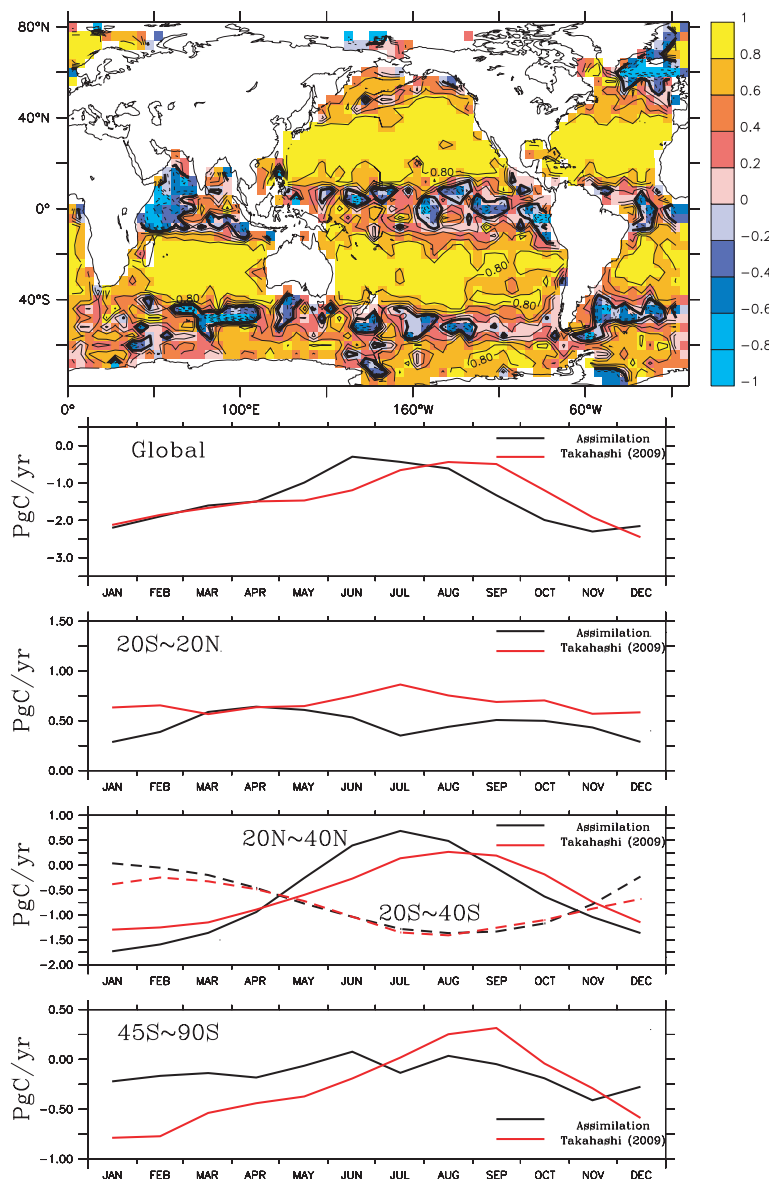


Fig. 10. (Top panel) Seasonal correlation coefficients calculated between assimilated sea-air CO₂ fluxes and Takahashi et al. (2009). The line plots compare the seasonal cycle of area integrated sea-air CO₂ fluxes from assimilation and Takahashi et al. (2009).

of air-sea CO₂ flux in the North Pacific, because our assimilated CO₂ flux data incorporate a large amount of observed data available from the North Pacific.

The seasonal cycle of globally integrated air-sea CO₂ fluxes differs considerably among the above four products (Fig. 12). The assimilated fluxes show a minimum sink during the boreal summer. The seasonal cycle of Le Quere et al. (2005) shows a minimum sink during September and October, and a maximum sink during April and May, consistent with the findings of Takahashi et al. (2009). Among the atmospheric inversions, Rödenbeck et al. (2003) reported a seasonal cycle that is largely opposite to that of the present study and to observations. The oceanic seasonal cycle in the inversion performed by Rödenbeck et al. (2003) is probably affected by land signals, as such sig-

nals are much larger than oceanic signals and it is generally difficult to separate the two. Moreover, the Rödenbeck et al. (2003) inversion does not employ any prior flux for seasonality. The inversion fluxes reported by Patra et al. (2005) show a seasonal cycle consistent with that reported by Takahashi et al. (2009).

The error bars in Fig. 12 show the interannual standard deviations for each month of the year. The interannual amplitudes are weakest in our assimilated air-sea CO₂ fluxes, possibly because of the weak mean constraint applied to the model. The standard deviations are comparable between our product and Le Quere et al. (2005). Interannual variability is greater in the atmospheric inversions performed by Patra et al. (2005) compared to other products compared here. The inversion fluxes

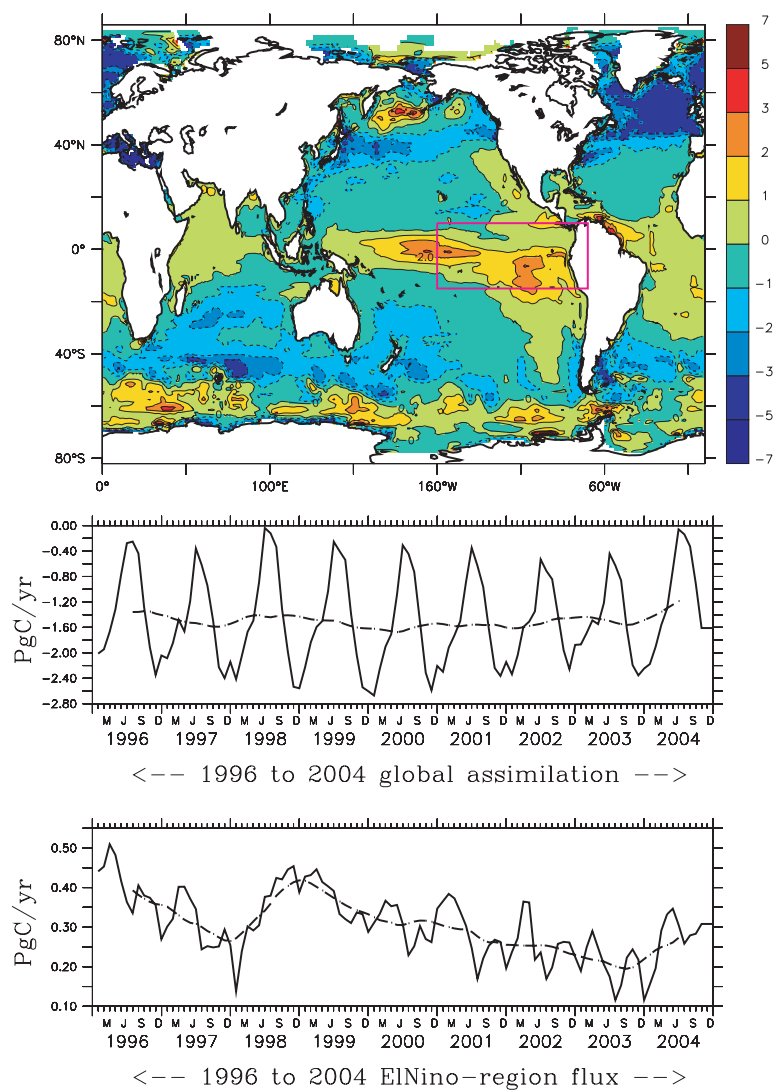


Fig. 11. (Top panel) 1996–2004 mean assimilated sea-air CO_2 fluxes. Units are in $\text{mol m}^{-2} \text{yr}^{-1}$. (Middle panel) Interannual CO_2 fluxes from the global ocean as given by the assimilation. (Bottom panel) Area integrated air–sea CO_2 flux from the Niño region (area is shown as box in the top panel). Dashed line represents a 12-month running mean.

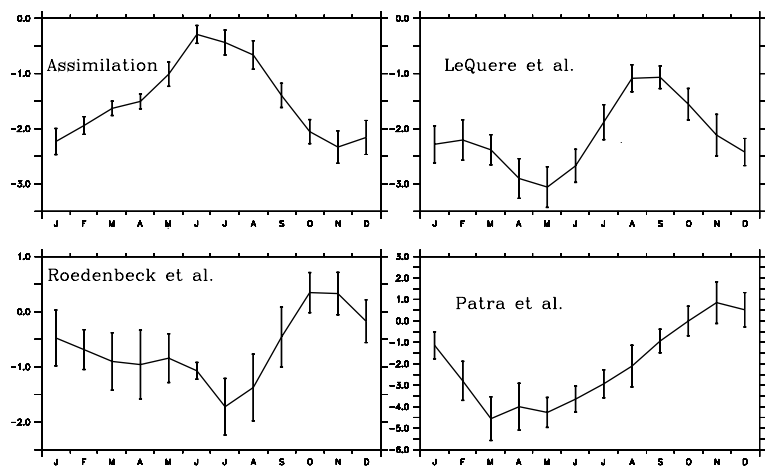


Fig. 12. Seasonal cycle of global air–sea CO_2 fluxes. Units are in PgC yr^{-1} . The error bars show the interannual standard deviations of each month calculated from a span of 1996–2004. The y-axis varies for each data product.

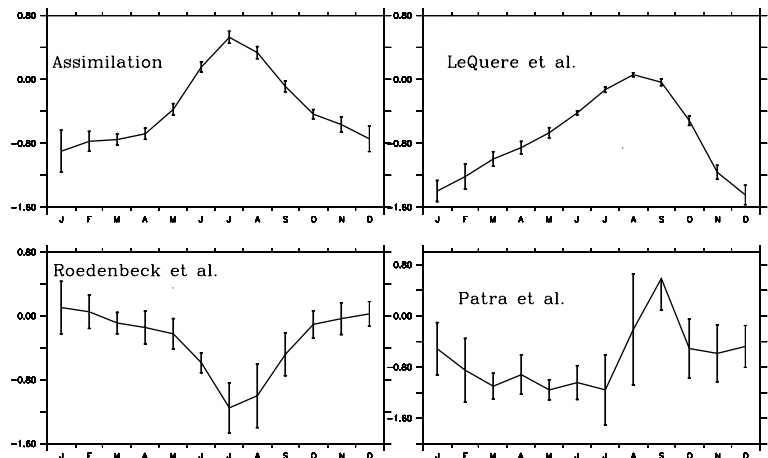


Fig. 13. Same as Fig. 12 but for fluxes from north Pacific alone.

calculated by Rödenbeck et al. (2003) show the largest interannual amplitudes among the four products.

The seasonal cycle of the North Pacific alone is worth comparing because our model benefited from a large number of observations in this region. Figure 13 shows the integrated air-sea CO₂ fluxes from the four data sources, for the North Pacific area. The ocean models show comparable seasonal cycles. The error bars in the figure indicate the interannual standard deviations for each month. The amplitude of the interannual signal in the North Pacific CO₂ flux is weaker in the OGCM of Le Quere et al. (2005), and is weaker during the boreal summer in our data product. This finding is consistent among the two ocean models. Among the atmospheric inversions, Patra et al. (2005) reported large interannual variability. The seasonal cycle reported by Patra et al. (2005) is comparable to that in the ocean models. However, the seasonal cycle in atmospheric inversion fluxes reported by Rödenbeck et al. (2003) is inconsistent with that of the other products.

4.1. Intercomparison of interannual variability

This section compares the interannual anomalies of area-integrated air-sea CO₂ fluxes obtained from four data sources. First, we compare the interannual variability in the North Pacific. Figure 14 shows a remarkable agreement between the assimilated air-sea CO₂ fluxes of the present study and atmospheric

inversions; the cycles of interannual anomalies are in phase in all cases. The magnitude of interannual CO₂ flux anomalies in the atmospheric inversion reported by Patra et al. (2005) is larger than those from other products, and is shown in the figure using a separate y-axis. Except for the relatively high amplitudes in Patra et al. (2005), the atmospheric inversions generally agree with the interannual variability of assimilated air-sea CO₂ fluxes. The inversion performed by Rödenbeck et al. (2003) yields a similar interannual cycle to that of the present assimilation, although the seasonal cycles are opposite. Given that the inversion method employed by Rödenbeck et al. (2003) is linear, seasonal and interannual variability are possibly independent of each other, meaning that the comparison of interannual variability may be reasonable even if the seasonality is different (C. Rödenbeck, 2009, personal communications). The comparison demonstrates that the global atmospheric inversions agree with the assimilated air-sea CO₂ fluxes of the present study. The interannual variability derived from an OGCM by Le Quere et al. (2005), in contrast, is weak and out of phase compared with the other data sources.

Figure 15 compares the globally integrated air-sea CO₂ flux interannual anomalies between three products: the assimilated air-sea CO₂ fluxes of the present study, the atmospheric inversions of Rödenbeck et al. (2003), and OGCM results presented by Le Quere et al. (2005). We excluded Patra et al. (2005) from this comparison because of the relatively large amplitude of the

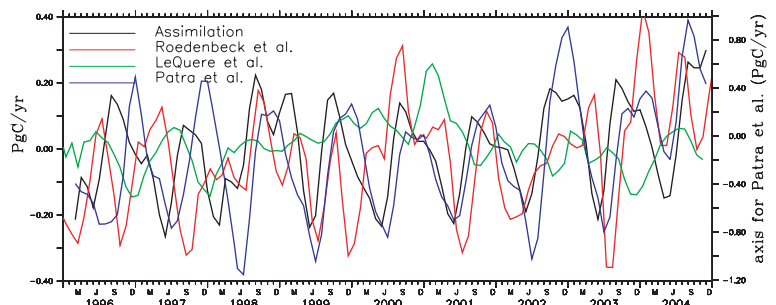


Fig. 14. Interannual variabilities of the north Pacific area integrated air-sea CO₂ flux anomalies derived from the four data sets.

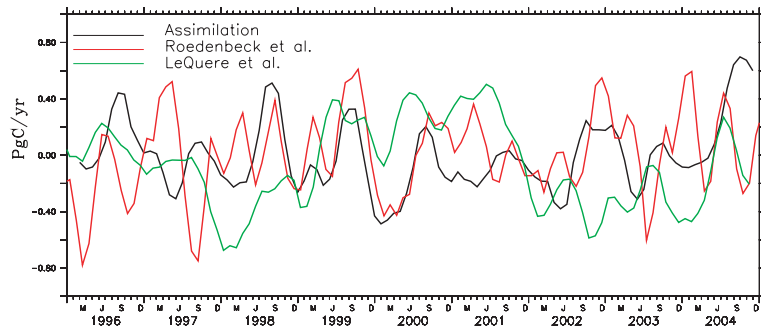


Fig. 15. Same as Fig. 14 but for global anomalies.

anomalies in their study. The three data products show comparable interannual variability (both in magnitude and phase). The assimilation system of the present study agrees with the atmospheric inversions on interannual scales. The assimilated fluxes offer a better comparison with atmospheric inversions than do the OGCM results of Le Quere et al. (2005).

5. Discussion and Conclusion

This study differs from previous attempts at modelling of the ocean carbon cycle in the following key aspects. (1) We used an offline tracer transport model for physical circulation, which uses observationally constrained reanalysis data products. This approach performs well in representing carbon transport in the ocean. (2) The ecosystem model, although a simple model derived from McKinley et al. (2004), is acceptable in terms of representing interannual variability in air–sea CO_2 fluxes and was implemented to constrain values of surface ocean $p\text{CO}_2$ with available observations. (3) The assimilation resulted in improved model performance, yielding a reduction in systematic biases and errors. (4) An intercomparison of the present data product with atmospheric inversions revealed reasonable agreement in the interannual variability of air–sea fluxes. (5) Ultimately, we obtained an observationally constrained air–sea CO_2 flux on an interannual time scale for the global ocean.

The use of reanalysis physical circulation enables us to extend the assimilated CO_2 flux data in a near-real-time basis. This approach provides a wealth of information in terms of ocean flux priors for upcoming atmospheric inversions using CO_2 measurements made by the on-board instrument attached to GOSAT. Thus, the proposed model has the potential to contribute inter-annually varying near-real-time ocean flux priors, constrained by available observations of $p\text{CO}_2$ and with reduced bias, for near-real-time atmospheric inversions.

The proposed model can be used to set up for the operational forecasts of CO_2 fluxes, depending on the availability of operational ocean circulation. The recent increase in the amount of $p\text{CO}_2$ observation data is helpful in setting up this model for operational forecasts of CO_2 fluxes. Moreover, autonomous instruments such as Argo may contain sensors for DIC and $p\text{CO}_2$ in the future. In such a case, the proposed model could be used

to provide continuously updated, observationally constrained air–sea CO_2 flux data.

5.1. Limitations

The method used to constrain values of surface ocean $p\text{CO}_2$ contains a few uncertainties. Accordingly, we discuss the limitations of the present system, with a view to improving the proposed CO_2 assimilation system.

First, the model uses only one adjoint, meaning that the control variable (DIC) is corrected irrespective of the source of errors. This assimilation system considers that DIC determines the surface ocean $p\text{CO}_2$ in the model, given that SST, SSS and surface alkalinity are close to observed values. However, this assumption has caveat because errors may occur in these variables even though they are derived from the reanalysis offline inputs. The modelled DIC is corrected to match $p\text{CO}_2$ values with observations; however, there exists little guidance regarding whether the correction is in the desired direction. To verify that the corrections applied to DIC are valid, we compared the assimilated DIC with observations in several sectors of the Pacific. In most cases, the assimilation corrected the DIC in the desired direction. As an example, Fig. 16 shows the surface ocean DIC from three cruises carried out in the North Pacific. The figure compares the assimilated DIC of the surface ocean with observations, revealing a reasonable agreement.

Another limitation of the system is the short term (2 months) of the assimilation windows. We found that the adjoint loses its sensitivity in longer backward runs. Here, the data are considered only at the surface ocean, where vertical mixing is maximized. Therefore, the model-data misfit shows a relatively rapid downward “dilution”. During the 2-month period of adjoint runs, the sensitivities are not propagated to depths greater than the euphotic zone. Therefore, the correction applied to the surface ocean DIC is confined to within 100 m of the surface ocean. This limits the assimilation system in terms of using surface ocean $p\text{CO}_2$ observations to eliminate errors in simulated DIC below 100 m depth. However, we cannot be sure that surface ocean $p\text{CO}_2$ is indeed controlled by deep DIC. Vertical mixing plays a crucial role in bringing the subsurface DIC signal to the surface and thereby influencing $p\text{CO}_2$. However, the present

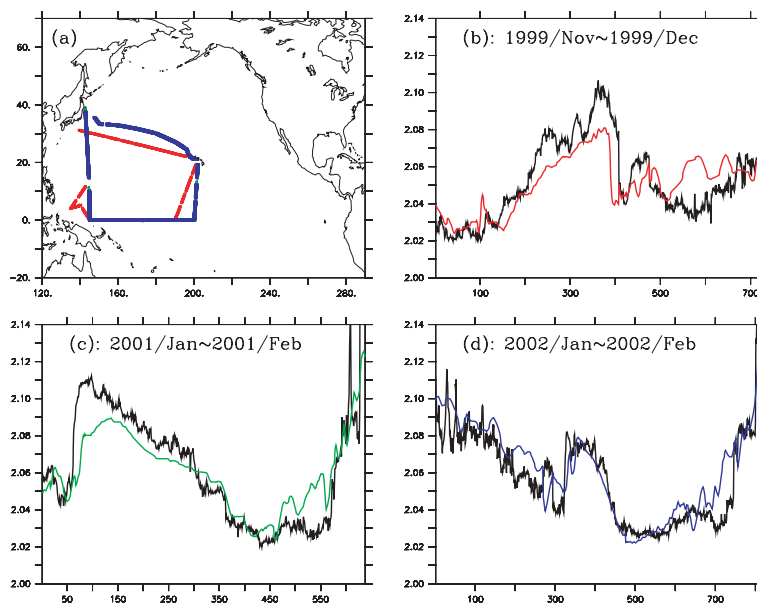


Fig. 16. (a) Three cruises from the western north Pacific which sampled the surface ocean DIC. (b–d) Comparison of observed (black) and assimilated (colour) DIC concentrations. Units are in mole m⁻³.

assimilation system only considers corrections that are possible within the distance across which adjoints can propagate in a 2-month iteration. Moreover, DIC is a non-conservative tracer, which may contribute to the weak sensitivity of adjoints to the longer backward integration.

Although the present method has limitations, it represents an important step toward additional future improvements. The incorporation of satellite-derived chlorophyll-*a* data into the assimilation would be a promising future step, along with expanding the ecosystem model to include the full dynamics of alkalinity.

6. Acknowledgments

This work was carried out as part of the GOSAT carbon cycle research project at CGER, NIES. Computational resources were provided by supercomputer facility at NIES. The offline fields for the physical model were provided by GFDL, Princeton, NJ. The authors are thankful to the comments received from Andrew Jacobson, NOAA, U.S.A. and one anonymous reviewer which were helpful to improve the quality of the paper. The paper is proof read by Stallard Scientific Editing, Nelson, New Zealand. Global ocean air–sea CO₂ flux data from 1996 to 2004 are available for research purposes; please contact vinu.valsala@nies.go.jp.

References

- Baker, D. F., Doney, S. C. and Schimel, D. S. 2006. Variational data assimilation for atmospheric CO₂. *Tellus* **58B**, 359–365.
- Bennett, A. F. 2002. *Inverse Modeling of the Ocean and Atmosphere*. Cambridge University Press, New York, NY, 234 pp.
- Bryan, K. and Lewis, L. J. 1979. A watermass model of the World ocean. *J. Geophys. Res.* **84**, 2503–2517.
- Delworth, T. L., Broccoli, A. J., Rosati, A., Stouffer, R. J., Balaji, V. and co-authors. 2006. GFDL's CM2 Global coupled climate models. Part I: Formulation and simulation characteristics. *J. Clim.* **19**, 643–674.
- Dutkiewicz, S., Marshall, M. F. J. and Gregg, W. W. 2001. Interannual variability of phytoplankton abundances in the north Atlantic. *Deep-Sea Res.* **48**, 2323–2344.
- Feely, R. A., Boutin, J., Cosca, E. C., Dandonneau, Y., Etcheto, J. and co-authors. 2002. Seasonal and interannual variability of CO₂ in the equatorial Pacific. *Deep-Sea Res.* **14**, 2443–2469.
- Feely, R. A., Wanninkhof, R., Takahashi, T. and Tans, P. 1999. Influence of El Niño on the equatorial Pacific contribution to atmospheric CO₂ accumulation. *Nature* **398**, 365–386.
- Fukumori, I., Cheng, T. L. and B. and Menemenlis, D. 2004. The origin, pathway and destination of Niño-3 water estimated by a simulated passive tracer and its adjoint. *J. Phys. Oceanogr.* **34**, 582–604.
- Gent, P. R. and McWilliams, J. C. 1990. Isopycnal mixing in ocean circulation models. *J. Phys. Oceanogr.* **20**, 150–155.
- Gnanadesikan, A., Dixon, K. W., Griffies, S. M., Balaji, V., Barreiro, M. and co-authors. 2006. GFDL's CM2 global coupled climate models. Part II: The baseline ocean simulation. *J. Clim.* **19**, 675–697.
- Griffies, S. M. and Hallberg, R. W. 2000. Bi-harmonic friction with a Smagorinsky-like viscosity for use in large-scale eddy-permitting ocean models. *J. Phys. Oceanogr.* **128**, 2935–2946.
- Gruber, M., Gloor, M., Fletcher, S. E. M., Doney, S. C., Dutkiewicz, S. and co-authors. 2009. Oceanic sources, sinks and transport of atmospheric CO₂. *Global Biogeochem. Cycles* **23**, doi:10.1029/2008GB003349.
- Gurney, K. R., Law, R. M., Denning, A. S., Rayner, P. J., Pak, B. and Transcom-3-L2-modelers 2004. Transcom-3 inversion intercomparison: control results for the estimation of seasonal carbon sources and sinks. *Global Biogeochem. Cycles* **18**, doi:10.1029/2003GB002111.
- Hourdin, F. and Talagrand, O. 2002. Eulerian backtracking of atmospheric tracers: I adjoint derivation and parametrization of subgrid-scale transport. *Quart. J. Roy. Meteor. Soc.* **128**, doi:10.1256/qj.yy.n.

- Hourdin, F., Talagrand, O. and Idelkadi, A. 2002. Eulerian backtracking of atmospheric tracers: II Numerical aspects. *Quart. J. Roy. Meteor. Soc.* **128**, doi:10.1256/qj.yy.n.
- Ikeda, M. and Sasai, Y. 2000. Reconstruction of subsurface DIC and alkalinity fields in the north pacific using assimilation of upper ocean data. *Mar. Chem.* **72**, 343–358.
- Jacobson, A. R., Fletcher, S. E. M., Gruber, N., Sarmiento, J. L. and Gloor, M. 2007. A joint atmosphere-ocean inversion for surface fluxes of carbon dioxide: 2. Regional results. *Global Biogeochem. Cycles* **21**, doi:10.1029/2006GB002703.
- Keeling, C. D., Bacastow, R. B. and Whorf, T. P. 1982. Measurements of the concentration of carbon dioxide at mauna loa observatory, Hawaii. *Carbon Dioxide Review* **110**, Oxford University Press, New York, NY, pp. 377–385.
- Keeling, C. D., Brix, H. and Gruber, N. 2004. Seasonal and long-term dynamics of the upper ocean carbon cycle at station ALOHA near Hawaii. *Global Biogeochem. Cycles* **110**, doi:10.1029/2004GBC002227.
- Keeling, C. D., Piper, S. C., Bacastow, R. B., Wahlen, M., Whorf, T. P. and co-authors. 2001. Exchanges of atmospheric CO₂ and $\delta^{13}\text{C}$ with the terrestrial biosphere and oceans from 1978 to 2001. Global aspects. Reference Series: 01-06 Scripps Institute of Oceanographs, La Jolla, California.
- Key, R. M., Kozyr, A., Sabine, C. L., Lee, K., Wanninkhof, R. and co-authors. 2004. A global ocean carbon climatology: results from Global Ocean Data Analysis Project (GLODAP). *J. Geophys. Res.* **18**, doi:10.1029/2004GB002247.
- Large, W. G., McWilliams, J. C. and Doney, S. C. 1994. Ocean vertical mixing: a review and a model with a nonlocal boundary layer parameterization. *Rev. Geophys.* **32**, 363–403.
- Large, W. G. and Yeager, S. G. 2008. The global climatology of an interannually varying air-sea flux data set. *Climate Dyn.* doi:10.1007/s00382-008-0441-3.
- Le Quere, C., Harrison, S. P., Prentice, I. C., Buitenhuis, E. T., Aumont, O. and co-authors. 2005. Ecosystem dynamics based on plankton functional types for global ocean biogeochemistry models. *Global Change Biol.* **11**, 2016–2040.
- Le Quere, C., Orr, J. C., Monfray, P., Aumont, O. and Madec, G. 2000. Interannual variability of the oceanic sink of CO₂ from 1979 through 1997. *Global Biogeochem. Cycles* **14**, 1247–1265.
- Le Quere, C., Rödenbeck, C., Buitenhuis, E. T., Conway, T. J., Gomez, R. L. and A. and co-authors. 2007. Saturation of the southern ocean CO₂ sink due to recent climate change. *Science* **316**, 1735–1738.
- Lee, K., Tong, L. T., Millero, F. J., Sabine, C. L., Dickson, A. G. and co-authors. 2006. Global relationships of total alkalinity with salinity and temperature in surface waters of the world's oceans. *Geophys. Res. Lett.* **33**, doi:10.1029/2006GL027207.
- Lenton, A. and Matear, R. 2007. Role of the Southern Annular Mode (SAM) in southern ocean CO₂ uptake. *Global Biogeochem. Cycles* **21**, doi:10.1029/2006GB002714.
- Maksyutov, S., Nakatsuka, Y., Valsala, V., Saito, M., Kadyrov, N. and co-authors. 2010. Algorithms for carbon flux estimation using GOSAT observational data. *CGER'S Supercomput. Monogr.* **15**, 1–112.
- McKinley, G. A., Follows, M. J. and Marshall, J. 2004. Mechanism of air-sea CO₂ flux variability in the equatorial Pacific and North Atlantic. *Global Biogeochem. Cycles* **18**, doi:10.1029/2003GB002179.
- Obata, A. and Kitamura, Y. 2003. Interannual variability of the air-sea exchange of CO₂ from 1961 to 1998 simulated with a global ocean circulation-biogeochemistry model. *J. Geophys. Res.* **108**, doi:10.1029/2001JC001088.
- Orr, J. C., Najjar, R., Sabine, C. L. and Joos, F. 1999. *Abiotic-HOWTO*. LSCE/CEA Saclay, Gif-sur-Yvette, France, 25 pp.
- Patra, P. K. and Maksyutov, S. 2002. Incremental approach to the optimal network design for CO₂ surface source inversion. *Geophys. Res. Lett.* **29**, doi:10.1029/2001GL013943.
- Patra, P. K., Maksyutov, S., Ishizawa, M., Nakazawa, T., Takahashi, T. and co-authors. 2005. Interannual and decadal changes in the air-sea CO₂ flux from atmospheric CO₂ inverse modeling. *Global Biogeochem. Cycles* **19**, doi:10.1029/2004GB002257.
- Rödenbeck, C., Houweling, S., Gloor, M. and Heimann, M. 2003. CO₂ flux history 1982–2001 inferred from atmospheric data using a global inversion of atmospheric transport. *Atmos. Chem. Phys.* **3**, 1919–1964.
- Takahashi, T., Sutherland, S. C. and Kozyr, A. 2007. Global ocean surface water partial pressure of CO₂ database: measurements performed during 1968–2006 (Version 1.0). ornl/cdiac-152 NDP-08. Carbon Dioxide Information Analysis Center 20.
- Takahashi, T., Sutherland, S. C., Wanninkhof, R., Sweeney, C., Feely, R. A. and co-authors. 2009. Climatological mean and decadal changes in surface ocean pCO₂ and net sea-air CO₂ flux over the global oceans. *Deep-Sea Res. II* **56**, 554–577.
- Thomas, H., Prowe, A. E. F., Lima, I. D., Doney, S. C., Wanninkhof, R. and co-authors. 2008. Changes in the north Atlantic oscillation influence CO₂ uptake in the north atlantic over the past 2 decades. *CGER'S Supercomput. Monogr.* **22**, doi:10.1029/2007GB003167.
- Tjiputra, J. F., Polzin, D. and Winguth, A. E. 2007. Assimilation of seasonal chlorophyll and nutrient data into an adjoint three-dimensional ocean carbon cycle model: Sensitivity analysis and ecosystem parameter optimization. *Global Biogeochem. Cycles* **21**, doi:10.1029/2006GB002745.
- Uppala, S., Kallberg, P., Hernandez, A., Saarinen, S., Fiorino, M. and co-authors. 2004. ERA-40. ECMWF 45-year reanalysis of the global atmosphere and surface conditions 1957–2002. *ECMWF Newsl.* **101**, 2–21.
- Valsala, K. V., Maksyutov, S. and Ikeda, M. 2008. Design and validation of an offline oceanic tracer transport model for a carbon cycle study. *J. Clim.* **21**, 2752–2769.
- Wanninkhof, R. 1992. Relationship between wind speed and gas exchange over the ocean. *J. Geophys. Res.* **97**, 7373–7382.
- Weiss, R. F. 1974. Carbon dioxide water and sea water: the solubility of a non-ideal gas. *Mar. Chem.* **2**, 203–215.
- Wetzel, P., Winguth, A. and Maier-Reimer, E. 2005. Sea to air CO₂ flux from 1948 to 2003. A model study. *Global Biogeochem. Cycles* **19**, doi:10.1029/2004GB002339.
- Yumimoto, K. and Uno, I. 2006. Adjoint inverse modeling of CO₂ emissions over eastern Asia using four-dimensional variational data assimilation. *Atmos. Environ.* **40**, doi:10.1016/j.atmosenv.2006.05.042.
- Zeng, J., Nojiri, Y., Murphy, P. P., Wong, C. S. and Fujinuma, Y. 2002. A comparison of $\delta p\text{CO}_2$ distributions in the northern north Pacific using results from a commercial vessel in 1995–1999. *Deep-Sea Res.* **49**, 5303–5315.