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Inverse estimates of anthropogenic CO₂ uptake, transport, and storage by the ocean

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[1] Regional air-sea fluxes of anthropogenic CO₂ are estimated using a Green's function inversion method that combines data-based estimates of anthropogenic CO₂ in the ocean with information about ocean transport and mixing from a suite of Ocean General Circulation Models (OGCMs). In order to quantify the uncertainty associated with the estimated fluxes owing to modeled transport and errors in the data, we employ 10 OGCMs and three scenarios representing biases in the data-based anthropogenic CO₂ estimates. On the basis of the prescribed anthropogenic CO₂ storage, we find a global uptake of 2.2 ± 0.25 Pg C yr⁻¹, scaled to 1995. This error estimate represents the standard deviation of the models weighted by a CFC-based model skill score, which reduces the error range and emphasizes those models that have been shown to reproduce observed tracer concentrations most accurately. The greatest anthropogenic CO₂ uptake occurs in the Southern Ocean and in the tropics. The flux estimates imply vigorous northward transport in the Southern Hemisphere, northward cross-equatorial transport, and equatorward transport at high northern latitudes. Compared with forward simulations, we find substantially more uptake in the Southern Ocean, less uptake in the Pacific Ocean, and less global uptake. The large-scale spatial pattern of the estimated flux is generally insensitive to possible biases in the data and the models employed. However, the global uptake scales approximately linearly with changes in the global anthropogenic CO₂ inventory. Considerable uncertainties remain in some regions, particularly the Southern Ocean.

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1. Introduction

[2] It is estimated that the Earth's oceans have absorbed about $48 \pm 9\%$ of the CO₂ emitted over the industrial period

(1880–1994) from fossil fuel consumption and cement production [Sabine *et al.*, 2004]. Accurate, quantitative assessments of the spatial pattern of the air-sea flux of anthropogenic CO₂ are needed to improve our understanding of the physical processes controlling this uptake. However, there are substantial uncertainties associated with current estimates of these fluxes.

[3] The exchange of anthropogenic CO₂ across the air-sea interface cannot be measured directly. However, the total air-sea CO₂ exchange can be determined from observations of the difference between the partial pressures of CO₂ in the atmosphere and the surface ocean, *p*CO₂, and a formulation of the air-sea gas exchange coefficient [e.g., Takahashi *et al.*, 2002]. No method is currently available to measure the component of the air-sea exchange that is attributable to the anthropogenic perturbation of the atmospheric CO₂ concentration, although this quantity has been separated from the observations in the Indian Ocean using a method related to the one presented here [Hall and Primeau, 2004]. The spatial pattern of the oceanic uptake of anthropogenic CO₂ has traditionally been estimated using Ocean General Circulation Models (OGCMs) [e.g., Orr *et al.*, 2001; Murnane *et al.*, 1999; Sarmiento *et al.*, 1992].

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[4] The tracer-based ΔC^* method is used extensively to separate the concentration of anthropogenic CO_2 in the ocean from ocean interior observations of dissolved inorganic carbon (DIC) and other tracers [Gruber *et al.*, 1996]. This technique has been employed to calculate regional and global inventories of anthropogenic CO_2 storage in the ocean [e.g., Lee *et al.*, 2003; Gruber, 1998; Sabine *et al.*, 1999, 2002], and a global summary was presented by Sabine *et al.* [2004]. However, while this method provided many new insights into anthropogenic CO_2 storage, by itself it cannot be used to quantitatively assess the air-sea fluxes and oceanic transport of anthropogenic CO_2 .

[5] Recently, an approach has been developed to estimate surface fluxes from ocean interior data [Gloor *et al.*, 2001; Gruber *et al.*, 2001; Gloor *et al.*, 2003]. This approach uses a Green's function inverse method analogous to atmospheric tracer inversions [e.g., Enting and Mansbridge, 1989; Tans *et al.*, 1990; Bousquet *et al.*, 2000] to infer regional air-sea fluxes from ocean interior observations and OGCMs that are used to determine how surface fluxes influence tracer concentrations in the interior ocean.

[6] The inverse approach is appealing because the flux estimates are driven by data and because it is independent of bulk formulations, such as the parameterization of the air-sea gas exchange coefficient needed to estimate air-sea fluxes from measurements of the air-sea partial pressure difference [e.g., Takahashi *et al.*, 2002]. The application of this inversion method to the anthropogenic CO_2 problem is aided by the fact that the large-scale spatial footprints of anthropogenic CO_2 uptake are well preserved in the oceans owing to the long timescales of ocean circulation. However, there are several important sources of uncertainty associated with this method that have not been addressed. Comparisons between heat and oxygen flux estimates using three different OGCMs suggested that model transport is one of the largest sources of uncertainty in the inverse estimates [Gloor *et al.*, 2001; Gruber *et al.*, 2001]. There are also several sources of uncertainty associated with the estimates of anthropogenic CO_2 used to constrain the inversion [Gruber *et al.*, 1996; Matsumoto and Gruber, 2005; Keeling, 2005; Sabine and Gruber, 2005]. A third issue that needs to be considered is the aggregation error, which is caused by the assumption that fluxes within a large spatial region are proportional to a prescribed spatial pattern [Kaminski *et al.*, 2001]. In addition, the inversion implicitly assumes that ocean circulation was approximately steady over the last 2 centuries and that the only source of temporal variability in the oceanic uptake of anthropogenic CO_2 is the atmospheric CO_2 perturbation.

[7] The aim of this paper is to extend the first estimates of Gloor *et al.* [2003] by estimating the air-sea fluxes of anthropogenic CO_2 with a refined method, address the uncertainties and robustness of these estimates, and explore the oceanic transport of anthropogenic CO_2 implied by the surface fluxes. We employ a suite of 10 OGCMs to estimate regional anthropogenic CO_2 fluxes from 24 regions. We discuss the features of the flux estimates and their implications for the global CO_2 cycle. We then explore the role of ocean transport in the inversion and assess the uncertainty due to differences among OGCMs. In addition, we quantify

the effect of likely sources of systematic error in the data-based estimates of anthropogenic CO_2 on the inversely estimated fluxes. Finally, the inverse results are compared with forward model simulations using the same suite of models permitting us to assess what we have learned using the inverse approach.

2. Methods

2.1. Anthropogenic CO_2 Estimates

[8] One of the primary components enabling this work is the recent availability of a high-density, global data set of DIC and other tracers in the ocean interior from the Global Ocean Data Analysis Project (GLODAP) [Key *et al.*, 2004]. This data set is composed of data collected from cruises conducted as part of the World Ocean Circulation Experiment (WOCE), the Joint Global Ocean Flux Study (JGOFS), and the National Oceanic and Atmospheric Administration (NOAA) Ocean-Atmosphere Exchange Study (OACES) as well as historical cruises. (Locations of the observations are shown in Figure fs01 of the auxiliary material¹.) As a result of this project, over 68,000 observations are available to constrain the flux estimates.

[9] For each of these observations, the component of the observed DIC concentration that is due to the atmospheric perturbation of CO_2 was estimated using the ΔC^* method [Gruber *et al.*, 1996]. In this study, we use individual data points rather than the gridded data set. The spatial and temporal inhomogeneity of these data are accounted for by sampling the model simulated basis functions at the grid box corresponding to the sampling site during the year the data was collected, as discussed in the following section.

[10] A zonally averaged section of the reconstructed anthropogenic CO_2 used to constrain the inversion is shown in Figure 1. The highest anthropogenic CO_2 concentrations occur near the surface with generally rapidly decreasing concentrations toward the interior of the ocean. This is a consequence of the long timescale of ocean transport from the surface to the deep ocean interior. The deepest penetration occurs in the North Atlantic, owing to the extensive deep water formation in this region, and at midlatitudes, owing to the convergence of intermediate waters and mode waters that were recently in contact with the surface. There is little penetration in the tropics owing to the shallow thermocline. The anthropogenic CO_2 data set is discussed in detail by Sabine *et al.* [2004].

2.2. Inverse Model

[11] We use the same approach used by Gloor *et al.* [2003], with a few adaptations. We provide here only an overview of the method and refer to the auxiliary material for further details. The surface of the ocean is divided into 30 regions, and later aggregated to 24 regions as shown in Figure 2. Ten OGCMs are used to simulate a basis functions for each surface region, describing how an arbitrary unit of flux at the surface impacts tracer concentrations in the interior ocean. (Basis functions for one OGCM

¹Auxiliary material is available at <ftp://ftp.agu.org/apend/gb/2005GB002530>.

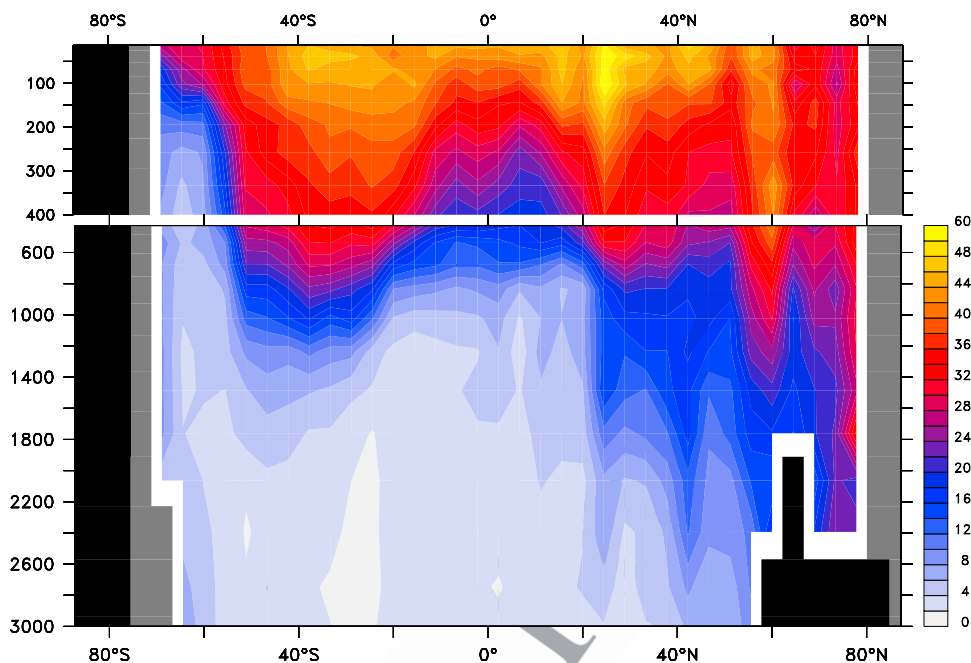


Figure 1. Meridional section of zonally averaged anthropogenic CO_2 ($\mu\text{mol kg}^{-1}$) used to constrain the inversion. Uniform gray areas bounded by a thick, white line represent locations where no observations are available and black areas represent topography. Anthropogenic CO_2 was estimated from dissolved inorganic CO_2 measurements using the ΔC^* method of Gruber *et al.* [1996]. Based on data provided by GLODAP [Key *et al.*, 2004].

176 corresponding to each region are shown in Figure fs02 of
 177 the auxiliary material.) The resulting simulated basis func-
 178 tions are then sampled at the location and time of each of
 179 the observations during the year that each observation was
 180 collected (Figure 1). Each of the observations, in this case
 181 data-based estimates of anthropogenic CO_2 , C_{ant} , is approx-
 182 imated as a linear combination of the $n_{reg} = 30$ basis
 183 functions,

$$C_{ant} = \sum_{i=1, n_{reg}} \lambda_i A_i + \epsilon, \quad (1)$$

185 where A_i is the modeled basis function concentration at the
 186 location of the observations; λ_i is a dimensionless factor that
 187 scales the unit surface flux into the region, and ϵ is a
 188 residual due to limitations of the method. In order to
 189 account for random errors in the data-based anthropogenic
 190 CO_2 estimates, each of the data-based estimates is weighted
 191 by the inverse of its random error, estimated by error
 192 propagation [see Gruber *et al.*, 1996]. Finally, the system of
 193 linear equations is solved for the combination of surface
 194 fluxes that is in optimal agreement with the data-based
 195 anthropogenic CO_2 estimates, using Singular Value decom-
 196 position (SVD). In cases where multiple observations occur
 197 in the same model grid box, each observation is treated as a
 198 separate constraint in the system of linear equations.

199 [12] The basis function for a given model region is
 200 generated by continuously injecting an arbitrary unit flux
 201 of a dye tracer into the surface of a this region and by
 202 running the OGCMs forward in time over the industrial
 203 period (1765–2005). This flux is distributed within the

region on the basis of the seasonal climatology of Takahashi
et al. [2002] and scaled with time on the basis of the
 atmospheric CO_2 perturbation using a scaling factor, $\phi(t)$.

[13] The temporal scaling, ϕ , is calculated from the
 atmospheric CO_2 mixing ratio as done by Gloor *et al.*
 [2003].

$$\phi(t) = \frac{\chi_{\text{CO}_2}(t) - \chi_{\text{CO}_2}^{\text{Preindustrial}}}{\int (\chi_{\text{CO}_2}(t) - \chi_{\text{CO}_2}^{\text{Preindustrial}}) dt}, \quad (2)$$

where χ_{CO_2} is the atmospheric mixing ratio of CO_2 ,
 assumed to be 280 ppm in preindustrial times [Etheridge
et al., 1996]. The time history of χ_{CO_2} is prescribed by a
 spline fit determined by Enting *et al.* [1994] on the basis of
 ice core data [Neftel *et al.*, 1985; Friedli *et al.*, 1986] and
 observations of atmospheric CO_2 at Mauna Loa Observa-
 tory [Keeling *et al.*, 1989]. We updated this time series to
 the year 2005 using observations from Mauna Loa provided
 by CMDL/NOAA and a scaled version of the IS92 scenario
 [Mikaloff Fletcher *et al.*, 2003]. Here $\chi_{\text{CO}_2}^{\text{Preindustrial}}$ is 280 ppm
 based on ice core data.

[14] This temporal scaling of the dye fluxes is possible
 owing to the nearly exponential growth of atmospheric CO_2
 during the industrial period. Theoretical considerations and
 a box model analysis show that when the mixing ratio of an
 atmospheric gas increases exponentially, the oceanic uptake
 is, to first order, proportional to the rate of growth. This is
 because the atmospheric growth rate of a trace gas at any
 point in time is proportional to the total amount of the trace
 gas in the atmosphere. We confirmed our scaling by plotting

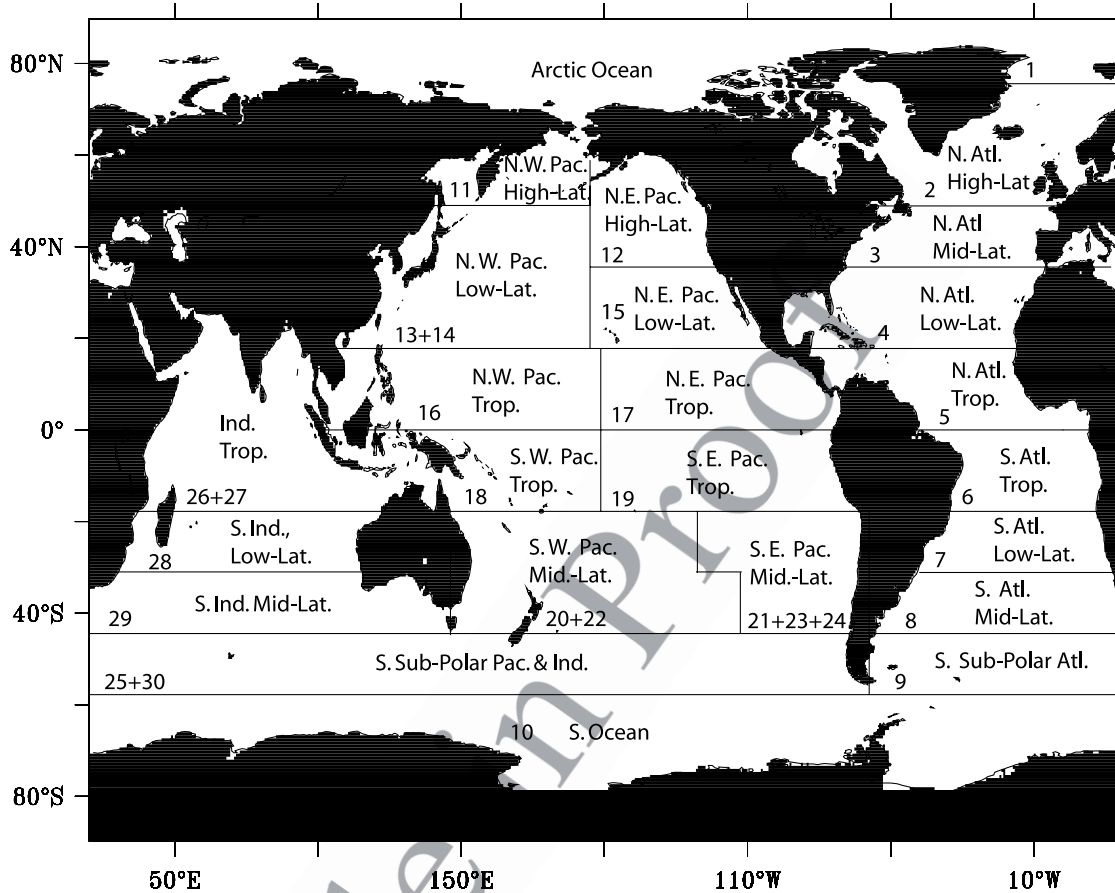


Figure 2. The 24 regions used for the ocean inversion. The region numbers show the aggregation from the original 30 regions [Mikaloff Fletcher *et al.*, 2003] to the 24 regions used in this study.

231 anthropogenic CO₂ uptake versus atmospheric CO₂ perturbation using results from the second phase of the Ocean
 232 Carbon-cycle Model Intercomparison Project (OCMIP-2)
 233 [Watson and Orr, 2003] (see Figure fs03 of the auxiliary
 234 material). This analysis also reveals some notable departures
 235 from our scaling around 1800 and 1940. These are caused
 236 by the large changes in atmospheric CO₂ growth rate that
 237 occurred during these periods. The results from the
 238 OCMIP-2 forward simulations also demonstrate that the
 239 increase in the buffer factor due to the accumulation of
 240 anthropogenic CO₂ in the surface ocean between 1765 and
 241 2005 is too small to have caused a detectable deviation from
 242 our assumed linear scaling.
 243

244 [15] Basis functions were computed for 30 surface regions
 245 [Mikaloff Fletcher *et al.*, 2003], and later aggregated to
 246 24 regions. These aggregations were selected to minimize
 247 the covariance between the modeled response to surface
 248 fluxes into each pair of regions. High covariances between
 249 regions indicate that the inversion cannot effectively distinguish
 250 between two regions either because the basis functions
 251 are too similar or because the observational data set is
 252 insufficient. The sum of the fluxes into two regions with
 253 high covariance may be well constrained, but the individual
 254 fluxes are highly uncertain.

2.3. OGCMs

256
 257 [16] We employ basis functions from 10 OGCMs in order
 258 to elucidate the role of differences in OGCM transport in the
 259 inversion. These model simulations were undertaken by six
 260 different modeling groups: Princeton (PRINCE) Massachusetts
 261 Institute of Technology (MIT), Bern-Switzerland (Bern3D), Jet
 262 Propulsion Laboratory (ECCO), National Center for Atmospheric
 263 Research (NCAR), and University of Liège-Belgium (UL) (described
 264 briefly in the auxiliary material). Princeton provided results from
 265 five different configurations of their model [Gnanadesikan *et al.*,
 266 2002, 2004], summarized in Table ts01 of the auxiliary material.
 267 Owing to the history of model development, several of these
 268 models share common numerical cores. However, comparison with
 269 data constraints have shown that differences in sub-grid-scale
 270 parameterizations and surface forcing are a stronger determinant
 271 of model differences than model architecture [Dutay *et al.*, 2002;
 272 Doney *et al.*, 2004; Matsumoto *et al.*, 2004]. This is well
 273 illustrated by the PRINCE family of models, which share the same
 274 fundamental numerical core setup, but have differing values of
 275 the vertical and along-isopycnal diffusivity, and in some cases
 276 also differing salinity restoring schemes, wind fields, and
 277 topography. These changes cause the resulting model
 278 279

t1.1 **Table 1.** Evaluation of Model Skill Based on Comparisons Between CFC-11 Model Simulations and the GLODAP Gridded CFC Data Set^a

t1.2		Correlation	Normalized Std. Dev. ^b	Model Skill ^c	Inverse Anthropogenic CO ₂ Uptake, Pg C yr ⁻¹	Forward Anthropogenic CO ₂ Uptake, Pg C yr ⁻¹
t1.3	BERN	0.89	1.04	0.81	2.05	N.A.
t1.4	ECCO	0.96	0.89	0.91	2.01	N.A.
t1.5	MIT	0.91	1.00	0.85	2.22	N.A.
t1.6	NCAR	0.95	0.98	0.91	2.18	2.36
t1.7	PRINCE-LL	0.90	1.18	0.80	1.85	1.90
t1.8	PRINCE-HH	0.93	1.05	0.87	2.33	2.43
t1.9	PRINCE-LHS	0.93	1.04	0.86	1.99	2.04
t1.10	PRINCE-2	0.93	1.03	0.87	2.17	2.24
t1.11	PRINCE-2a	0.91	1.05	0.85	2.25	2.35
t1.12	UL	0.87	1.0	0.77	2.81	2.95
t1.13	Mean	0.92	1.02	0.85	2.18	2.32

^aAlso tabulated are forward and inverse estimates of the global total anthropogenic CO₂ uptake (Pg C yr⁻¹, scaled to 1995).

t1.14 Forward results are from OCMIP-2 [Dutay et al., 2002; Watson and Orr, 2003].

^bStd. Dev. is defined as the standard deviation of the modeled field divided by the corresponding standard deviation of the observed field.

t1.15 ^cTaylor [2001].

280 configurations to span nearly the entire range of model
281 behavior seen in the global coarse-resolution models that
282 participated in OCMIP-2 [Matsumoto et al., 2004].

283 [17] Four of the models used here have been compared in
284 OCMIP-2 [Dutay et al., 2002; Doney et al., 2004; Watson
285 and Orr, 2003]: the LL configuration of PRINCE, and the
286 MIT, NCAR, and UL models. The MIT model used here
287 has a slightly different configuration from the version used
288 in OCMIP-2.

289 [18] In order to determine which models are likely to have
290 the most accurate transport on the timescale of anthropo-
291 genic CO₂ perturbation, we compare the GLODAP gridded
292 CFC-11 data set with simulations of CFC-11 from that
293 followed the OCMIP-2 protocol [Dutay et al., 2002]. Table 1
294 shows the correlation between the gridded CFC-11 data
295 and the modeled CFC-11, the standard deviation of the
296 modeled CFC-11 normalized by the standard deviation of
297 the gridded CFC-11 data, and a CFC-11 model skill score
298 based on these two quantities [Taylor, 2001]. We use
299 these CFC-11 skill scores to weight the different models
300 when calculating the between-model means and standard
301 deviations, such that models that simulate the distribution
302 of CFC-11 more accurately have a stronger effect on the
303 reported results.

305 3. Results

306 3.1. Anthropogenic CO₂ Uptake

307 [19] The inversion finds a global anthropogenic CO₂
308 uptake of 2.2 Pg C yr⁻¹, with a weighted standard deviation
309 of 0.25 Pg C yr⁻¹, scaled to a nominal year of 1995. The
310 range across all models is 1.85 to 2.81 Pg C yr⁻¹ (Table 1).
311 This substantial range is due in part to differences between
312 the effective vertical diffusivities in the models. Highly
313 diffusive models distribute the dye over a larger portion
314 of the ocean. This requires larger anthropogenic CO₂ fluxes
315 in order to match the high observed anthropogenic CO₂
316 concentrations in the upper ocean. The OGCMs providing
317 the high and low ends of this range (UL and PRINCE-LL)
318 also have lower CFC-11 skill scores than the other OGCMs

used in this study. This suggests that the cross-model range 319
can be considered an upper estimate of the uncertainty 320
associated with the inversely estimated global anthro- 321
pogenic CO₂ uptake. 322

[20] The greatest anthropogenic CO₂ uptake occurs in the 323
Southern Ocean, particularly in the subpolar regions (44°S 324
to 58°S), where the weighted mean anthropogenic CO₂ 325
uptake is 0.51 Pg C yr⁻¹ with a standard deviation of 326
0.17 Pg C yr⁻¹ (Figure 3). This flux represents 23% of the 327
global total anthropogenic CO₂ uptake. In addition, the 328
inversion finds considerable anthropogenic CO₂ uptake in 329
the tropics. In contrast, anthropogenic CO₂ uptake at mid 330
latitudes is found to be low, despite the fact that the greatest 331
anthropogenic CO₂ storage occurs there (Figure 1). 332

[21] These broad features in the spatial pattern of the 333
fluxes are consistent across all of the models that partici- 334
pated in this study. However, there exists considerable 335
model differences between the anthropogenic flux estimates 336
for some regions, leading to substantial uncertainties in the 337
weighted means. The greatest anthropogenic CO₂ uncer- 338
tainty occurs in the Southern Ocean, with a weighted 339
standard deviation from the weighted mean uptake of 340
0.10 Pg C yr⁻¹ for the region south of 58°S and 0.17 Pg 341
C yr⁻¹ for the region between 44°S and 58°S. As a 342
percentage of the total signal, the range in the high-latitude 343
North Atlantic is also very high. The inverse estimates are 344
the most consistent in the North Atlantic and North Pacific. 345

[22] This uptake pattern is in good agreement with pre- 346
vious forward modeling studies. In some of the first 3-D 347
OGCM studies of the oceanic uptake of anthropogenic CO₂, 348
Sarmiento et al. [1992] and Maier-Reimer and Hasselmann 349
[1987] found a similar pattern of vigorous anthropogenic 350
CO₂ uptake at high latitudes and at the equator, and low 351
anthropogenic CO₂ uptake at midlatitudes. They attributed 352
the high uptake in the tropics and in the high latitudes 353
primarily to these regions being characterized by high rates 354
of transport and mixing of subsurface waters depleted in 355
anthropogenic CO₂ to the surface. Although variations in 356
gas transfer velocity were found by Sarmiento et al. [1992] 357
to be of second importance for the global uptake of 358

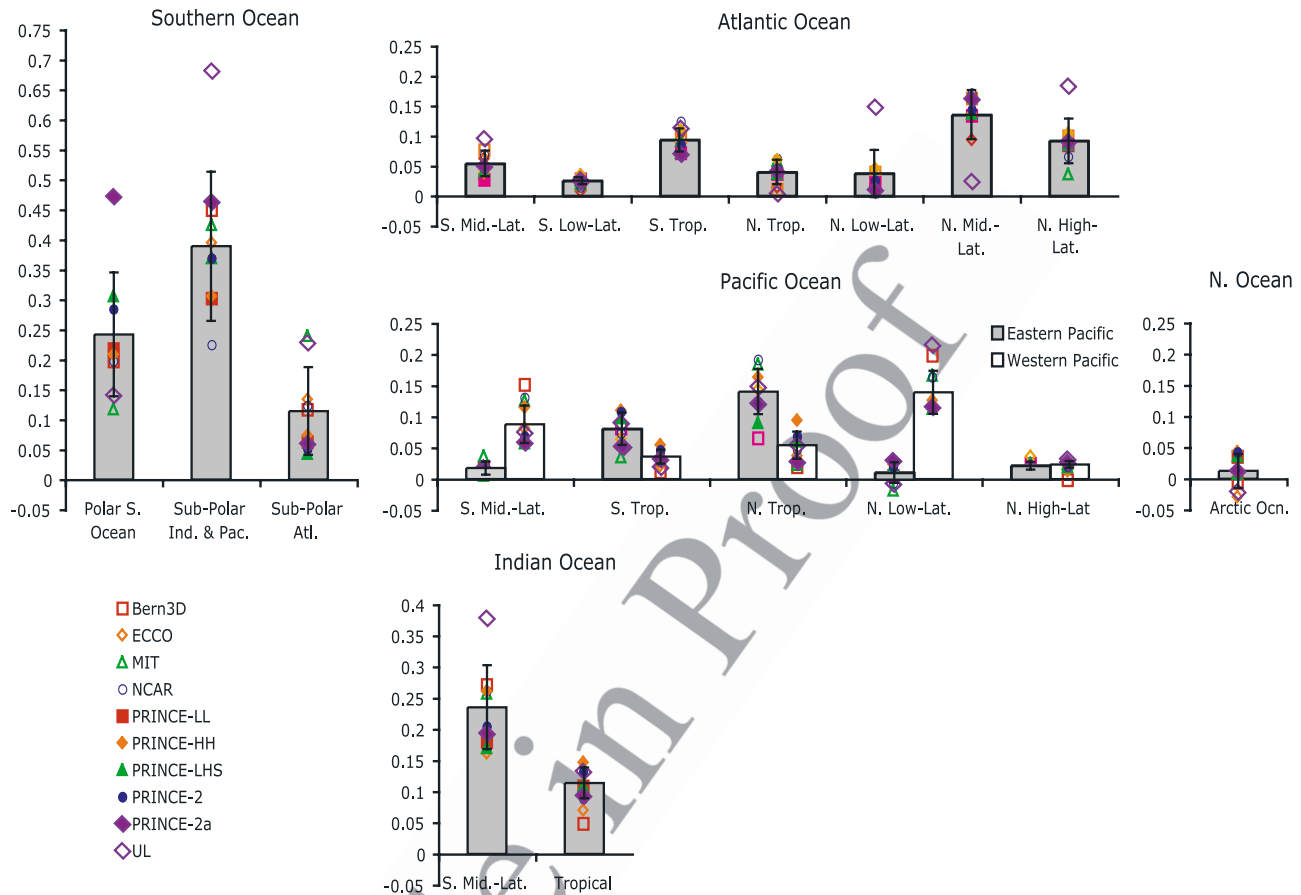


Figure 3. Inverse estimates of anthropogenic CO_2 uptake by the ocean (Pg C yr^{-1}) for a nominal year of 1995 (positive values indicate flux into the ocean). The columns show the cross-model weighted means, and the error bars represent the weighted standard deviation. The weights were provided by the model's CFC-11 skill scores (see Table 1). The flux estimates for individual models are shown as symbols.

359 anthropogenic CO_2 , the higher wind speeds in high-latitude
 360 regions were found to have some enhancing effect on
 361 greater anthropogenic CO_2 uptake there. Owing to the long
 362 residence of upper ocean waters in the midlatitudes, anthro-
 363 pogenic CO_2 in the surface waters of these regions generally
 364 follows the atmospheric perturbation quite closely [see, e.g.,
 365 Gruber *et al.*, 2002; Keeling *et al.*, 2004; Takahashi, 2004].
 366 This leads to low uptake.

367 [23] Sarmiento *et al.* [1992] found an anthropogenic CO_2
 368 uptake of 1.9 Pg C yr^{-1} for the decade from 1980 to 1989.
 369 This is comparable with our weighted estimate of $1.82 \pm$
 370 $0.21 \text{ Pg C yr}^{-1}$ when scaled to the same time period. Orr *et al.*
 371 [2001] simulated anthropogenic CO_2 uptake using four
 372 3-D OGCMs and found a 1980–1989 uptake of $1.85 \pm$
 373 $0.35 \text{ Pg C yr}^{-1}$. Like this study, they found the greatest
 374 anthropogenic CO_2 uptake and the greatest range between
 375 models in the Southern Ocean. The inverse estimates will be
 376 compared in more detail with the forward simulations in
 377 section 5.

378 [24] This study is also in good agreement with the earlier
 379 inversion study of Gloor *et al.* [2003] (Figure fs05 of the

auxiliary material), as the latter estimates generally fall 380
 within the model range of this study. Since the methodology 381
 is the same, the primary causes for the differences between 382
 the two studies are the choice of OGCM and the selection of 383
 model regions. Gloor *et al.* [2003] used only one model 384
 (PRINCE-LL, also used here), while we report the weighted 385
 mean of 10 models, including the PRINCE-LL model. We 386
 estimate fluxes into 24 surface regions while Gloor *et al.* 387
 [2003] used only 13 regions. The larger number of model 388
 regions in this study is expected to reduce the aggregation 389
 error [Kaminski *et al.*, 2001], giving our results more 390
 confidence. In addition, we employ a spatial and temporal 391
 flux pattern modeled after the observationally based air-sea 392
 CO_2 flux estimates of Takahashi *et al.* [2002], which is 393
 likely a better assumption than the annual mean pattern 394
 based on heat fluxes employed by Gloor *et al.* [2003]. 395
 Additional but likely smaller differences between the two 396
 studies arise because we weight the data-based anthropo- 397
 genic CO_2 estimates with an estimate of the random error, 398
 which is different for every observation, while Gloor *et al.* 399
 [2003] weighted all of the observations equally. Finally, a 400

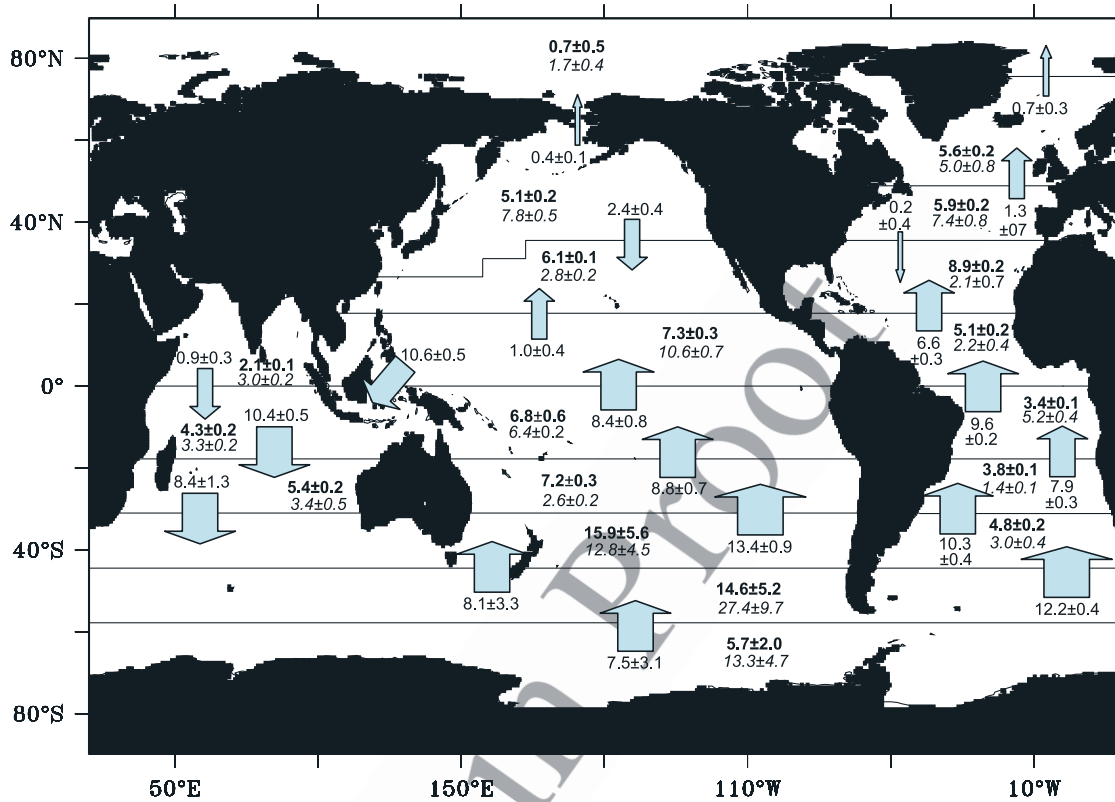


Figure 4. Global map of the time integrated (1765–1995) transport (shown above or below arrows) of anthropogenic CO₂ based on the inverse flux estimates (italics) and their implied storage (bold) in Pg C. Shown are the weighted mean estimates and their weighted standard deviation.

401 larger anthropogenic CO₂ data set is available to constrain
 402 the inverse estimates in this study. Owing to the large
 403 number of observations used in both studies, this latter
 404 difference has little impact on the inverse estimates.

406 3.2. Oceanic Transport of Anthropogenic CO₂

407 [25] The transport of anthropogenic CO₂ can be calcu-
 408 lated from the divergence of the regional fluxes integrated in
 409 time (1765–1995) and the inverse storage estimates. In
 410 order to be consistent with the estimated fluxes, we calcu-
 411 lated this storage from the sum of the regional scaling
 412 factors multiplied by the basis functions (equation (1)),
 413 rather than using observed storage.

414 [26] Globally, the vigorous anthropogenic CO₂ uptake in
 415 the Southern Ocean and the absence of large storage there
 416 drive a substantial equatorward transport in most of the
 417 Southern Hemisphere (Figure 4). Only about half of the
 418 anthropogenic CO₂ taken up in the high-latitude Southern
 419 Ocean is stored there, while the rest is transported equator-
 420 ward. This leads to a considerable anthropogenic CO₂
 421 storage at midlatitudes in the Southern Hemisphere and a
 422 northward cross-equatorial transport. In the Northern Hemi-
 423 sphere, anthropogenic CO₂ is transported poleward from the
 424 tropics and equatorward from midlatitudes, leading to
 425 convergence and storage in the subtropics. We find a small
 426 amount of poleward transport from high latitudes into the
 427 Arctic Ocean. This general pattern of anthropogenic uptake
 428 at high latitudes and in the tropics with subsequent transport

to midlatitudes, where the anthropogenic CO₂ is stored, is in
 good agreement with previous modeling studies [Sarmiento
 et al., 1992].

[27] The largest portion of the anthropogenic CO₂ trans-
 ported equatorward from the Southern Ocean is going into
 the Atlantic Ocean. Some of it is transported northward
 along the surface, and some of it is transported at depth,
 mostly associated with the equatorward and downward
 spreading of Sub-Antarctic Mode Water (SAMW) and
 Antarctic Intermediate Water (AAIW). The bulk of this
 Southern Ocean derived anthropogenic CO₂ then accumu-
 lates in the South Atlantic Subtropical Gyre (basis functions
 shown in Figures fs06 and fs07 of the auxiliary material). A
 portion of the anthropogenic CO₂ taken up in the tropics is
 transported southward, but most is either stored there or
 transported northward along the surface and then stored in
 the subtropical North Atlantic (Figure fs02 of the auxiliary
 material, regions 5 and 6).

[28] In the North Atlantic, the greatest anthropogenic CO₂
 uptake occurs at mid and high latitudes. Anthropogenic CO₂
 taken up in these regions is either transported equatorward
 to midlatitudes or poleward, where it is entrained into North
 Atlantic Deep Water (NADW) (Figure fs02 of the auxiliary
 material, regions 2 and 3). This leads to convergence and
 storage in the Northern Subtropics (Figure 4).

[29] About 40% of the anthropogenic CO₂ transported
 poleward from the Southern Ocean is going into the Pacific
 Ocean or into the Indian Oceans (Figure fs02 of the

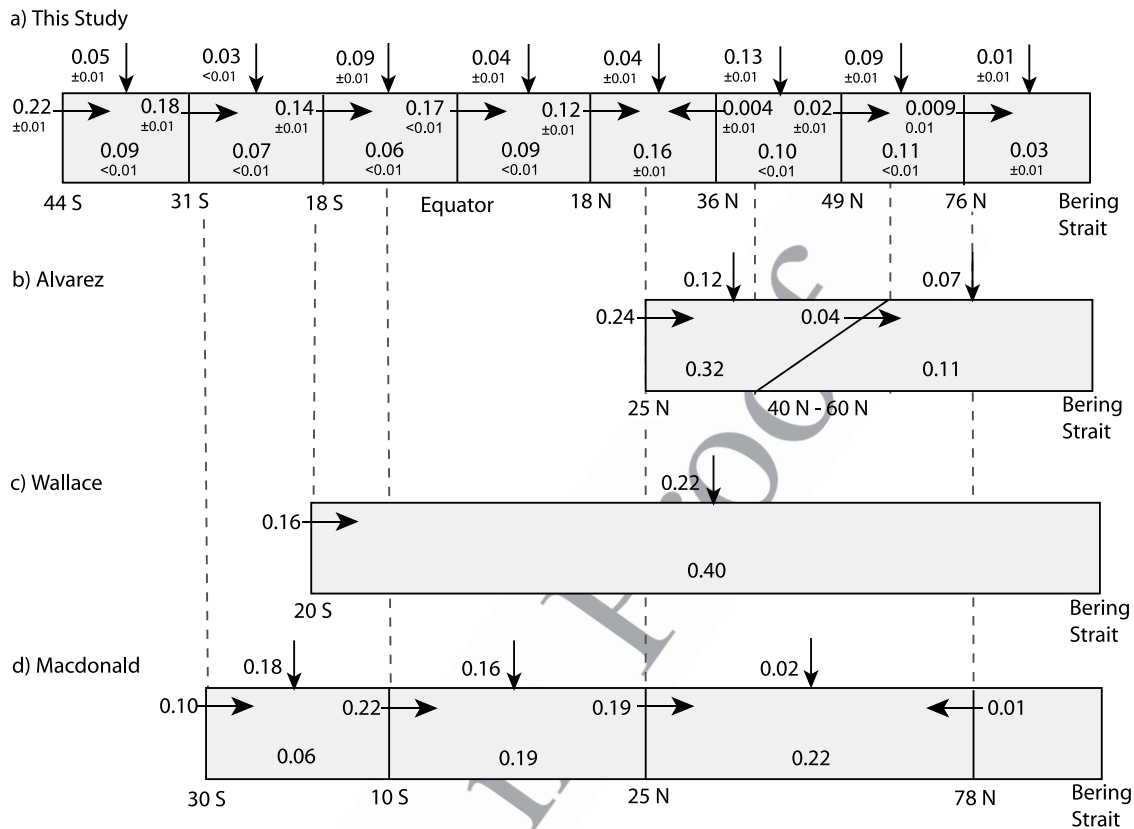


Figure 5. Uptake, storage, and transport of anthropogenic CO_2 in the Atlantic Ocean (Pg C yr^{-1}) based on (a) this study (weighted mean and standard deviation scaled to 1995), (b) the estimates of [Alvarez *et al.*, 2003], where the transport across 24°N was taken from Rosón *et al.* [2003], (c) Wallace [2001], where the transport across 20°S was taken from Holfort *et al.* [1998], and (d) Macdonald *et al.* [2003], where the transports across 10°S and 30°S were taken from Holfort *et al.* [1998], and the transport across 78°N was taken from Lundberg and Haugan [1996]. This figure is not to scale.

457 auxiliary material, regions 25 and 30). Since this transport
 458 exceeds storage in the South Pacific, it drives equatorward
 459 transport of anthropogenic CO_2 throughout the South Pa-
 460 cific and substantial northward cross-equatorial transport
 461 (Figure 4). In the North Pacific, the greatest anthropogenic
 462 CO_2 uptake occurs at high latitudes and in the tropics.
 463 Anthropogenic CO_2 taken up in the North Pacific is trans-
 464 ported equatorward (Figure fs02 of the auxiliary material,
 465 regions 11 and 12), and anthropogenic CO_2 from the tropics
 466 is transported poleward (Figure fs02 of the auxiliary mate-
 467 rial, regions 16 and 17), leading to convergence and storage
 468 in the subtropical North Pacific.

469 [30] The Indonesian throughflow plays a critical role in
 470 determining the transports in the Indian and Pacific oceans
 471 south of 18°N (Figure 4) as it sets up a transport loop that
 472 involves strong northward transport in the South Pacific and
 473 southward transport in the southern Indian Ocean. We
 474 computed the anthropogenic CO_2 transport by the Indone-
 475 sian throughflow for each model by multiplying at each
 476 model depth the diagnosed volume flux in the model with
 477 the anthropogenic CO_2 concentration estimate from the
 478 GLODAP gridded data set, interpolated to the throughflow
 479 point in each model. The OGCM simulated volume fluxes
 480 across the straight are generally within the range of obser-

481 vational estimates [e.g., Gordon and Fine, 1996], but these
 482 estimates are themselves rather uncertain since this transport
 483 is not well understood and may have significant interannual
 484 variability. We therefore regard our estimated time-integrated
 485 transport of $10.6 \pm 0.5 \text{ Pg C}$ by the Indonesian throughflow as
 486 an uncertain component of our transport estimates.

487 [31] In Figure 5, we compare our transport estimates for
 488 the Atlantic with those estimated from hydrographic data
 489 and data-based anthropogenic CO_2 estimates [e.g.,
 490 Lundberg and Haugan, 1996; Holfort *et al.*, 1998; Alvarez
 491 *et al.*, 2003; Rosón *et al.*, 2003; Macdonald *et al.*, 2003].
 492 This comparison remains somewhat qualitative, as these
 493 hydrographic estimates are subject to substantial uncertain-
 494 ties from a variety of factors [e.g., Macdonald *et al.*, 2003].
 495 In addition, the hydrographic data-based estimates deter-
 496 mine the transport at a single point in time and could be
 497 substantially biased owing to the neglect of seasonal varia-
 498 tions in transport [e.g., Wilkin *et al.*, 1995]. In contrast, our
 499 estimate of the anthropogenic CO_2 transport is scaled from
 500 the time-integrated transport from 1765 to 1995, and reflects
 501 a long-term mean transport. Therefore, even if the hydro-
 502 graphic data-based estimates were insensitive to seasonal
 503 biases, the two transports are not directly comparable as
 504 they pertain to very different time periods. In addition, there

are sources of uncertainty associated with the inverse estimates that have not been quantified, as discussed in section 4. These caveats need to be considered when comparing the results.

[32] In order to arrive at transport estimates for a particular year, we scaled the time integrated transports to 1995 using the atmospheric perturbation. We assumed inventory in each region increases proportionally with the perturbation to atmospheric CO₂, such that the regional transports scale proportionally. This scaling is supported by an analysis of forward model simulations (Figure fs08 of the auxiliary material).

[33] Both our estimates and hydrographic transects find substantial northward transport throughout the South Atlantic (Figure 5). Our transport estimate across 31°S is 70% larger than the estimate of 0.1 ± 0.02 Pg C yr⁻¹ across 30°S determined by *Holfort et al.* [1998]. However, our estimate of 0.14 ± 0.01 Pg C yr⁻¹ northward transport across 18°S is in reasonable agreement with *Wallace* [2001], who found 0.16 ± 0.02 Pg C yr⁻¹ northward transport across 20°S.

[34] In the North Atlantic, we find a northward transport of 0.12 ± 0.01 Pg C yr⁻¹ across 18°N and no significant transport across 36°N. This is substantially smaller than the northward transport of 0.24 ± 0.08 Pg C yr⁻¹ and 0.19 ± 0.08 Pg C yr⁻¹ across 25°N estimated by *Rosón et al.* [2003] and *Macdonald et al.* [2003], respectively. However, owing to the large uncertainties associated with the hydrographic estimates, the differences are only marginally statistically significant. We find a small northward transport across 49°N of 0.02 ± 0.01 Pg C yr⁻¹ that is in good agreement with the transport estimated across a diagonal transect between 40°N and 60°N [*Álvarez et al.*, 2003]. Finally, we find a marginally significant northward transport at 76°N, whereas *Lundberg and Haugan* [1996] estimated a southward transport at 78°N. The small northward transport across 76°N is very sensitive to the choice of OGCM, as will be shown in the following section. Therefore we conclude that our northward transport at 76°N is not a robust result of the inversion, while the transports at the more southern latitudes in the Atlantic are found to be generally invariant across the models investigated.

4. Sensitivity and Error Analysis

[35] In this section, we address and quantify two sources of error in the inversion. First, we use basis functions from 10 OGCMs to assess the sensitivity of the estimates to the choice of transport model. Then we address the sensitivity of the inversion to biases in the data-based estimates of the anthropogenic CO₂ concentrations.

[36] There are other potential sources of error that will not be addressed here. The most important is our assumption that the ocean circulation has remained constant over time. There is substantial evidence for decadal variability in ocean circulation from repeat hydrography studies [e.g., *García et al.*, 2002; *Bryden et al.*, 2003; *Johnson and Gruber*, 2006; *McPhaden and Zhang*, 2002], which could lead to biases in the inverse estimates. For example, if the ventilation in a given region were weakening progressively over time, a basis function generated for that region using constant

present-day circulation would underestimate the fraction of dye near the surface relative to the portion of dye in deeper waters. We are currently unable to quantitatively assess the possible impact of long-term changes in ocean circulation on our inverse results. Forward simulations by *Raynaud et al.* [2005] suggest that variations in ocean circulation have a relatively small impact on the air-sea flux of anthropogenic CO₂ on interannual timescales, but may be more substantial on decadal timescales. However, comparisons between simulations of CFCs with constant circulation and observations do not indicate major problems as a result of decadal variability [*Dutay et al.*, 2002].

[37] There are also potential methodological sources of errors. For example, the relatively small number of model regions used here may cause aggregation errors [*Kaminski et al.*, 2001]. However, on the basis of the analysis of the covariance matrix (see text01 section of the auxiliary material), we conclude that a larger number of model regions is likely to yield a solution that is not adequately constrained by the observations. A second issue is the spatial and temporal pattern used to prescribe the distribution of the fluxes within the model region. Inverse estimates using several different spatial patterns indicate that the flux estimates are not particularly sensitive to the choice of spatial pattern or whether the pattern includes seasonal variations [*Gloor et al.*, 2001].

4.1. Sensitivity to the Choice of OGCM

[38] On the basis of a comparison of the 10 OGCMs considered in this study, we find that most of the major features of the spatial pattern of the anthropogenic CO₂ uptake and transport estimates are generally robust. However, there are substantial between-model differences in some regions.

[39] The largest variability differences between models occurs in the Southern Ocean (see Figures fs03, fs06, and fs09 in the auxiliary material) as found by OCMIP-2 [*Orr et al.*, 2001; *Watson and Orr*, 2003; *Doney et al.*, 2004]. *Doney et al.* [2004] cite limitations of the models in accurately representing along-isopycnal transport, brine rejection due to sea ice formation, boundary conditions, the role of eddies and how they are parameterized, and the lack of data available to validate the models in this region as the major reasons for this large spread in model behavior. In our inversion, the UL and MIT models give the largest anthropogenic CO₂ uptake and storage in the Southern Ocean. The UL model has the poorest CFC skill score, but the MIT skill score is close to the average of all models used here. These two models entrain a larger portion of the anthropogenic CO₂ injected between 44°S and 58°S into deep waters and transport a smaller portion to the midlatitudes than all of the other models (see, for example, basis functions for the subpolar Atlantic in Figures fs06 and fs07 of the auxiliary material). The midlatitude basis functions have relatively shallow dye penetration. Therefore a greater anthropogenic CO₂ uptake is required at high latitudes to match the observed storage in midlatitude intermediate waters.

[40] The Arctic Ocean is the second region showing high between-model differences in the estimated fluxes. This is

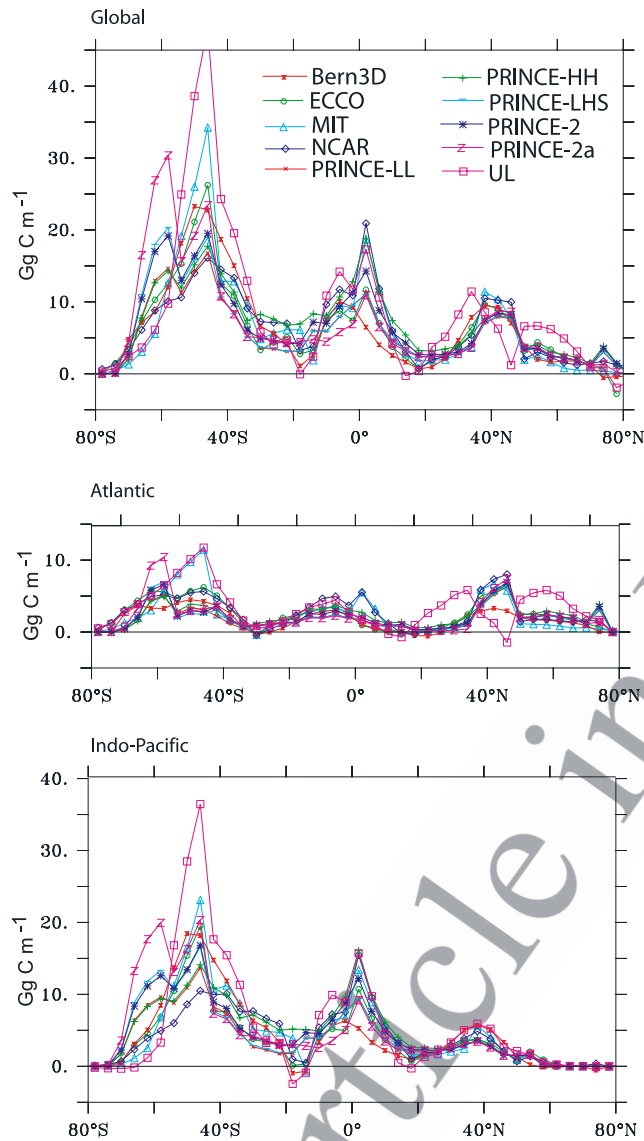


Figure 6. Zonally and temporally integrated anthropogenic CO₂ uptake by (top) the global ocean, (middle) the Atlantic Ocean, and (bottom) the Indo-Pacific Ocean from 1765–1995.

623 likely due to the large differences between models in the
 624 representation of this basin. Some models have a well-
 625 resolved Arctic basin due to their shifting the North Pole
 626 over land; others do not resolve it at all. We therefore have
 627 little confidence in the estimated fluxes for this basin.
 628 Fortunately, this has little influence on our results, as the
 629 fluxes are expected to be small owing to sea-ice inhibiting
 630 the uptake for a large portion of the Arctic.

631 [41] The inverse estimates for most of the OGCMs used
 632 in this study are in excellent agreement in the Atlantic.
 633 However, the UL model exhibits a different latitudinal
 634 distribution of anthropogenic CO₂ uptake (Figure 6). This
 635 can be traced back to this model storing a large portion of
 636 the dye tracer injected into the high-latitude North Atlantic
 637 near the surface. As discussed further in the auxiliary

material, this leads to a rearrangement of the flux distribu- 638
 tion in order to match the data-based estimates of anthro- 639
 pogenic CO₂. 640

[42] One way to evaluate the different models and to 641
 assess biases in the inverse estimates is to examine the 642
 residuals between the data-based anthropogenic CO₂ esti- 643
 mates and the anthropogenic CO₂ storage calculated from 644
 the inverse flux estimates (Figure 7). The models underes- 645
 timate the mean anthropogenic CO₂ concentration by about 646
 1 to 2.5 $\mu\text{mol kg}^{-1}$. In addition, all of the models underes- 647
 timate anthropogenic CO₂ storage in the thermocline (500 648
 to 1000 m). This suggests that they either do not sufficiently 649
 ventilate this region, or that the anthropogenic CO₂ esti- 650
 mates in this region are biased high. As discussed in more 651
 detail in the following section, this region has not been 652
 identified as a region of substantial possible biases in the 653
 data-based estimates of anthropogenic CO₂ [Matsumoto and 654
Gruber, 2005], so that an overly weak ventilation in the 655
 models is the more likely cause of the positive residuals in 656
 the deeper thermocline. 657

[43] In waters shallower than 500 m, most of the models 658
 show negative residuals at around 20°N and 30°S and 659
 positive residuals in the tropics and at around 40°N and 660
 40°S. One possible explanation for this structure is that the 661
 models have excessive poleward transport out of the tropics 662
 and too strong equatorward transport out of the high 663
 latitudes. If this were the case, the uptake might be over- 664
 estimated in the tropics and at high latitudes in order to 665
 match the substantial anthropogenic CO₂ concentrations in 666
 these areas. A large portion of this excessive flux would 667
 then be transported to midlatitudes, leading to an overesti- 668
 mate of the anthropogenic CO₂ storage. An alternative 669
 explanation is a bias in the reconstructed anthropogenic 670
 CO₂ concentrations. *Matsumoto and Gruber [2005]* showed 671
 that the ΔC^* method tends to be biased high in the upper 672
 thermocline, explaining at least part of the positive residuals 673
 in this region. 674

[44] In the Southern Ocean, most of the models have 675
 negative residuals between about 200 m and 1000 m and 676
 positive residuals in the deep waters. If the data-based 677
 estimates of anthropogenic CO₂ were correct, this would 678
 suggest that the models tend to overestimate the vertical 679
 transport of anthropogenic CO₂ in the upper 1000 m of the 680
 Southern Ocean and that they are unable to represent the 681
 small anthropogenic CO₂ concentrations found in the deep 682
 Southern Ocean. Since the identified possible biases in the 683
 data-based estimates of anthropogenic CO₂ are an overes- 684
 timation in the upper ocean and an underestimation in the 685
 deep ocean [Matsumoto and Gruber, 2005], the adjustment 686
 for this possible error in anthropogenic CO₂ would actually 687
 accentuate the residuals rather than ameliorate them. This 688
 points to a persistent problem in the employed OGCMs in 689
 how they simulate the circulation in the Southern Ocean. 690
 The UL and PRINCE-LL models, which have the lowest 691
 CFC-11 skill scores (Table 1), represent the two extreme 692
 cases. The UL model, which finds substantially more 693
 anthropogenic CO₂ uptake than any of the other contrib- 694
 uting models, has large negative residuals throughout most of 695
 the Southern Ocean, suggesting that its inversely estimated 696
 uptake is too large. In contrast, the PRINCE-LL model, 697

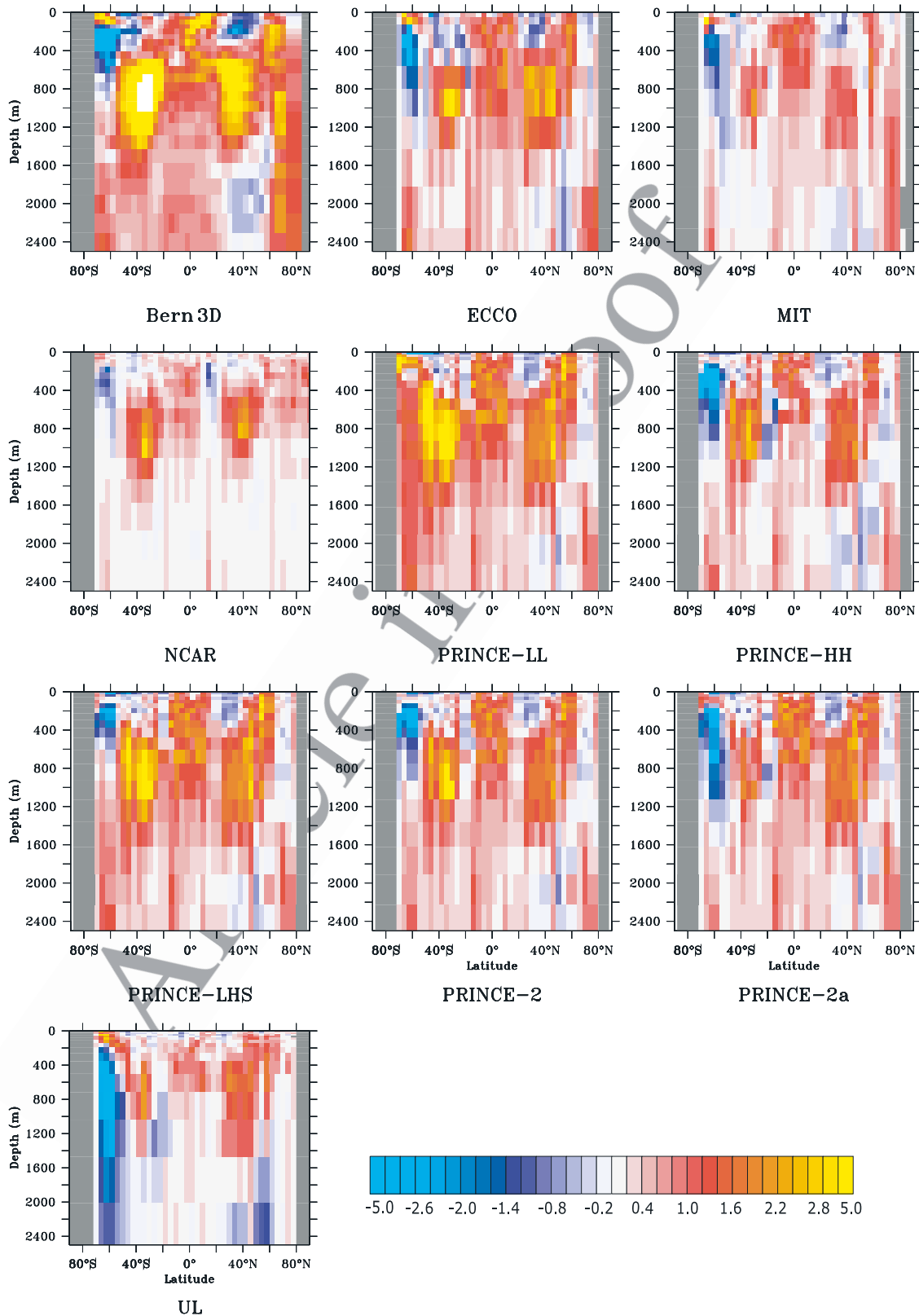


Figure 7. Meridional section of the zonal mean of the difference between the data-based anthropogenic CO₂ estimates and the inverse anthropogenic CO₂ storage estimates ($\mu\text{mol kg}^{-1}$) for the 10 models that participated in this study. Solid gray areas represent locations where no observations are available or that are outside the model grid. Little spatial structure to the residuals exists below 2500 m.

698 which has the lowest global anthropogenic CO₂ uptake, has
 699 positive residuals throughout the Southern Ocean. This
 700 model is characterized by very low vertical and along-
 701 isopycnal diffusivity, so that a much greater portion of the
 702 anthropogenic CO₂ remains near the surface. The resulting
 703 underestimation of the data-based estimates points to this
 704 model being deficient in its uptake. Thus the large residuals
 705 exhibited by these two models confirm their having a low
 706 CFC-11 skill score. Their large residuals also confirm our
 707 use of these skill scores as weights for computing means
 708 and standard deviations, as the likelihood of these two
 709 models being accurate is smaller than that of the other
 710 models.

712 4.2. Sensitivity to Errors in the Anthropogenic CO₂ 713 Estimates

714 [45] Inverse estimates rest on the assumption that the
 715 observations used to constrain the inversion are accurate.
 716 However, we constrain our inversion with an estimated
 717 quantity, which may contain biases. This makes it necessary
 718 to test the sensitivity of the inverse flux estimates to such
 719 biases. First, we examine the impact of a density-dependent
 720 bias modeled after that identified by *Matsumoto and Gruber*
 721 [2005]. Then we investigate the effect of biases in the
 722 stoichiometric ratios used to remove the effects of biology
 723 from the observed CO₂ concentration. *Gruber* [1998] ar-
 724 gued that biases in this ratio could substantially alter the
 725 distribution of anthropogenic CO₂ as well as the total
 726 inventory.

727 [46] We will not investigate the impact of possible biases
 728 in anthropogenic CO₂ emerging from the fact that possible
 729 changes in ocean circulation due to ocean warming were not
 730 taken into account in estimating the anthropogenic CO₂
 731 inventory [*Keeling*, 2005]. *Matsumoto and Gruber* [2005]
 732 showed, however, that changes in ocean circulation and
 733 biogeochemistry have relatively little impact on the esti-
 734 mated anthropogenic CO₂ concentrations [see also *Sabine*
 735 and *Gruber*, 2005].

736 [47] *Matsumoto and Gruber* [2005] examined the accu-
 737 racy of the anthropogenic CO₂ estimates by applying the
 738 ΔC^* method to results from a forward model simulation
 739 with known anthropogenic CO₂ concentrations. The authors
 740 identified substantial biases in the ΔC^* method stemming
 741 from the neglected time evolution of the air-sea disequilib-
 742 rium, biases in the *p*CFC ventilation age, and errors in
 743 identifying water masses that contribute to a given water
 744 parcel. As a result, they suggested that the ΔC^* method
 745 tends to overestimate the anthropogenic CO₂ inventory in
 746 shallower waters by about 10% and underestimate it in
 747 deeper waters. Globally, the ΔC^* method inferred anthro-
 748 pogenic CO₂ inventory was about 7% larger than the true
 749 inventory.

750 [48] It is not within the scope of this paper to reassess the
 751 anthropogenic CO₂ data set based on the findings of
 752 *Matsumoto and Gruber* [2005]. However, it is critical to
 753 address the impact these biases might have on the inverse
 754 estimates. To this end, we constructed a “*Matsumoto and*
 755 *Gruber corrected*” scenario, in which a hypothetical correc-
 756 tion factor was applied to the data-based anthropogenic CO₂
 757 estimates and these corrected anthropogenic CO₂ estimates

were used in the inversion. The correction factor was 758
 determined as a function of density in such a way that it 759
 reduced anthropogenic CO₂ in the upper ocean by about 760
 10% and increased it in the deep ocean slightly, while 761
 reducing the global inventory by 7%. In addition, two 762
 scenarios were constructed to assess the impact of a globally 763
 uniform shift in the oxygen to carbon remineralization ratio, 764
 $r_{C:O_2}$, used to remove the effects of biology. The construc- 765
 tion of these scenarios is described in section 4 of the text01 766
 file in the auxiliary material. 767

[49] The spatial pattern of the inversely estimated air-sea 768
 fluxes is remarkably insensitive to these biases (Figure 8); 769
 however, the net global anthropogenic CO₂ uptake scales 770
 approximately linearly with changes in the estimated global 771
 inventory of anthropogenic CO₂ (8, numerical results 772
 shown in Table ts04 of the auxiliary material). The Matsu- 773
 moto and Gruber scenario leads to a global reduction in the 774
 anthropogenic CO₂ uptake of 8%, reflecting the global 7% 775
 decrease in the anthropogenic CO₂ inventory. Relative to 776
 the global uptake, the anthropogenic uptake at high latitudes 777
 (north of 49°N and south of 58°S) is increased slightly and 778
 the uptake in all other regions is decreased slightly by the 779
 hypothetical correction. Increasing the stoichiometric ratio, 780
 $r_{C:O_2}$, by 13% decreases the global anthropogenic CO₂ flux 781
 by 7%, and decreasing $r_{C:O_2}$ by 13% increases the global 782
 flux by 8%. In the ΔC^* method, $r_{C:O_2}$ together with AOU is 783
 used to subtract the effect of changes in DIC as a result of 784
 biological processes. Therefore increases in $r_{C:O_2}$ are 785
 expected to lead to decreases in the estimated anthropogenic 786
 CO₂, and vice versa. The inverse estimates are least sensi- 787
 tive to changes in $r_{C:O_2}$ at midlatitudes, where AOU is 788
 lowest, and most sensitive at high latitudes and in the 789
 tropical Pacific. 790

[50] We conclude from these analyses that the inverse flux 791
 estimates generally tend to be more sensitive to the choice 792
 of model than to biases in the anthropogenic CO₂ estimates. 793
 Therefore, despite the fact we employed 10 different 794
 OGCMs and used CFC skill scores to weight the different 795
 models, possible biases in model transport still tends to 796
 dominate the overall uncertainty in our flux estimates. 797

799 5. Comparison of Forward and Inverse Models

[51] Traditionally, the spatial distribution of the air-sea 800
 flux of anthropogenic CO₂ has been estimated using for- 801
 ward simulations of OGCMs forced by the observed atmo- 802
 spheric CO₂ perturbation [e.g., *Orr et al.*, 2001; *Murnane et* 803
al., 1999; *Sarmiento et al.*, 1992]. In this section, the inverse 804
 estimates of each OGCM are compared with their 805
 corresponding forward estimates undertaken as part of 806
 OCMIP-2 [*Watson and Orr*, 2003] in order to assess what 807
 we have learned by constraining the models with the data- 808
 based anthropogenic CO₂ estimates. 809

[52] The difference between the forward simulations and 810
 the corresponding inverse estimates of anthropogenic CO₂ 811
 for 1995 from seven of the ten models used in this study are 812
 shown in Figure 9 (complete numerical results shown in 813
 Table ts05 of the auxiliary material). Positive values indicate 814
 that the forward model simulates more anthropogenic CO₂ 815
 uptake than the inversion and vice versa. The Bern3D, 816

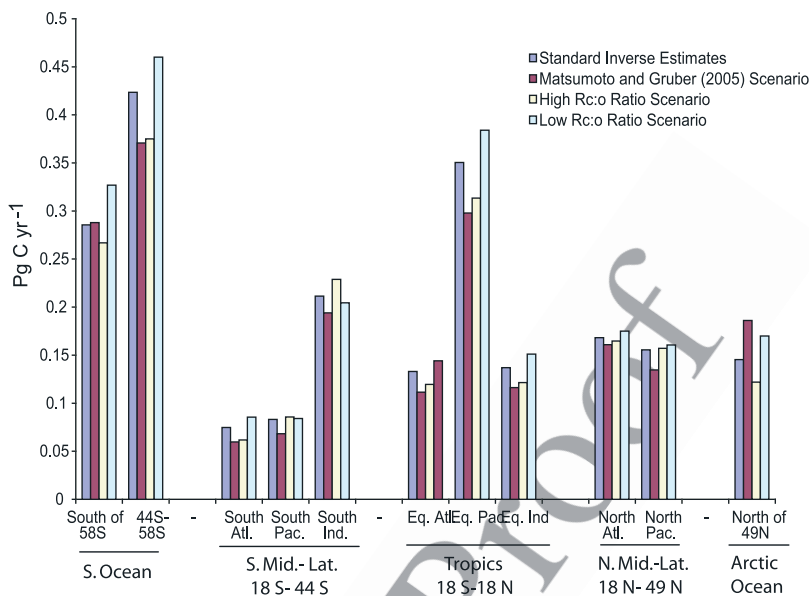


Figure 8. Sensitivity of the inverse estimates of the anthropogenic CO₂ fluxes (Pg C yr⁻¹, scaled to 1995) to errors in the data-based anthropogenic CO₂ estimates used to constrain the inversion. The anthropogenic CO₂ fluxes have been estimated using the standard data-based anthropogenic CO₂ estimates from GLODAP, anthropogenic CO₂ estimates with a hypothetical correction based on work by *Matsumoto and Gruber* [2005], and anthropogenic estimates based on the high and low end of the range associated with the CO₂ to oxygen ratio [*Anderson and Sarmiento, 1994*]. The inverse estimates are aggregated to 11 regions for clarity.

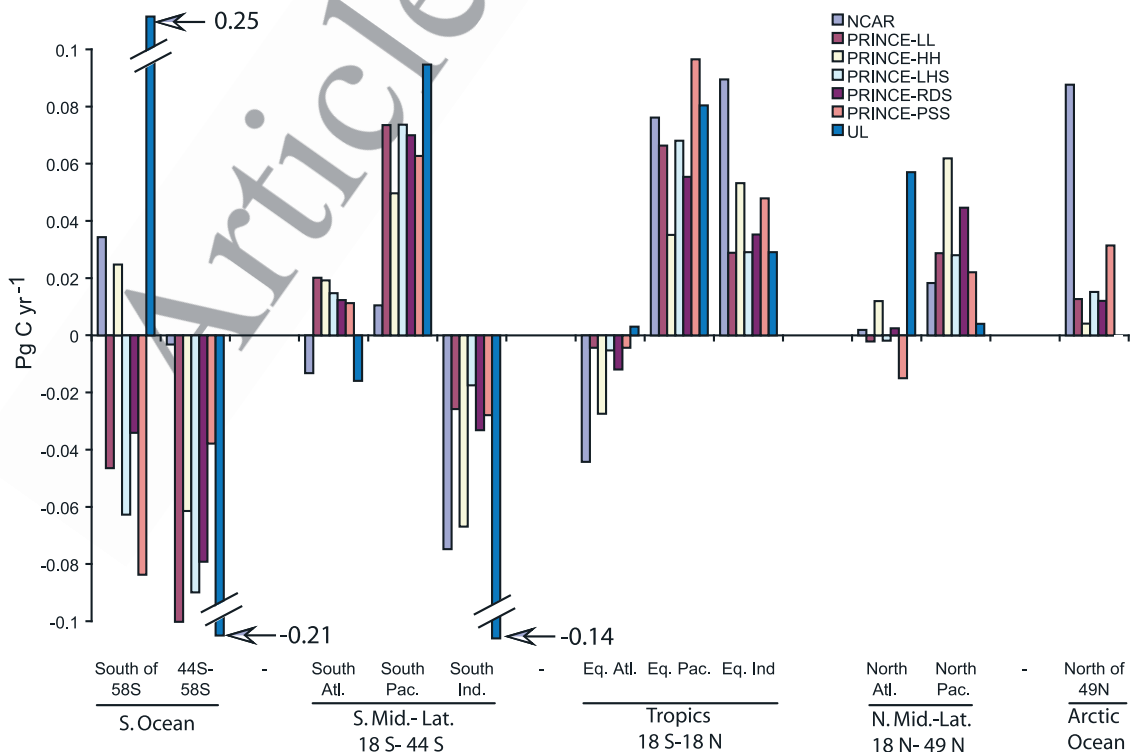


Figure 9. Zonally integrated difference between the forward and inverse anthropogenic CO₂ uptake estimates for 1995 (a positive value indicate that the forward uptake flux is larger than the inverse). Forward simulations are from OCMIP-2 [*Watson and Orr, 2003*]. Positive (negative) values indicate that the forward simulation finds more (less) anthropogenic CO₂ uptake than the inversion.

817 ECCO, and MIT models are not included because their
818 forward simulations were not available at the time of this
819 writing.

820 [53] There are clearly trends in the difference between the
821 forward and inverse estimates across all models (Figure 9).
822 The forward model simulations for those models included
823 both in this study and in OCMIP-2 find a global anthropo-
824 genic CO₂ uptake of 2.3 ± 0.32 Pg C yr⁻¹ when the mean
825 and standard deviation are weighted in the same way as the
826 inverse estimates. In comparison, the inverse estimates find
827 0.1 Pg C yr⁻¹ less uptake than the forward simulations and
828 reduce the uncertainty estimate by 22%. For most of the
829 models, the inverse anthropogenic CO₂ uptake estimates are
830 substantially larger than those of the forward model esti-
831 mates in the Southern Ocean between 44°S and 58°S and in
832 the Indian Ocean south of 18°S. This is primarily driven by
833 all of the forward models simulating a smaller anthropo-
834 genic CO₂ storage in the midlatitudes of the Southern
835 Hemisphere, particularly in the Indo-Pacific (Figure fs11
836 of the auxiliary material). In order to match the data-based
837 estimates, the inversion requires a more vigorous flux into
838 the subpolar South Atlantic and subpolar Indo-Pacific,
839 whose signal is then transported equatorward to midlati-
840 tudes. An exception to this pattern is the NCAR model, for
841 which the inversion finds a smaller anthropogenic CO₂
842 uptake in these regions compared to the forward simula-
843 tions. However, in southern midlatitudes, where most of the
844 inverse models show decreased uptake compared to the
845 forward models, the NCAR model finds increased anthro-
846 pogenic CO₂ uptake in the Atlantic and only slightly
847 decreased anthropogenic CO₂ uptake in the Pacific. This
848 suggests that fluxes from these regions contribute strongly
849 to matching the observed midlatitude storage in the NCAR
850 inversion. The other large exception is the UL model, for
851 which the inversion suggests a strong equatorward shift of
852 uptake, away from the high latitudes in the Southern Ocean.

853 [54] In the Atlantic, most of the models find more
854 anthropogenic uptake than the forward models around
855 40°N and from 18°S to the equator (Figure 9). The
856 inversion generally finds less anthropogenic CO₂ uptake
857 from 18°N to 36°N and at high northern latitudes.

858 [55] These consistent differences between the forward and
859 inverse estimates suggest that using the data-based anthropo-
860 genic CO₂ estimates to constrain the flux estimates adds new,
861 quantitative information about the spatial distribution of the
862 anthropogenic CO₂ fluxes that cannot be gained using
863 OGCMs alone. There are three possible causes for differences
864 between the forward estimates and the inverse estimates.
865 Differences could be a result of deficiencies in the model's
866 underlying physical circulation. There could be large-scale
867 biases in the data-based anthropogenic CO₂ estimates used to
868 constrain the inversion; however, the spatial pattern of the
869 inverse flux estimates have been shown to be insensitive to
870 several potential biases in the ΔC^* method. Finally, there may
871 be errors in the air-sea gas exchange in the forward models.

872 6. Conclusions

873 [56] The Green's function inverse approach presented
874 here is currently the only method that has been applied

globally to estimate the air-sea flux of anthropogenic CO₂ 875
from data-based estimates of its ocean interior distribution. 876
A related tracer-based method, the transit time distribution 877
method, has recently been developed to do this as well [Hall 878
and Primeau, 2004], but it has not yet been applied 879
globally. Other promising methods include the adjoint 880
method [Schlitzer, 2004], but this approach has not been 881
applied to estimating air-sea fluxes of anthropogenic CO₂. 882

[57] A previous inversion study employing the same 883
Green's function technique suggested that while the uncer- 884
tainty of the inversely estimated fluxes due to random errors 885
is remarkably small, substantial potential for bias exists 886
because of the uncertainty in the OGCMs used to represent 887
ocean transport and mixing [Gloor *et al.*, 2001]. Our 888
investigation using a suite of ten OGCMs suggests that 889
the inversely estimated fluxes of anthropogenic CO₂ are 890
generally insensitive to potential biases introduced by 891
OGCM transport and mixing. This is not the case for all 892
regions, though, as substantial uncertainties persist in a few 893
of them, particularly in the Southern Ocean. We also find 894
that the spatial pattern of the air-sea fluxes is remarkably 895
robust with respect to three scenarios for biases in the data- 896
based estimates of anthropogenic CO₂, but the net global 897
uptake flux scales approximately linearly with changes in 898
the global anthropogenic CO₂ inventory. We did not inves- 899
tigate the potential impact of long-term changes in ocean 900
circulation and biogeochemistry on our inversion results, 901
but on the basis of our current understanding we believe that 902
this impact has remained small so far. Given the near- 903
exponential growth of atmospheric CO₂ and radiative forc- 904
ing, we expect this impact to grow with time, however. This 905
will require the development of new methods to determine 906
the anthropogenic CO₂, as well as the use of time varying 907
circulation models in order to use this method in the future. 908

[58] On the basis of our relatively broad investigation of 909
errors and biases in data and models, we conclude that our 910
best estimate for the oceanic uptake rate of anthropogenic 911
CO₂ for a nominal year of 1995 is 2.2 Pg C yr⁻¹, with an 912
uncertainty due to errors in OGCM transport of ± 0.25 Pg C 913
yr⁻¹ (1-sigma). This represents a 22% improvement in error 914
estimates over forward simulations when the same method 915
is used to weight the standard deviation of the models. We 916
estimate that the uncertainty due to potential biases in the 917
data-based estimates is somewhat smaller than the uncer- 918
tainty due to errors in OGCM transport. The ocean inver- 919
sion provides strong constraints for the global budget of 920
anthropogenic CO₂, in particular the net uptake by the 921
terrestrial biosphere (see A. R. Jacobson *et al.*, A joint 922
atmosphere-ocean inversion for surface fluxes of carbon 923
dioxide: 2. Results, submitted to *Global Biogeochemical* 924
Cycles, 2006). 925

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