Carbon sources and sinks from an Ensemble Kalman Filter ocean data assimilation

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Abstract. We quantify contemporary and preindustrial net air-sea CO_2 8 fluxes by an Ensemble Kalman Filter assimilation of interior ocean obser-9 vations and compare results with published estimates in the light of data and 10 model uncertainties. Four different published reconstructions of anthropogenic 11 carbon and the ΔC_{qasex} tracer are assimilated into different versions of the 12 Bern3D ocean model. The two tracers represent the components of dissolved 13 inorganic carbon due to the anthropogenic perturbation and due to the air-14 sea gas exchange of natural CO₂. Contemporary air-sea fluxes for broad lat-15 itudinal bands are consistent with those from earlier ocean inversions and 16 the observed air-sea CO_2 partial pressure differences. Best agreement with 17 the pCO₂-based contemporary fluxes is found for the TTD anthropogenic 18 carbon reconstruction. We infer modest meridional transport rates of up to 19 $0.5 {\rm ~GtC~yr^{-1}}$ for the preindustrial and the contemporary ocean and a small 20 carbon transport across the equator. The anthropogenic perturbation off-21 sets the preindustrial net sea-to-air flux yielding a weak contemporary car-22 bon sink in the Southern Ocean (south of 44° S) of 0.15 ± 0.25 GtC yr⁻¹. Prein-23 dustrial Southern Ocean outgassing varies by almost a factor of two among 24 the four ΔC_{gasex} reconstructions. Large differences in regional fluxes are found 25 between an earlier ocean inversion using Green's function and this study for 26 the same model and input data calculation. Systematic differences in assim-27 ilated and optimized ΔC_{qasex} fields are large in both inversions and the con-28 temporary, anthropogenic, and preindustrial air-sea CO_2 flux in the high and 29 mid-latitude Southern Hemisphere remain uncertain. 30

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1. Introduction

Human activities cause atmospheric carbon dioxide (CO₂) and its radiative forcing to rise at at speed that is unprecedented for at least the last 20,000 years and well above the natural range of at least the past 800,000 years [*Lüthi et al.*, 2008; *Joos and Spahni*, 2008]. The main cause for the perturbation in CO₂ and climate are carbon emissions from fossil fuel use and land use changes. A quantitative understanding of the marine sources and sinks of CO₂ is an important element for understanding the role of the carbon cycle and climate-carbon cycle interactions in global warming projections [*Denman et al.*, 2007].

The contemporary air-sea fluxes of CO_2 are conceptually often separated into "natural" and "anthropogenic" components to discuss and quantify physical and biogeochemical mechanisms driving carbon sources and sink fluxes and changes in carbon inventories. The natural part represents the air-sea flux before the beginning of the industrialization (~1750 AD), when the carbon cycle was relatively close to equilibrium on decadal to centennial time scales. The anthropogenic component is the perturbation from the preindustrial state.

It has remained challenging to accurately quantify carbon sources and sink fluxes and underlying mechanisms, in part because of the large spatio-temporal variability of CO_2 fluxes [e.g. *Bakker et al.*, 2001], in part because it is not possible to measure the natural and anthropogenic components of the CO_2 air-sea flux, and of the carbon fluxes and stocks within the ocean separately.

⁵⁰ Major scientific efforts have been dedicated to quantify natural and anthropogenic and ⁵¹ contemporary carbon fluxes and stocks using both data and models. A set of studies

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aims to separate the natural and anthropogenic component of the inorganic carbon in 52 the ocean from tracer data and to reconstruct the detailed spatial distribution of natural 53 and anthropogenic carbon (C_{anth}) in the ocean [e.g. Chen and Millero, 1979; Heimann 54 and Maier-Reimer, 1996; Gruber et al., 1996; Sabine et al., 2004; Álvarez et al., 2009; 55 Vázquez Rodríguez et al., 2009]. Reconstructions yield consistently large C_{anth} inventories 56 in the North Atlantic and in the southern mid-latitude ocean, but considerable discrepan-57 cies in Southern Ocean inventories are found between different observation-based methods 58 Álvarez et al., 2009; Vázquez Rodríquez et al., 2009]. 59

⁶⁰ Another string of work is directed to establish the spatio-temporal distribution of the air-⁶¹ sea difference in the partial pressure of CO_2 (p CO_2) to calculate, in combination with an ⁶² air-sea transfer velocity [e.g. Wanninkhof, 1992; Müller et al., 2008], carbon fluxes [Tans ⁶³ et al., 1990; Takahashi et al, 2008].

Alternatively, inverse approaches are utilized to infer inter alia contemporary air-sea fluxes 64 of carbon from atmospheric and oceanic data in combination with transport models [e.g. 65 Enting and Mansbridge, 1989; Gloor et al., 2003; Baker et al, 2006; Jacobson et al., 2007; 66 C. Rödenbeck et al., 2008]. Inversions of the observed atmospheric CO_2 gradient yield 67 agreement in flux for large latitudinal bands [Denman et al., 2007], but reveal a large 68 sensitivity of inferred regional fluxes to model details and suffer from the sparse CO_2 69 sampling network with 60-120 stations [Kaminski et al., 2001]. In contrast, the oceanic 70 carbon distribution has been established by thousands of measurements [Key et al., 2004]. 71 Recently, the natural and anthropogenic air-sea fluxes of CO_2 have been quantified 72 from data-based reconstructions of C_{anth} and the tracer ΔC_{gasex} [Gloor et al., 2003; 73 Mikaloff Fletcher et al., 2006, 2007; Gerber et al., 2008]. The ΔC_{gasex} tracer reflects 74

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⁷⁵ the component of the total inorganic carbon which is due to the natural air-sea exchange ⁷⁶ of CO₂ (see section 2.1). *Gruber et al.* [2009], summarizing results from *Mikaloff Fletcher* ⁷⁷ *et al.* [2006, 2007], found in general consistency between the contemporary air-sea fluxes ⁷⁸ from the ocean inversion and the fluxes derived from the surface ocean partial pressure ⁷⁹ field of CO₂ [*Takahashi et al*, 2008] and atmospheric inversions [*Baker et al*, 2006].

Uncertainties from input data or ocean transport have been quantified [Mikaloff Fletcher] 80 et al., 2006, 2007; Gerber et al., 2008]. Mikaloff Fletcher et al. [2006, 2007] use the Green's 81 function of ten different ocean model or model setups to quantify uncertainties related 82 to ocean transport. An area of concern is that the assimilated data are not directly 83 observed, but computed from oceanic tracer distributions using constant stochiometric 84 atios. Mikaloff Fletcher et al. [2007] found that depth-dependent ratios strongly affect 85 air-sea fluxes in regions that ventilate the deep ocean, most notably the Southern Ocean. 86 Gerber et al. [2008] assessed uncertainties from systematic biases in the reconstructions of 87 anthropogenic carbon (C_{anth}) by assimilating four global and six Atlantic reconstructions 88 in their Ensemble Kalman Filter approach. The results indicate that the uptake and 89 partitioning of carbon fluxes in the Southern Ocean remain uncertain and the explicit 90 consideration of uncertainties in the C_{anth} reconstructions yields larger error bars for the 91 inferred fluxes than derived by [Mikaloff Fletcher et al., 2006] and discussed by [Gruber 92 et al., 2009]. A disturbing feature of the inversion for preindustrial air-sea fluxes is the 93 presence of substantial residuals in the ΔC_{gasex} tracer [Mikaloff Fletcher et al., 2007; Gru-94 ber et al., 2009 pointing to unsolved problems and rising a number of questions. How 95 robust are inverse estimates for the Southern Ocean region where large discrepancies in 96 reconstructed anthropogenic carbon are found? How do uncertainties in published data-97

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⁹⁸ based estimates of C_{anth} impact on the reconstruction of ΔC_{gasex} and preindustrial carbon ⁹⁹ fluxes? Are the results of ocean inversion studies as sensitive to the details of the experi-¹⁰⁰ mental setup and methods as those from atmospheric inversions?

The aim of this study to quantify preindustrial and contemporary air-sea and meridional 101 ocean carbon fluxes on the regional scale and to explore their uncertainties. We com-102 plement our earlier inversion of C_{anth} data [Gerber et al., 2008]) by assimilating ΔC_{gasex} 103 data in the Bern3D model [Müller et al., 2006] using an Ensemble Kalman Filter method 104 (EnKF) [Evensen, 2003]. There are a number of new elements compared to earlier stud-105 ies. Four different published reconstructions of C_{anth} are utilized to compute ΔC_{qasex} . 106 These reconstruction methods are: The ΔC^* method [Gruber et al., 1996], the TTD 107 method [Waugh et al., 2006], the CFC-shortcut method [Thomas and Ittekkot, 2001] and 108 the TrOCA method [Touratier and Goyet, 2004; Touratier et al., 2007]. The data are 109 assimilated into different dynamical setups of the Bern3D ocean model. The ΔC_{gasex} is 110 usually normalized to have on average a surface concentration of zero. Here, we explore 111 the impact on results of an alternative normalization. The influence of a potential bias or 112 mismatch in the seasonality of the air-sea CO_2 flux and ocean transport is investigated. 113 Last but not least, the application of an EnKF provides an alternative to the Green's func-114 tion approach applied by Mikaloff Fletcher et al. [2006, 2007] or to other data assimilation 115 methods [Schlitzer, 2002, 2007]. 116

2. Method

2.1. Data

We use a quasi-conservative tracer, ΔC_{gasex} [Sarmiento and Gruber, 2006] to infer preindustrial air-sea CO₂ fluxes. The underlying hypothesis is that gradients in ΔC_{gasex}

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are only driven by air-sea CO₂ fluxes and by physical (advection, convection, diffusion) transport within the ocean. ΔC_{gasex} is computed from measured total inorganic carbon (C_T) by removing the reconstructed anthropogenic component C_{ant}, and the estimated signals from the remineralization of organic material and calcium carbonate by assuming constant stochiometric ratios between carbon and phosphate, $\mathbf{r}_{C:P}$, and between phosphate (PO_4^{3-}) and nitrate, $\mathbf{r}_{N:P}$ in the remineralization flux of organic matter and a ratio of 0.5 between carbon and alkalinity (Alk) in calcium carbonate:

$$\Delta C_{gasex} = \frac{S_0}{S} (C_T - r_{C:P} P O_4^{3-} - 0.5 (Alk + r_{N:P} P O_4^{3-})) - C_{anth} - const.$$
(1)

 ΔC_{gasex} is computed for all available section-data points of the GLODAP database [Key 117 et al., 2004] (http://cdiac.esd.ornl.gov/oceans/glodap/GlopDV.htm) and the values as-118 signed to the appropriate grid cell of the Bern3D model. $r_{C:P}$ and $r_{N:P}$ are set to 117 and 119 16 [Anderson and Sarmiento, 1994]. The constant const is chosen to yield a mean surface 120 concentration, or alternatively global ocean inventory, of zero. ΔC_{gasex} is normalized to 121 a reference salinity $S_0 = 35psu$. The anthropogenic perturbation is removed using four 122 observation-based reconstructions of C_{anth} . The C_{anth} estimates from the ΔC^* , TTD and 123 CFC-shortcut reconstructions are available from the GLODAP site. The TrOCA C_{anth} 124 estimates are calculated following *Touratier et al.* [2007] from the GLODAP section data. 125 A detailed comparison of different C_{anth} data can be found in Vázquez Rodríguez et al. 126 [2009], a short overview of the methods is presented in *Gerber et al.* [2008], and further 127 details are available from the original publications [Gruber et al., 1996; Thomas and It-128 tekkot, 2001; Waugh et al., 2004; Touratier et al., 2007]. Three of the reconstruction 129 methods (ΔC^* , TTD and CFC-shortcut) have been investigated in the framework of an 130 OGCM [Matsumoto and Gruber, 2005; Waugh et al., 2006; Matear et al., 2003]. The ΔC^* 131

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method shows a tendency towards overestimation of C_{anth} in young water masses, but 132 underestimation in old water [Matsumoto and Gruber, 2005]. The TTD method tends 133 to be biased high in the Southern Ocean due to the assumed temporally constant air-sea 134 CO_2 disequilibrium [Waugh et al., 2006]. The CFC-shortcut method is not very reliable 135 for old waters masses such as those in the deep ocean and those found in the Southern 136 Ocean [Matear et al., 2003; Waugh et al., 2006]. Two recent studies comparing different 137 C_{anth} reconstructions in the Atlantic and the Indian suggest that the ΔC^* method yields 138 too low C_{anth} inventories in the Southern Ocean [Vázquez Rodríguez et al., 2009; Álvarez 139 et al., 2009] 140

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2.2. Model Setup

¹⁴² 2.2.1. Optimization scheme

The approach of *Gerber et al.* [2008] is used with a few adaptation and a brief overview is given next. Further detail are found elsewhere [*Gerber et al.*, 2008; *Evensen*, 2003, 2004]. The global ocean surface of the Bern3D model is divided into 17 regions (Figure S1) for which the natural net air-sea flux of carbon is optimized. The air-sea flux of CO₂ for each model region F_l is described as the product of a spatio-temporal pattern, P(i, j, t), and a scaling parameter, ψ :

$$F_l(i,j,t) = P(i,j,t)\psi(l)$$
(2)

¹⁴³ P is scaled to a unit flux (integrated over the region and year). ψ is the magnitude of the ¹⁴⁴ air-sea flux as optimized in the EnKF analysis scheme for each region l. The seasonal pat-¹⁴⁵ tern P(i, j, t) is based on the CO₂ air-sea flux climatology of *Takahashi et al.* [2002]. The

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air-sea fluxes of carbon act as a source or sink of ΔC_{gasex} in the Bern3D model. ΔC_{gasex} 146 is transported as a conservative tracer within the model ocean. After spin up of the 147 circulation, the Bern3D is initialized with a ΔC_{qasex} concentration of zero and integrated 148 over 3000 years. In the EnKF, an ensemble of 32 members is applied and each member is 149 forced with a set of the 17 flux scaling parameters ψ_l . The ensemble members are opti-150 mized in a way that minimizes the deviation between data-based estimates and modeled 151 distribution of the ΔC_{gasex} tracer. The optimization is repeated until convergence of the 152 solution, typically reached after a few iteration steps. The initial scaling parameters for 153 any region are normally distributed around zero. The ensemble size has been tested in 154 a simulation, where we included 64 instead of 32 ensemble members. Deviations in the 155 inferred air-sea fluxes are less than 0.05 GtC yr^{-1} , except for the Southern Ocean, where 156 we find a deviation up to 0.08 GtC yr⁻¹. The root-mean-square-error (RMSE) between 157 optimized and observation-based ΔC_{gasex} remains the same. 158

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¹⁶⁰ 2.2.2. Bern3D Model configurations

The Bern3D ocean model [Müller et al., 2006] is a cost-efficient coarse resolution global 161 circulation model based on the ocean model of [Edwards and Marsh, 2005]. Model results 162 are found to be in good agreement with observed distribution of different tracers Müller 163 et al., 2006, 2008; Parekh et al., 2008; Tschumi et al., 2008]. As in Gerber et al. [2008], 164 four different model setups with different circulation patterns or mixing are used to assess 165 uncertainties associated with ocean transport. These have been built to investigate exist-166 ing shortcomings in the circulation of the Bern3D model. The four model configurations 167 include the (i) Standard setup as described in Müller et al. [2006]; (ii) the $ACC \times 3$, 168

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which includes a strengthening of the Antarctic Circumpolar Current (ACC) by a factor 169 of three and a salt flux from the Pacific to the North Atlantic. Increasing the strength 170 of the ACC results in a northward expansion of the ACC and lowers the strength of the 171 southern subtropical gyres. The inserted salt flux increases the formation and propaga-172 tion of the North Atlantic Deep Water (NADW); (iii) the $PSI \times 3$ setup, which is the 173 standard setup modified by increasing the barotropic streamfunction globally by a fac-174 tor of three and applying the same salt flux as in the ACC \times 3 setup. The increased 175 barotropic streamfunction leads to strong horizontal and vertical mixing of tracers in all 176 basins. (iv) the *High Diffusion* setup is the same as the standard setup except that di-177 apycnal diffusion is increased by a factor of four for passive tracer and is set to 4×10^{-5} 178 m s⁻². Figure (S2) shows the overturning of the different circulation setups. The Green's 179 function of the standard setup has been used earlier in the inversions of *Mikaloff Fletcher* 180 et al. [2006, 2007]. The skill of the Bern3D model in representing the observation-based 181 radiocarbon distribution compares with a score of 0.93 favorably to that of general circu-182 lation ocean models with a range of skill scores from 0.65 to 0.94 [Mikaloff Fletcher et al., 183 2007]. 184

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2.3. Calculation of contemporary air-sea fluxes and of best estimates

¹⁸⁶ Contemporary air-sea fluxes for each region are calculated as the sum of the natural ¹⁸⁷ fluxes inferred from the ΔC_{gasex} tracer, the anthropogenic air-sea fluxes of *Gerber et al.* ¹⁸⁸ [2008] for the nominal year 1995, and a river-derived outgassing taken from *Jacobson et al.* ¹⁸⁹ [2007] and as used in *Gruber et al.* [2009]. The input of organic and inorganic carbon by

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¹⁹⁰ rivers causes an outgassing of CO₂ [Sarmiento and Sundquist, 1992] that is not resolved ¹⁹¹ by the inversion.

¹⁹² Best estimates are calculated by averaging the results obtained for the four different ¹⁹³ ΔC_{gasex} fields. Individual estimates are weighted using the same skill score as *Gerber et al.* ¹⁹⁴ [2008] and derived from the assimilated and modeled C_{anth} fields. For ocean transport ¹⁹⁵ and river outgassing, we apply the same uncertainties as *Gruber et al.* [2009]. Overall un-¹⁹⁶ certainties are estimated by adding uncertainties from the ΔC_{gasex} fields, ocean transport, ¹⁹⁷ and river-driven outgassing using Gaussian error propagation. All uncertainties provided ¹⁹⁸ represent ± 1 standard deviation.

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3. Results

3.1. Large-scale carbon sources and sink fluxes

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The results of the Ensemble Kalman Filter (EnKF) inversion are aggregated for each 201 basin into broad latitudinal bands to ease comparison with other studies. The aggregated 202 Bern3D-EnKF inversion yields the familiar pattern of outgassing of CO_2 in the tropics 203 and ocean uptake in the temperate and high latitude ocean (Figure 1). The contempo-204 rary (1995 AD) tropical source (19°S to 16°N) is estimated to be 0.60 ± 0.16 GtC yr⁻¹. 205 The contemporary high-latitude sinks are 0.15 ± 0.25 GtC yr⁻¹ in the Southern Ocean 206 and 0.25 ± 0.07 GtC yr⁻¹ in the northern North Atlantic. The sink in the mid latitude 207 ocean amounts to 1.20 ± 0.16 GtC yr⁻¹ in Southern and to 0.64 ± 0.13 GtC yr⁻¹ in the 208 Northern Hemisphere. 209

²¹⁰ The attribution of the contemporary fluxes to a preindustrial flux (natural plus river-

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²¹¹ ine outgassing), and to an anthropogenic perturbation reveals underlying oceanographic ²¹² mechanisms (Table 1 and Figure 2). The perturbation flux is into the ocean in all re-²¹³ gions, with largest sink fluxes of C_{anth} into the Southern Ocean. At preindustrial time, ²¹⁴ the inferred tropical source is larger than today, whereas the temperate and northern ²¹⁵ high-latitude sink fluxes are considerably smaller than today. In the Southern Ocean, the ²¹⁶ natural (preindustrial) component corresponds to a source of 0.69 ± 0.15 GtC yr⁻¹ that ²¹⁷ is more than offset by an uptake flux of 0.86 ± 0.25 GtC yr⁻¹ of C_{anth}.

The flux patterns are consistent with the generally accepted picture of ocean circulation 218 and the observed surface temperature and nutrient distributions. The preindustrial pat-219 tern is the result of the interplay of the solubility pump and the biological pump [Volk and 220 Hoffert, 1985; Maier-Reimer, 1993; Murnane et al., 1999; Sarmiento et al., 2000]. Warm-221 ing causes the pCO_2 to rise favoring outgassing in the warm tropical region and uptake 222 in the mid- and high-latitude ocean. Formation of organic material tends to lower pCO_2 , 223 whereas remineralization tends to enhance pCO_2 . Consequently, we expect a tendency 224 towards CO_2 outgassing in regions where nutrient and carbon rich water is brought to the 225 surface and uptake from the atmosphere where organic matter is formed. Thus, the CO_2 226 outgassing in the tropics and in the Southern Ocean is explained by upwelling of nutri-227 ent rich waters, whereas the sink in mid-latitude regions and in the high latitude North 228 Atlantic is consistent with the low nutrient concentrations and with cooling of poleward 229 flowing waters. 230

In the inversion, the preindustrial air-sea flux is a direct consequence of the gradients in ΔC_{gasex} and ocean model transport. For example, Circumpolar Deep Water (CPDW) with high ΔC_{gasex} concentrations is transported towards the surface, moving northward

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²³⁴ by the Ekman drift and subsequently transformed into Antarctic Intermediate Water ²³⁵ (AAIW). Thus, the decrease in ΔC_{gasex} concentration of order 50 μ mol between CPDW ²³⁶ to AAIW implies a strong CO₂ outgassing in the Southern Ocean.

Turning to the anthropogenic perturbation, the accelerating atmospheric CO_2 growth, 237 causes a positive perturbation in the surface atmosphere-ocean partial pressure difference 238 relative to the natural, preindustrial situation and uptake of C_{anth} in all regions. The 239 uptake of C_{anth} is expected to be particularly large in regions were "old" water, that has a 240 low concentration of C_{anth} and a large potential for C_{anth} uptake, is upwelled or mixed by 241 convection into the surface. A confounding factor is warming and cooling. Warming has 242 the tendency to enhance C_{anth} uptake as the Revelle factor and thus the uptake capacity 243 for C_{anth} is lower for warm than for cold water. The strong anthropogenic uptake flux in 244 the Southern Ocean appears to be driven by upwelling of Circumpolar Deep Water and 245 subsequent warming as the water is moved northwards by the Ekman drift before being 246 subducted as AAIW and Subantarctic Mode Water (SAMW). This results in large C_{anth} 247 inventories in the Southern Hemisphere mid-latitude ocean [Sabine et al., 2004]. Uptake 248 fluxes per unit area are also relatively high in the upwelling regions of the tropics and in 249 the Nordic Seas where convection and North Atlantic Deep Water formation carry C_{anth} 250 efficiently to the abyss. (Figure S2). 251

The reconstructed contemporary fluxes for broad latitudinal bands of individual ocean basins are broadly consistent with the results of the Green's function ocean inversion (GFOI) [*Gruber et al.*, 2009], the fluxes from the pCO₂ climatology [*Takahashi et al*, 2008], and the atmospheric inversion of *Baker et al* [2006]. The EnKF results agree with the results from the GFOI for each aggregated region within their uncertainties. Dif-

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ferences between the two studies are large for the Southern Ocean and the Pacific Mid 257 Latitude region. The EnKF inversion, as well as the GFOI, yield higher contemporary 258 uptake fluxes in the southern mid-latitudes than the pCO_2 climatology. The disagreement 259 is particularly striking for the temperate Pacific region, a region where sampling density 260 for the pCO_2 climatology is low [Takahashi et al, 2008]. Interestingly, the atmospheric 261 inversion of *Baker et al* [2006] suggests a small net sea-to-air flux in the mid-latitude 262 Southern Pacific, a finding that is in strong contrast to the uptake flux inferred by all 263 oceanographic methods. 264

Next, we follow the setup of *Gerber et al.* [2008] for anthropogenic fluxes and investigate uncertainties in natural fluxes by applying different ocean transport fields and different reconstructions of ΔC_{gasex} . In addition, we explore sensitivity of air-sea fluxes to the prescribed seasonal flux pattern.

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3.2. Uncertainties in inferred natural air-sea CO_2 fluxes

3.2.1. Sensitivity of natural air-sea fluxes to ocean transport and to prescribed seasonal flux pattern

Four different circulation setups of the Bern3D ocean model are applied with the same ΔC_{gasex} data. The latter are computed by removing the anthropogenic signal reconstructed with the ΔC^* method [*Gruber et al.*, 1996; *Key et al.*, 2004] and by normalizing the ΔC_{gasex} field to have zero average surface concentration. A few regions are particularly sensitive to changes in circulation: the polar and subpolar Southern Ocean, the high-latitude North Atlantic, and the southern mid-latitude regions (Table 2).

The more vigorous Atlantic overturning and the deeper penetration of NADW in the

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"ACC" and "PSI × 3" setups leads to a higher uptake flux in the high- and mid-latitude North Atlantic. The inferred uptake flux north of 34°N in the Atlantic is more than twice as high for these two settings than for the standard setup. Consequently, the export of ΔC^* from the deep North Atlantic to the Southern Hemisphere is enhanced. The enhanced strength of the circumpolar currents causes a more vigorous exchange between the Southern Ocean and the mid-latitude ocean, enhancing uptake in the mid-latitude Pacific and mid-latitude Indian regions and outgassing in the Southern Ocean. The increased uptake in the North Atlantic and the Pacific and Indian southern mid-latitude regions is mostly balanced by enhanced outgassing in the Southern Ocean region by 0.19 and 0.30 GtC yr⁻¹, and a decrease in the aggregated net air-to-sea flux in the mid-latitude and tropical Atlantic (46°S to 34°N) by 0.14 and 0.18 GtC yr⁻¹ relative to the standard setup. Increasing vertical diffusivity for passive tracers by a factor of four has a small effect (< 0.06 GtC yr⁻¹) on inferred sources and sinks (Table 2).

Observation-based concentrations of ΔC_{gasex} are much higher than model results in the deep Pacific and Atlantic (Figure 3). We have modified the seasonal pattern for the northern high latitude Atlantic and for the polar Southern Ocean towards larger uptake rates during the winter time and less uptake or even outgassing during summer time. Thereby, we tend to increase the ΔC_{gasex} concentrations in the surface during times of deep water formation in the Nordic Seas and the Southern Ocean and to reduce the data-model difference in the deep. Technically, the seasonal pattern of the two regions has been shifted by an offset of $0.95 \times \bar{p_n}$ where $\bar{p_n}$ is the temporal and spatial average flux of each grid box:

$$\tilde{p}(i,j,t)_n = \frac{p(i,j,t)_n - 0.95\bar{p}}{\int_{i,j,t} (p(i,j,t) - 0.95\bar{p})}$$
(3)

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We emphasize that there is no reason to modify the prescribed spatio-temporal pattern 272 from the pCO_2 climatology in such a drastic way and that this modification represents an 273 extreme case. The inferred uptake pattern in the North Atlantic is changing towards less 274 uptake in the North Atlantic High Latitude region, entirely compensated by increased up-275 take in the Northern Mid Latitude region. The Southern Ocean outgassing is reduced by 276 0.07 GtC yr^{-1} relative to the standard and uptake in the Atlantic Southern Mid-latitude 277 region is reduced from 0.23 to 0.09 GtC yr⁻¹, whereas results are similar as in the standard 278 setup for other regions. The modification in seasonal forcing did not improve the modeled 279 distribution of ΔC_{aasex} and residuals in the deep Atlantic and deep Pacific are still high. 280 Thus, a potential mismatch in the seasonality of the air-sea flux and ocean transport is 281 not an explanation for the large residuals. 282

An intriguing finding of this first set of sensitivity simulations is that the global average 283 air-sea CO_2 flux does not vanish. The inversion yields a global uptake flux of 0.2 GtC 284 yr^{-1} (Table 2). This is inconsistent with the assumption of an ocean in steady state at 285 preindustrial time. We attribute this to the choice of the normalization constant in the 286 computation of ΔC_{qasex} . Following earlier work, the constant has been set to yield an 287 average surface concentration of zero and a whole ocean inventory of the ΔC_{qasex} tracer 288 of around 600 GtC. The implication is that a global uptake of 0.2 GtC yr^{-1} is required 289 in our 3000 year long simulations to match the global ΔC_{gasex} inventory. Non-vanishing 290 global air-to-sea net fluxes, within the range of 0.14 to -0.1 GtC yr^{-1} across the different 291 ocean transport models, have also been found by *Mikaloff Fletcher et al.* [2007]; they also 292 normalized ΔC_{qasex} to a zero mean surface concentration. 293

²⁹⁴ The ΔC_{gasex} field is renormalized to yield a zero ocean inventory to avoid the implicit

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requirement of a net ocean uptake. This is permissible as ocean tracer transport is described by a set of linear equations and the addition of a constant does not change tracer divergence. As expected, the global net air-sea flux is reduced close to zero in the EnKF (Table 2). Surprisingly, this reduction is almost entirely achieved by an increase in Southern Ocean outgassing by 0.19 GtC yr⁻¹.

In summary, the air-sea flux for the Southern Ocean region appears to be particularly sensitive for the details of the model setup and the ΔC_{gasex} field should preferentially be normalized to yield a zero global inventory. We note that the results presented in the Figures and in Tables 1 and 3 and further discussed below have all been obtained by applying ΔC_{gasex} fields normalized to a zero inventory.

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$_{306}$ 3.2.2. Sensitivity of natural air-sea fluxes to different reconstructions of $_{307}$ ΔC_{gasex}

Next, we investigate the sensitivity of the natural air-sea fluxes to the four different ΔC_{gasex} fields. At this stage, we only use the *Standard* circulation setup of the Bern3D model.

³¹¹ The four ΔC_{gasex} reconstructions differ remarkably. The root-mean-square-error (RMSE) ³¹² between the different ΔC_{gasex} fields ranges from 6.91 μ mol l⁻¹ (between TTD and CFC) ³¹³ to 13.17 μ mol l⁻¹ (between TrOCA and CFC). Figure 4 shows the zonally-averaged dis-³¹⁴ tribution for each of the four reconstructions in the upper Pacific. Differences are only ³¹⁵ due to the use of different C_{anth} reconstructions. Large differences in ΔC_{gasex} of order 20 ³¹⁶ μ mol l⁻¹ are found in various regions. The TrOCA reconstruction yields higher values in ³¹⁷ the Pacific equatorial thermocline and lower values in the Pacific AAIW than any of the

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other three reconstructions. In the North Pacific thermocline around 50°N, ΔC_{qasex} con-318 centrations are higher for the ΔC^* and the TrOCA reconstructions than for the TTD and 319 CFC reconstructions; CFC concentrations suggest the presence of a substantial amount 320 of anthropogenic carbon and thus a relatively low ΔC_{gasex} concentrations in this region. 321 As with the different circulations, the magnitude of flux in the Southern Ocean, the south-322 ern mid-latitudes, and in the high latitude Atlantic are particularly sensitive to the choice 323 of the ΔC_{gasex} fields (Figure 5, Table 2). For all other regions, deviations are less than 324 0.06 GtC yr^{-1} . Inferred outgassing in the Southern Ocean varies almost by a factor of 325 two for the different reconstructions and ranges from 0.53 (TTD, CFC) to 0.98 GtC yr⁻¹ 326 (TrOCA). The TrOCA- and the ΔC^* - based reconstructions yield a higher concentra-327 tion of C_{anth} and thus a lower ΔC_{gasex} in the AAIW compared to the CFC and TTD 328 reconstructions. This implies a stronger preindustrial Southern Ocean outgassing for the 329 TrOCA and the ΔC^* reconstructions. The TrOCA reconstruction yields also a very high 330 uptake flux in the southern mid-latitude Indian ocean, probably as a result of relatively 331 high ΔC_{qasex} concentrations in SAMW. 332

It might be instructive to compare the fluxes deduced from the different reconstructions and model setups with those derived from the pCO₂ climatology of *Takahashi et al* [2008]. The TTD-based fluxes show the best agreement in terms of correlation and relative standard deviation with the pCO₂-based fluxes [*Taylor*, 2001]. The CFC estimates yield a larger standard deviation and the ΔC^* and TrOCA estimates also a smaller correlation. Results from the standard circulation setup compare better with the pCO₂-derived fluxes than those from alternative circulation setups (Figure S3).

 $_{340}$ In conclusion, the explicit consideration of uncertainties in ΔC_{gasex} explains the larger

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error bars given in this study compared to those estimated by *Gruber et al.* [2009] and *Mikaloff Fletcher et al.* [2007]. Best agreement with the pCO₂-based contemporary fluxes is found for the standard circulation setup and the TTD reconstruction.

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345 3.2.3. Sensitivity of natural air-sea fluxes to the inversion method

The Green's function of the Bern3D model have also been used in the inversion of 346 Mikaloff Fletcher et al. [2007] allowing us to compare the sensitivity of the flux to the 347 choice of inversion method and experimental details. Surprisingly large differences are 348 found given that the same ΔC_{qasex} data calculation and the same ocean model are ap-349 plied in both inversions (Table 2). Southern Ocean outgassing is 0.4 GtC yr^{-1} larger in 350 the EnKF inversion than in the GFOI. Uptake in the southern mid-latitude Pacific and 351 Indian is also larger in the EnKF inversion, but results are similar for the southern mid-352 latitude Pacific. The GFOI yields 0.2 GtC yr^{-1} more outgassing in the southern tropical 353 Pacific than the EnKF. 354

The GFOI yields a relatively low preindustrial uptake flux in the northern high latitude 355 Atlantic and a substantial uptake in the northern mid latitude Atlantic. This has been 356 previously attributed to a tendency of the ocean models applied in the GFOI to produce 357 North Atlantic Deep Water too far south, with little or no formation in the Nordic Seas 358 [Gruber et al., 2009]. However, this shortcoming is not evident in the Bern3D-EnKF in-359 version that yields no preindustrial uptake flux in the mid latitude North Atlantic and 360 a contemporary northern high-latitude flux that is almost identical to the flux from the 361 pCO_2 climatology (Figure 1). 362

³⁶³ It is beyond the scope of this study to identify in detail the experimental differences

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that lead to the different results for the EnKF and the GFOI. However, residuals be-364 tween assimilated and modeled data may provide an indication (Figure 3 and Figure 7 365 in *Mikaloff Fletcher et al.* [2007]). Both inversions yield substantial root mean square 366 deviations between assimilated and optimized ΔC_{qasex} for all ocean models and for all 367 experimental setups. The inversions yield a smoother distribution in ΔC_{qasex} with weaker 368 gradients than reconstructed. We have implemented the fluxes from the GFOI in a for-369 ward simulation with the Bern3D model and found a root mean square deviation between 370 reconstructed and simulated ΔC_{qasex} field of 18.7 μ mol l⁻¹ compared to the 16.86 μ mol 371 $^{-1}$ for EnKF inversion. The difference is likely explained by different weighting of the 372 assimilated data in the GFOI and the EnKF and should not be interpreted as an indi-373 cation for the quality of the two inversions. Both inversions have difficulties to represent 374 the reconstructed ΔC_{gasex} and the qualitatively similar flux patterns of the EnKF and of 375 the GFOI yield similar root mean square deviations. 376

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$_{378}$ 3.2.4. Deviations between assimilated and modeled ΔC_{gasex} concentrations

The RMSE between the reconstructed and modeled ΔC_{qasex} fields are large for all setups 379 Table 2, 3). RMSE are between 18 (standard) and 14 μ mol l⁻¹ ("PSI × 3") for different 380 circulations and between 16 (standard) and 20 μ mol l⁻¹ for the different ΔC_{qasex} recon-381 struction and standard circulation. The zonally-averaged residuals in the Atlantic, Pacific 382 and Indian range between -40 and +40 μ mol l⁻¹, comparable to reconstructed ΔC_{qasex} 383 range of -60 to +60 μ mol l⁻¹ (Figure 3). Part of the data-model mismatch is very likely 384 related to deficiencies in the Bern3D transport. Most notable is a too weak formation 385 and northward penetration of AAIW and a too shallow penetration of NADW in the 386

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³⁸⁷ standard setup. However, large residuals and RMSE are not unique to the Bern3D-EnKF ³⁸⁸ assimilation, but have also been found for each of the ten model or model versions used ³⁸⁹ by *Mikaloff Fletcher et al.* [2007] with a reported RMSE range between 16 and 21 μ mol ³⁹⁰ kg⁻¹. These large residuals may point to some fundamental problems with the ΔC_{gasex} ³⁹¹ tracer.

 ΔC_{gasex} is advected and mixed from the surface and has no sources and sinks in the 392 interior ocean implying that concentrations in the interior should be within the concen-393 tration range of the source regions. However, interior concentrations often appear to be 394 outside the concentration range of potential source regions. For the ΔC^* reconstruction, 395 concentrations are below 40 μ mol l⁻¹ between 500 and 1000 m, whereas higher concen-396 trations (up to 68 μ mol l⁻¹) are again found at greater depths in the North Atlantic. 397 The relatively high concentrations in the thermocline and the deep northern Pacific in 398 the ΔC^* reconstruction appear also not to be reflected in any of the potential source 399 waters. Concentrations in the deep (> 2000 m) Pacific are up to 35 μ mol l⁻¹, whereas 400 concentrations in the Southern Ocean surface and in the deep Indian and in the Atlantic 401 sector of the Southern Ocean are clearly lower. Similarly, the relatively high values of 402 5 μ mol l⁻¹ in the upper thermocline around 35°S in the Indian and Atlantic appear to 403 have no correspondence in adjacent source regions. As a consequence, the assimilation 404 is not able to match the ΔC_{qasex} concentrations simultaneously. Data-model misfits are 405 also found in the GFOI of *Mikaloff Fletcher et al.* [2007]. 406

⁴⁰⁷ How can these apparent inconsistencies in ΔC_{gasex} be explained? Non-representative ⁴⁰⁸ sampling may add uncertainties, but we rely on around 47'000 samples to yield 15897 ⁴⁰⁹ data points on the Bern3D model grid. Analytical uncertainties in the measurements of

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 C_T , Alk, and PO_4^{3-} are too small and uncertainties in the reconstructed C_{anth} fields can 410 also not explain the data-model mismatch. Potentially important are uncertainties in the 411 Redfield factors used in equation 1. For example, typical concentrations of PO_4^{3-} in the 412 North Pacific are around 3 μ mol l⁻¹. A decrease in $(R_{C:P}+0.5\times R_{N:P})$, used in equation 1, 413 by 10% yields an increase in ΔC_{gasex} of 37 μ mol l⁻¹. Alternatively, interior concentrations 414 outside the source region range may be a consequence of past variability in air-sea CO_2 415 fluxes or ocean transport, e.g. in response to volcanic eruptions or as part of the internal 416 climate variability [Frölicher et al., 2009]. 417

In summary, large residuals are found for all individual setups. This may point to fundamental problems with the ΔC_{gasex} tracer, possibly related to spatial variations in the Redfield ratio between PO_4^{3-} and C_T of order 5 to 10 percent or related to ocean variability.

421

422 **3.2.5.** Meridional ocean carbon transport

Meridional ocean transport rates are linked to the pattern of air-sea gas exchange of 423 CO₂. For the preindustrial steady-state the net air-sea flux corresponds to the divergence 424 in transport. The southward transport increases steadily from the northern high latitudes 425 to reach 0.6 GtC yr^{-1} at 19°N. In other words, the temperate and high-latitude Northern 426 Hemisphere ocean absorbs on average 0.6 GtC yr^{-1} at preindustrial times. Maxima in 427 equatorwards-directed ocean transport are 0.6 GtC yr^{-1} and 0.3 GtC yr^{-1} in the Northern 428 and Southern Hemisphere, supporting a tropical outgassing of 0.9 GtC yr^{-1} . Transport 429 across the equator is small $(0.24 \text{ GtC yr}^{-1})$. The carbon uptake in the southern mid-430 latitudes is reflected in the decrease in northward transport by 0.84 GtC yr^{-1} from 19°S 431 to 44° N. The outgassing of the Southern Ocean is supported by a corresponding prein-432

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433 dustrial southward transport.

For the individual basins, we infer an extensive preindustrial southwards transport of CO₂ throughout the Atlantic (Table S1 and Figure 6), whereas transport rates are equatorwards in the Indo-Pacific (north of 40°S). The southward transport in the Atlantic is mainly driven by the formation and export of NADW. In the upper Atlantic (> 1000 meter) carbon transport is directed polewards in the northern high and mid latitudes.

The transport of anthropogenic carbon is primarily northwards. The most important transport pathway is from the Southern Ocean to the mid-latitudes and subtropics in connection with the northward spreading of AAIW and Subantarctic Mode Water, while transport rates are somewhat smaller in the Northern Hemisphere. A substantial fraction of the anthropogenic carbon taken up in the Southern Ocean is exported to the midlatitudes.

⁴⁴⁵ Contemporary meridional ocean carbon transport is directed southwards in the Northern ⁴⁴⁶ Hemisphere and south of 42°S, and northwards in the temperate and tropical Southern ⁴⁴⁷ Hemisphere (Figure 6). The large transport of carbon into the Southern Ocean inferred ⁴⁴⁸ for the preindustrial state is largely offset by the northward transport of anthropogenic ⁴⁴⁹ carbon and reduced by about a factor of three. Here, we neglect any possible contribution ⁴⁵⁰ from river-derived carbon.

The spread in transport among the different reconstruction methods is small in the tropical regions (0.05 GtC yr⁻¹), but large for the transport from the southern mid latitudes to the Southern Ocean. The range in the contemporary flux into the Southern Ocean region is from zero flux to 0.33 GtC yr⁻¹ across the four different input data sets. The range (0.54 to 0.98 GtC yr⁻¹) is even larger for the preindustrial flux.

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In summary, we infer relatively modest meridional transport rates for both the preindustrial and the contemporary ocean and a small carbon transport across the equator. The preindustrial carbon transport into the Southern Ocean is partly offset by the export of anthropogenic carbon out of the Southern Ocean.

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4. Discussion and Conclusion

An Ensemble Kalman Filter is combined with the Bern3D ocean model and tracer data 461 to quantify preindustrial, anthropogenic, and contemporary air-sea fluxes of CO_2 and 462 meridional carbon transport within the ocean. We find a substantial preindustrial out-463 gassing in the tropical and in the Southern Ocean, and carbon uptake in the mid - and 464 northern high latitudes as well as a small interhemispheric carbon transport. The anthro-465 pogenic carbon uptake offsets the preindustrial net sea-to-air CO_2 flux in the Southern 466 Ocean, implying that the Southern Ocean is currently on average a weak carbon sink 467 for the atmosphere. The reconstructed contemporary fluxes for broad latitudinal bands 468 are largely consistent with results from Green's function ocean inversions [Gloor et al., 469 2003; Jacobson et al., 2007; Gruber et al., 2009], the fluxes computed from the surface 470 ocean pCO₂ climatology [Takahashi et al, 2008], and atmospheric inversions [Baker et al, 471 2006. However, sensitivity simulations and the analysis of deviations between assimilated 472 and optimized tracer fields reveal large uncertainties in regional air-sea flux, in particular 473 for the Southern Ocean. Improvements in quantification and understanding of Southern 474 Ocean processes is high on the research agenda as this region plays a critical role for the 475 fate of anthropogenic carbon and for the glacial-interglacial CO_2 variations. 476

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The reconstruction of anthropogenic carbon is a potential source of biases in the esti-477 mates of ΔC_{gasex} . Another bias may be imposed by the normalization of the ΔC_{gasex} 478 field. We recommend to normalize the ΔC_{gasex} reconstruction to a zero mean ocean 479 inventory instead to a zero mean surface concentration to avoid non-vanishing global air-480 sea fluxes in the assimilation of ΔC_{qasex} data. Regional air-sea fluxes from the EnKF 481 assimilations are compared to those inferred from observations of the air-sea CO_2 partial 482 pressure differences [Takahashi et al, 2008]. Overall, best agreement is found when relying 483 on the TTD reconstruction [Waugh et al., 2006] in the EnKF assimilation. The explicit 484 consideration of uncertainties in the reconstructions of C_{anth} and ΔC_{qasex} explains the 485 larger error bars given in this study compared to those estimated by Gruber et al. [2009] 486 and Mikaloff Fletcher et al. [2007]. 487

⁴⁸⁸ Optimized air-sea fluxes are also sensitive to the choice of the inversion method. For ⁴⁸⁹ example, preindustrial outgassing in the Southern Ocean is with 0.7 GtC yr⁻¹ more than ⁴⁹⁰ twice as large in the EnKF inversion than in the corresponding Green's function inversion ⁴⁹¹ when applying the same ocean model and ΔC_{gasex} data calculation. This suggests that ⁴⁹² the air-sea fluxes are not tightly constrained by the assimilated data and details of the ⁴⁹³ inversion methods matter.

Large root mean square deviations between the assimilated and the optimized ΔC_{gasex} fields of 15 to 20 μ mol l⁻¹ are found for all individual setups of this study or by *Mikaloff Fletcher et al.* [2007] who applied ten different models and model versions in their Green's function inversion. The range of zonal mean residuals in ΔC_{gasex} is comparable to the reconstructed range in ΔC_{gasex} . The inversions yield a much smoother distribution in ΔC_{gasex} and gradients are much weaker than reconstructed. This may

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⁵⁰⁰ point to fundamental problems with the ΔC_{gasex} tracer, possibly related to spatial varia-⁵⁰¹ tions in the stochiometric ratios between phosphate and carbon of order 5 to 10 percent ⁵⁰² or related to ocean variability. These two factors should be more explicitly addressed in ⁵⁰³ future work.

In conclusion, oceanic tracer data have been successfully assimilated with an Ensemble 50 Kalman Filter to quantify regional air-sea fluxes and meridional ocean transport of car-505 bon. Results for the Southern Ocean, southern mid-latitudes and the northern North 506 Atlantic regions are particularly sensitive to uncertainties in input data, to uncertain-507 ties in ocean transport, and to the choice of inverse method and experimental details. 508 Systematic differences in assimilated and optimized ΔC_{gasex} fields remain uncomfortably 509 large, suggesting that the error estimates given in this study represent lower bounds. The 510 contemporary, anthropogenic, and preindustrial air-sea CO_2 flux in the Southern Ocean 511 remain uncertain. 512

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Acknowledgments. This study was financially supported by the European Union through the Integrated Project CarboOcean(511106-2), the Swiss National Science Foundation, and the Swiss Staatsekretariat für Bildung und Forschung (#C07.0068; COST Action 735).

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preindustrial time, and the present in GtC yr⁻¹. Fluxes of this study are compared to those from the ocean inversion of *Mikaloff Fletcher et al.* [2006, 2007] and those derived from a climatology of the air-sea CO₂ partial pressure difference [*Takahashi et al*, 2008] as published by *Gruber et al.* [2009]. Fluxes of this study represent weighted averages of the fluxes shown in Table 3 for four different reconstructions of C_{ant} and C_{gasex} . Uncertainties represent ±1 standard deviation. Uncertainties for this study include those from uncertainties in input data, ocean transport, and river-driven outgassing

Table 1.

Region	This study			Miklot	f Fletcher et al	Takahashi et al. 2008	
	C_{anth}	Natural	Total	C_{anth}	Natural	Total	Total
Atlantic N.High Lat.	0.13 ± 0.06	$0.16\pm~0.04$	0.25 ± 0.07	0.09 ± 0.04	0.11 ± 0.02	0.17 ± 0.06	0.25
Atlantic N. Mid Lat.	0.10 ± 0.05	0.02 ± 0.05	0.11 ± 0.06	0.13 ± 0.04	0.12 ± 0.04	0.24 ± 0.06	0.15
Atlantic N. Low Lat.	0.04 ± 0.05	0.16 ± 0.04	0.16 ± 0.06	0.04 ± 0.04	0.08 ± 0.04	0.08 ± 0.06	0.05
Atlantic N. Tropics	0.03 ± 0.02	-0.11 ± 0.02	-0.17 ± 0.05	0.04 ± 0.02	-0.03 ± 0.01	-0.08 ± 0.05	-0.03
Atlantic S. Tropics	0.11 ± 0.03	-0.09 ± 0.02	0 ± 0.04	0.09 ± 0.02	-0.14 ± 0.02	-0.06 ± 0.03	-0.09
Atlantic S. Low Lat.	0 ± 0.01	0 ± 0.01	0 ± 0.01	0.02 ± 0.01	-0.02 ± 0.01	0.01 ± 0.01	-0.02
Atlantic S. Mid Lat.	$0.05 {\pm} 0.04$	$0.17 {\pm} 0.08$	0.19 ± 0.08	0.05 ± 0.02	0.11 ± 0.05	0.16 ± 0.05	0.13
Pacific N. High Lat.	$0.05 {\pm} 0.03$	0 ± 0.04	0.02 ± 0.05	0.04 ± 0.01	-0.02 ± 0.04	0 ± 0.04	0.12
Pacific N. Mid Lat.	$0.05 {\pm} 0.06$	$0.25 {\pm} 0.06$	0.35 ± 0.08	0.15 ± 0.04	0.31 ± 0.05	0.42 ± 0.07	0.37
Pacific N. Tropics	0.18 ± 0.05	-0.30 ± 0.06	-0.15 ± 0.08	0.18 ± 0.04	-0.22 ± 0.06	-0.06 ± 0.07	-0.12
Pacific S. Tropics	0.12 ± 0.05	-0.25 ± 0.08	-0.14 ± 0.10	0.11 ± 0.02	-0.41 ± 0.08	-0.31 ± 0.09	-0.32
Pacific S. Mid Lat.	$0.09 {\pm} 0.06$	$0.52 {\pm} 0.10$	0.61 ± 0.11	0.11 ± 0.04	0.36 ± 0.09	0.46 ± 0.10	0.28
Indian Tropics	$0.07 {\pm} 0.04$	-0.12 ± 0.03	-0.14 ± 0.06	0.11 ± 0.03	-0.14 ± 0.02	-0.12 ± 0.06	-0.13
Indian S. Mid Lat.	0.12 ± 0.12	$0.29 {\pm} 0.12$	0.40 ± 0.09	0.25 ± 0.08	0.22 ± 0.06	0.46 ± 0.09	0.37
Polar and Subpolar Ocean	0.86 ± 0.25	-0.69 ± 0.15	0.15 ± 0.25	0.74 ± 0.17	-0.40 ± 0.11	0.34 ± 0.20	0.30
Global	2.10 ± 0.30	-0.01 ± 0.09	1.67 ± 0.38	2.18 ± 0.25	-0.05 ± 0.08	1.70 ± 0.35	1.31

Regional net air-to-sea fluxes of CO_2 for the anthropogenic perturbation, the

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Table 2. Regional net air-to-sea fluxes in GtC yr⁻¹ of natural (preindustrial without riverderived) CO₂ for four different Bern3D transport versions and two modified setups with the standard Bern3D model. The root mean square error (RMSE) between optimized and databased fields of C_{gasex} are calculated using all data points assimilated to the optimization. The column "zero inventory" shows the air-sea fluxes inferred from the ΔC^* reconstruction with an offset such that the total inventory of the ΔC_{gasex} is zero (see equation 1). The "seasonally forced" column shows air-sea fluxes with a modified flux pattern in the northern high Atlantic and Southern Ocean to increase the uptake during the winter months. The "Ocean Inversion"

column shows the results from the Bern3D model in Mikaloff Fletcher et al. [2007]

Region	Area in $10^6 km^2$	Standard	ACC	PSI $\times 3$	High	Seasonally	zero	Ocean
			$\times 3$		Diffusion	Forced	Inventory	Inversion
Atlantic N.High Lat.	9.45	0.21	0.32	0.43	0.21	0.12	0.18	0.14
Atlantic N. Mid Lat.	11.81	-0.01	0.10	0.09	0.02	0.07	0	0.09
Atlantic N. Low Lat.	16.53	0.16	0.15	0.14	0.13	0.15	0.15	0.09
Atlantic N. Tropics	14.17	-0.10	-0.11	-0.11	-0.13	-0.09	-0.09	-0.03
Atlantic S. Tropics	10.23	-0.06	-0.09	-0.10	-0.06	-0.10	-0.09	-0.10
Atlantic S. Low Lat.	10.23	0	0	0	0	0	0	-0.01
Atlantic S. Mid Lat.	9.45	0.23	0.14	0.13	0.19	0.09	0.21	0.05
Pacific N. High Lat.	12.20	-0.02	-0.01	0.0	-0.004	-0.02	-0.01	0.01
Pacific N. Mid Lat.	37.78	0.28	0.27	0.28	0.29	0.26	0.25	0.20
Pacific N. Tropics	38.96	-0.28	-0.30	-0.32	-0.35	-0.30	-0.29	-0.18
Pacific S. Tropics	28.33	-0.26	-0.26	-0.26	-0.20	-0.25	-0.24	-0.44
Pacific S. Mid Lat.	39.36	0.45	0.57	0.60	0.46	0.44	0.48	0.47
Indian Tropics	31.88	-0.10	-0.11	-0.12	-0.10	-0.10	-0.11	-0.11
Indian S. Mid Lat.	24.01	0.22	0.25	0.27	0.21	0.17	0.26	0.11
Polar and Subpolar Ocean	58.64	-0.53	-0.72	-0.83	-0.47	-0.46	-0.72	-0.32
Global		0.19	0.20	0.20	0.20	-0.02	-0.01	-0.03
RMSE $\Delta C_{gasex} \; (\mu \text{mol } l^{-1})$		17.82	15.29	14.15	16.94	17.27	16.86	18.7

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Table 3. Regional net sea-to-air fluxes in GtC yr⁻¹ of natural, anthropogenic and contemporary CO₂ inferred with four different reconstructions of anthropogenic carbon and the ΔC_{gasex} tracer. The contemporary air-sea flux is calculated by adding the river-derived outgassing [Jacobson et al., 2007] to the natural and anthropogenic flux. The skill scores and the RMSE for

 C_{anth} are taken from Gerber et al. [2008].

Region	River Derived		ΔC^*			TTD			TrOCA			CFC	
	Outgassing	C_{anth}	Natural	Total	C_{anth}	Natural	Total	C_{anth}	Natural	Total	C_{anth}	Natural	Total
Atlantic N.High Lat.	-0.03 ± 0.01	0.12	0.18	0.27	0.09	0.16	0.22	0.17	0.16	0.30	0.16	0.11	0.24
Atlantic N. Mid Lat.	-0.01 ± 0.01	0.11	0	0.10	0.08	0.03	0.10	0.15	0	0.14	0.07	0.06	0.12
Atlantic N. Low Lat.	-0.04 ± 0.02	0.04	0.15	0.15	0.06	0.16	0.18	0.01	0.17	0.14	0.06	0.15	0.17
Atlantic N. Tropics	-0.09 ± 0.05	0.03	-0.09	-0.15	0.04	-0.13	-0.18	0.02	-0.10	-0.17	0.03	-0.13	-0.19
Atlantic S. Tropics	-0.02 ± 0.01	0.09	-0.09	-0.02	0.10	-0.09	-0.01	0.15	-0.08	0.05	0.12	-0.10	0
Atlantic S. Low Lat.	0 ± 0	0	0	0	0	0.01	0.01	0.01	-0.01	0	0	0.01	0.01
Atlantic S. Mid Lat.	-0.01 ± 0	0.08	0.21	0.28	0.07	0.09	0.15	-0.01	0.22	0.20	0.08	0.08	0.15
Pacific N. High Lat.	-0.03 ± 0.01	0.05	-0.01	0.01	0.03	0	0	0.10	0	0.07	0.03	0.02	0.02
Pacific N. Mid Lat.	-0.05 ± 0.02	0.14	0.25	0.34	0.17	0.23	0.35	0.08	0.29	0.32	0.21	0.23	0.41
Pacific N. Tropics	-0.03 ± 0.01	0.15	-0.29	-0.17	0.21	-0.29	-0.11	0.15	-0.33	-0.21	0.20	-0.30	-0.13
Pacific S. Tropics	-0.01 ± 0.01	0.07	-0.24	-0.18	0.16	-0.25	-0.10	0.09	-0.24	-0.16	0.16	-0.27	-0.12
Pacific S. Mid Lat.	0 ± 0	0.14	0.48	0.62	0.05	0.50	0.55	0.12	0.58	0.70	0.06	0.54	0.60
Indian Tropics	-0.09 ± 0.04	0.05	-0.11	-0.15	0.06	-0.11	-0.14	0.10	-0.16	-0.15	0.07	-0.11	-0.13
Indian S. Mid Lat.	-0.01 ± 0.00	0.16	0.26	0.41	0.16	0.23	0.38	-0.03	0.46	0.42	0.17	0.24	0.40
Polar and Subpolar Ocean	-0.01 ± 0.00	0.72	-0.72	-0.01	0.75	-0.54	0.20	1.09	-0.98	0.10	0.88	-0.54	0.33
Global	-0.41 \pm 0.21	1.95	-0.01	1.53	2.04	0	1.62	2.20	-0.02	1.77	2.30	-0.01	1.88
Skill score		0.839			0.870			0.760			0.686		
RMSE $\Delta C_{qasex} \; (\mu \text{mol } l^{-1})$			16.86			16.39			19.44			17.21	
RMSE $C_{anth} \; (\mu \text{mol } l^{-1})$		7.20			6.76			9.85			13.37		

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Figure 1. Contemporary net air-to-sea fluxes of CO_2 for aggregated regions as inferred by assimilating ocean tracer data into the Bern3D transport model (red). Error bars represent ± 1 standard deviation and include uncertainties from input data, ocean transport, and riverderived outgassing. Results obtained by the Green's function ocean inversion of *Mikaloff Fletcher et al.* [2006, 2007] (black) and from a climatology of the air-sea CO_2 partial pressure difference [*Takahashi et al*, 2008] as published by *Gruber et al.* [2009] (blue) are shown for comparison.



Figure 2. Annual mean net air-to-sea flux (mol m⁻²) of CO₂ for the anthropogenic perturbation D R A F T March 27, 2009, 1:39pm D R A F T (left), the preindustrial (middle), and the present (1995, right) as inferred by assimilating four different reconstructions of anthropogenic carbon and of ΔC_{gasex} based on the ΔC^* , the TTD, the TrOCA, and the CFC methods into the Bern3D model.



Figure 3. Zonal average of data-based and modeled distribution of ΔC_{gasex} and of the corresponding residuals (data minus model) in μ mol l⁻¹ for the Atlantic, Pacific, and Indian.



Figure 4. Zonal average of data-based ΔC_{gasex} in μ mol l^{-1} in the upper Pacific for four reconstruction methods.

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Figure 5. Regional net air-to-sea fluxes in GtC yr⁻¹ for four different reconstruction methods compared to the estimates of *Mikaloff Fletcher et al.* [2007]. The error bar shows the weighted standard deviation of the models participating in *Mikaloff Fletcher et al.* [2007].



Figure 6. Meridional ocean carbon transport for the (a) preindustrial, (b) the anthropogenic perturbation, and (c) the present for the global ocean (blue), the Atlantic (black) and the Indo-Pacific (red). Rates are inferred by assimilating four different reconstructions of C_{anth} and ΔC_{gasex} into the standard version of the Bern3D model. Solid lines show best estimates and the shading the spread from the different input data.

- ¹ Carbon sources and sinks from an Ensemble Kalman
- ² Filter ocean data assimilation. Supplemental material

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Table 1. Supplement material Table S1: Meridional transport rates in GtC yr^{-1} for the different data-based estimates. A positive sign indicates a southwards transport. The flux is across the southern border of each region. The values in parentheses represent the net eastward flux across the eastern border of the box. A negative sign represents a westward flux.

Region	Meridional transport in $GtC yr^{-1}$						
	TTD	CFC	TrOCA	ΔC^*			
Atlantic N.High Lat. (46 N)	0.16	0.11	0.16	0.18			
Atlantic N. Mid Lat. (34 N)	0.19	0.17	0.17	0.18			
Atlantic N. Low Lat. (16 N)	0.35	0.32	0.34	0.33			
Atlantic N. Tropics (3 S)	0.22	0.19	0.23	0.24			
Atlantic S. Tropics (19 S)	0.13	0.10	0.16	0.16			
Atlantic S. Low Lat. (34 S)	0.14	0.10	0.15	0.16			
Atlantic S. Mid Lat. (46 S)	0.21(0.02)	0.17(0.01)	0.35(0.01)	0.35(0.02)			
Pacific N. High Lat. (51 N)	0	0.02	0	-0.02			
Pacific N. Mid Lat. (16 N)	0.24	0.25	0.29	0.24			
Pacific N. Tropics (3 S)	-0.05	-0.05	-0.04	-0.05			
Pacific S. Tropics (19 S)	-0.31	-0.33	-0.28	-0.30			
Pacific S. Mid Lat. (46 S)	0.19	0.22	0.30	0.19			
Indian Tropics (19 S)	-0.11	-0.11	-0.15	-0.10			
Indian S. Mid Lat. (42 S)	0.14	0.15	0.32	0.18			





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Figure 2. Supplement material Figure S2: Global streamfunction and Global Overturning for

the Standard, ACC \times 3 and PSI \times 3 Bern3D model setup.



Figure 3. Supplement material Figure S3: Taylor diagram for the different Bern3D model setups and the different data-based reconstructions compared with the pCO_2 based estimates of *Takahashi et al* [2008]. The red symbols are the different data-based simulations, the black ones represent the different Bern3D model setups.