



## **An Assessment of CO<sub>2</sub> Storage and Sea-Air Fluxes for the Atlantic Ocean and Mediterranean Sea Between 1985 and 2018**

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# Global Biogeochemical Cycles<sup>\*</sup>

## RESEARCH ARTICLE

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### Special Section:

Regional Carbon Cycle Assessment and Processes - 2

### Key Points:

- From 1985 to 2018, pCO<sub>2</sub> products suggest a lower mean CO<sub>2</sub> uptake ( $-0.36 \pm 0.06$  PgC yr<sup>-1</sup>) than ocean models ( $-0.47 \pm 0.15$  PgC yr<sup>-1</sup>)
- Since 2000, the CO<sub>2</sub> uptake is increasing twice as fast in the pCO<sub>2</sub> products compared to the models
- Major differences between models and pCO<sub>2</sub> products are attributed to the outgassing of riverine carbon and the seasonal cycle of pCO<sub>2</sub>

### Supporting Information:

Supporting Information may be found in the online version of this article.

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
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## An Assessment of CO<sub>2</sub> Storage and Sea-Air Fluxes for the Atlantic Ocean and Mediterranean Sea Between 1985 and 2018

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**Abstract** As part of the second phase of the Regional Carbon Cycle Assessment and Processes project (RECCAP2), we present an assessment of the carbon cycle of the Atlantic Ocean, including the Mediterranean Sea, between 1985 and 2018 using global ocean biogeochemical models (GOBMs) and estimates based on surface ocean carbon dioxide (CO<sub>2</sub>) partial pressure (pCO<sub>2</sub> products) and ocean interior dissolved inorganic carbon observations. Estimates of the basin-wide long-term mean net annual CO<sub>2</sub> uptake based on GOBMs and pCO<sub>2</sub> products are in reasonable agreement ( $-0.47 \pm 0.15$  PgC yr<sup>-1</sup> and  $-0.36 \pm 0.06$  PgC yr<sup>-1</sup>, respectively), with the higher uptake in the GOBM-based estimates likely being a consequence of a deficit in the representation of natural outgassing of land derived carbon. In the GOBMs, the CO<sub>2</sub> uptake increases with time at rates close to what one would expect from the atmospheric CO<sub>2</sub> increase, but pCO<sub>2</sub> products estimate a rate twice as fast. The largest disagreement in the CO<sub>2</sub> flux between GOBMs and pCO<sub>2</sub> products is found north of 50°N, coinciding with the largest disagreement in the seasonal cycle and interannual variability. The mean accumulation rate of anthropogenic CO<sub>2</sub> (C<sub>ant</sub>) over 1994–2007 in the Atlantic Ocean is  $0.52 \pm 0.11$  PgC yr<sup>-1</sup> according to the GOBMs, 28%  $\pm$  20% lower than that derived from observations. Around 70% of this C<sub>ant</sub> is taken up from the atmosphere, while the remainder is imported from the Southern Ocean through lateral transport.

**Plain Language Summary** This study contributes to the second Regional Carbon Cycle Assessment and Processes project by presenting a carbon cycle evaluation of the Atlantic Ocean including the Mediterranean Sea between 1985 and 2018. The assessment draws on output from global ocean biogeochemical models along with estimates based on observations of surface ocean carbon dioxide (CO<sub>2</sub>) partial pressure (pCO<sub>2</sub> products) and ocean interior dissolved inorganic carbon. The models suggest that the Atlantic took up  $-0.47 \pm 0.15$  Pg of carbon per year, in reasonable agreement with an uptake of  $-0.36 \pm 0.06$  Pg carbon per year computed from pCO<sub>2</sub> products. In the models, the rate of CO<sub>2</sub> uptake is keeping pace with the increase in atmospheric CO<sub>2</sub>, but it is twice as fast in the pCO<sub>2</sub> products. Most of the uptake of CO<sub>2</sub> by the ocean occurs in response to excess CO<sub>2</sub> released into the atmosphere from human activities. The so-called anthropogenic carbon accumulates in the Atlantic Ocean at a rate of  $0.52 \pm 0.11$  Pg carbon per year according to the models. This estimate is 28%  $\pm$  20% lower than that derived from observations. Further investigation reveals that about 70% of the accumulated anthropogenic carbon is taken up from the atmosphere, while the remainder is imported from the Southern Ocean.

## 1. Introduction

During the International Geophysical Year 1957–1958, Taro Takahashi (1930–2019) made the first systematic and accurate measurements of carbon dioxide gas (CO<sub>2</sub>) partial pressure in the air and sea surface along an

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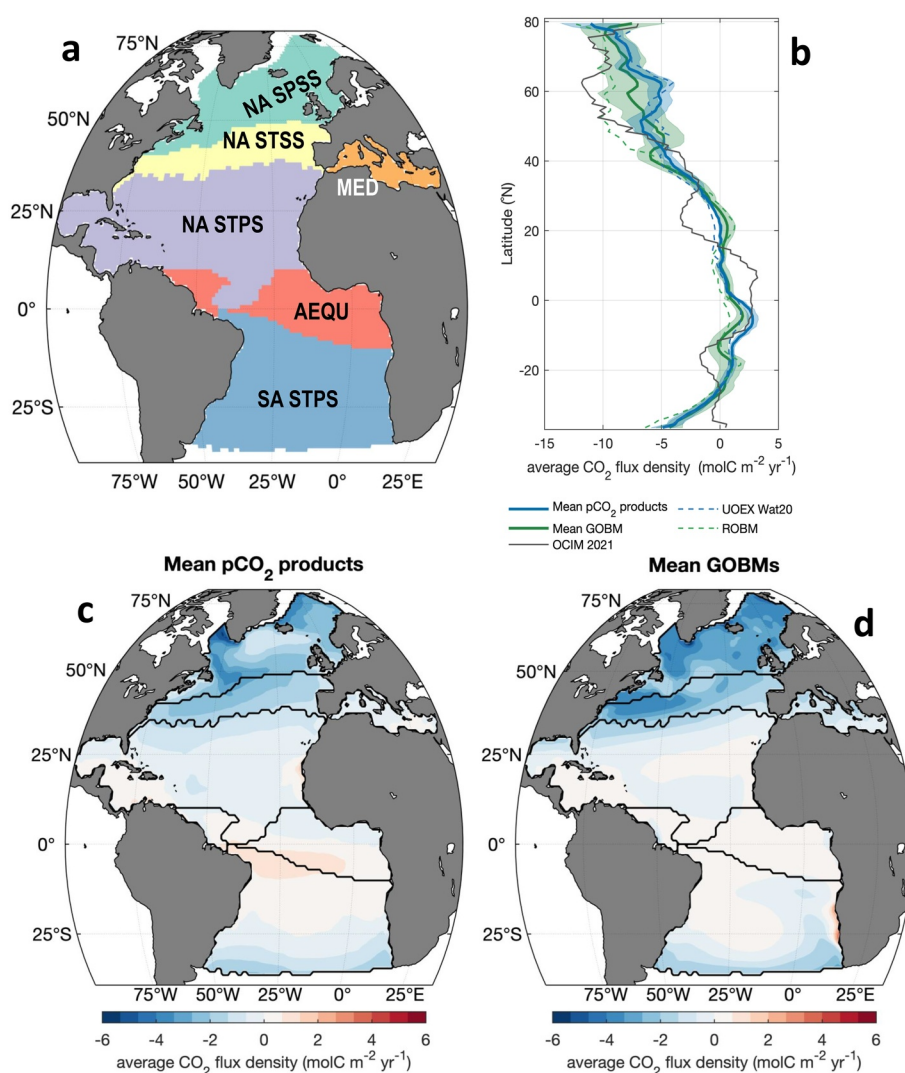
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Atlantic Ocean transect from Greenland to Cape Town (Takahashi, 1961). Since these early times, the importance of monitoring seawater CO<sub>2</sub> partial pressure (pCO<sub>2</sub>) for the assessment of sea-air exchanges of CO<sub>2</sub> has been increasingly recognized. Today, measurements of pCO<sub>2</sub> have become an integral part of ocean monitoring programs including Eulerian time series stations (Bates et al., 2014), oceanographic buoy arrays, and Ships of Opportunity (SOOP) programs (Bakker et al., 2016; Pfeil et al., 2013; Sabine et al., 2013; Wanninkhof et al., 2019). Early measurements of pCO<sub>2</sub> highlighted spatial patterns that were confirmed later by time-series measurements (Bates et al., 1998; Gruber et al., 1998; Keeling, 1993; Michaels et al., 1994), large-scale data compilations and the development of surface ocean CO<sub>2</sub> climatology (Takahashi et al., 2002, 2009). The North Atlantic between 25°N and 76°N stands out as a region of intense CO<sub>2</sub> uptake by the ocean. It represents only 7% of the ocean surface, but accounts for 23% of the global uptake (Schuster et al., 2013; Takahashi et al., 2009). Approximately two thirds of its contemporary uptake is caused by natural processes, such as heat loss and export production, while the remaining one third is caused by the increasing concentrations of CO<sub>2</sub> in the atmosphere and is therefore called uptake of anthropogenic carbon, C<sub>ant</sub> (Gruber et al., 2009; Keeling & Peng, 1995; Mikaloff Fletcher et al., 2007; Watson et al., 1995). The local uptake of C<sub>ant</sub> by sea-air exchange in the Atlantic combined with the net northward transport of C<sub>ant</sub>-rich southern latitude waters by the upper limb of the Atlantic meridional overturning circulation (AMOC) (Brown et al., 2021; MacDonald et al., 2003; Pérez et al., 2013; Rosón et al., 2003) leads to a high accumulation of C<sub>ant</sub> throughout the water column of the Atlantic, accounting for approximately 35% of the global total storage (Gruber et al., 2019; Sabine et al., 2004). Earlier studies highlighted the role of the AMOC, a key component of the global ocean circulation and a distinctive dynamic element of the Atlantic circulation, in the redistribution of CO<sub>2</sub> (Holfort et al., 1998; Wallace, 2001). The AMOC further links the upper ocean thermohaline circulation with the intense Deep Western Boundary Current (DWBC) connecting the waters formed in the subpolar North Atlantic with the Southern Ocean (Haine, 2016; Hirschi et al., 2020; Rhein et al., 2015). The DWBC contributes to natural interhemispheric carbon exchanges by transporting between 0.5 and 1 PgC yr<sup>-1</sup> from North Atlantic uptake regions southward (Aumont et al., 2001; Macdonald et al., 2003; Resplandy et al., 2018).

As part of the second phase of the Regional Carbon Cycle Assessment and Processes project (RECCAP2), we complement these earlier studies about the Atlantic carbon budget with an analysis of the latest observation- and model-based estimates of the Atlantic Ocean including sea-air fluxes (natural and anthropogenic), storage, and transport of CO<sub>2</sub> for the years 1985–2018. Following Fay and McKinley (2014), RECCAP2 divides the Atlantic into five regions or biomes (Figure 1a), namely the North Atlantic subpolar gyre (NA SPSS), the seasonally and permanently stratified regions of the North Atlantic subtropical gyre (NA STSS and NA STPS), the Atlantic equatorial upwelling region (AEQU), and the permanently stratified South Atlantic subtropical gyre extending southward to ~35°S (SA STPS). The Mediterranean Sea is also included as a single, sixth biome (MED). Among these regions, the NA SPSS stands out as a biome with high spatial and temporal variability, which still challenges our understanding, assessments, and modeling efforts despite the increase in observational capacity over the last two decades. During the RECCAP1 period (1990–2009), Schuster et al. (2013) estimated an average CO<sub>2</sub> uptake of  $-0.21 \pm 0.06$  PgC yr<sup>-1</sup> (*positive sign indicating flux into the atmosphere (outgassing), and negative sign a flux into the ocean (uptake)*) between 49°N and 79°N, consistent across observation-based estimates and numerical models used. This flux amounts to 10% of the global uptake and makes the NA subpolar region one of the regions with the highest CO<sub>2</sub> uptake density (see Text S1 in Supporting Information S2). Understanding the seasonal, interannual and long-term variability of the high latitude Atlantic CO<sub>2</sub> sink has been the focus of many observational and modeling studies (e.g., Breeden & McKinley, 2016; Goris et al., 2015; Leseurre et al., 2020; Macovei et al., 2020; Thomas et al., 2008; Tjiputra et al., 2012; Ullman et al., 2009; Watson et al., 2009). It has become clear that the variability of sea-air fluxes of CO<sub>2</sub> and C<sub>ant</sub> storage rates in this region is influenced by regional modes of climate variability, such as the North Atlantic Oscillation (NAO), through its effect on wind patterns and ocean heat loss, mixing, and deep water formation. During the time period from the 1990s to the 2000s, C<sub>ant</sub> storage rates decreased in the subpolar NA in response to the shift from predominantly high (1990–1995) to low (2002–2007) NAO (Gruber et al., 2019; Pérez et al., 2008, 2013; Steinfeldt et al., 2009). As part of their global assessment, Müller et al. (2023) found that only the North Atlantic exhibited a trend toward weaker C<sub>ant</sub> accumulation relative to the atmospheric CO<sub>2</sub> increase by comparing their inventory changes from 2004 to 2014 and from 1994 to 2004 with previous estimates for the period 1800 to 1994 from Sabine et al. (2004). However, recent observations show a reinvigoration of the C<sub>ant</sub> accumulation at the local scale associated with increased convection in the mid 2010s (Fröb et al., 2016), as a consequence of a shift back to positive NAO conditions.



**Figure 1.** (a) RECCAP2 biomes in the Atlantic including the Mediterranean Sea. (b) Latitudinal variation of CO<sub>2</sub> flux densities displayed for the ensemble mean of the pCO<sub>2</sub> products, the GOBM ensemble mean, the UOEX data product that corrects for skin temperature effects, the regional hindcast model (ROBM), and the inverse model OCIMv2021. Average CO<sub>2</sub> flux density from 1985 to 2018, illustrated on maps for the ensemble means of (c) nine pCO<sub>2</sub> products and (d) 11 GOBMs. Negative values indicate oceanic uptake of CO<sub>2</sub>. The biomes are the seasonally stratified North Atlantic subpolar gyre (NA SPSS), the seasonally and permanently stratified regions of the North Atlantic subtropical gyre (NA STSS and NA STPS), the Atlantic equatorial upwelling region (AEQU), the seasonally stratified South Atlantic subtropical gyre (SA STPS), and the Mediterranean Sea (MED). Note that the GOBMs do not adequately represent the RCO (Riverine CO<sub>2</sub> outgassing) fluxes and thus we did not adjust those with other available estimates.

The subtropical North Atlantic (in RECCAP1 defined to be 18° to 49°N, 7.2% of the ocean surface, see Text S1 in Supporting Information S2) was shown to be a CO<sub>2</sub> sink, with a net uptake of  $-0.26 \pm 0.06$  PgC yr<sup>-1</sup> between 1990 and 2009 (Schuster et al., 2013), comprising approximately equal parts to C<sub>ant</sub> and natural CO<sub>2</sub> uptake, where the latter is driven mainly by net heat loss, with limited contributions from biological activity (Gruber et al., 2009). The mean subtropical gyre uptake rate ( $-0.91$  mol C m<sup>-2</sup> yr<sup>-1</sup>) is similar to that observed in the Bermuda Atlantic Time-series Study (Bates et al., 2014), even though the eastern return branch of the subtropical gyre showed lower uptake values (Santana-Casiano et al., 2009). At both sites, the interannual variability of CO<sub>2</sub> flux correlates with sea surface temperature (SST) and mixed layer depth anomalies (González-Dávila et al., 2010; Gruber et al., 2002; Santana-Casiano et al., 2009). SST is the main driver of the seasonal cycle in the subtropics, driving an outgassing of CO<sub>2</sub> in summer and uptake in winter.



The tropical Atlantic is the second largest oceanic source of CO<sub>2</sub> to the atmosphere, after the tropical Pacific, with an annual emission of 0.10–0.11 PgC yr<sup>−1</sup> (Landschützer et al., 2014; Takahashi et al., 2009) due to frequent upwelling of cold, CO<sub>2</sub>-rich water in the eastern parts. Based on six different methodologies, the RECCAP1 estimate for this region converged on an outgassing of  $0.12 \pm 0.04$  PgC yr<sup>−1</sup> between 1990 and 2009 (Schuster et al., 2013; see Text S1 in Supporting Information S2). The increase in atmospheric CO<sub>2</sub> has decreased the net outgassing since preindustrial times, as the ocean supersaturation is reduced by about 50% (Gruber et al., 2009). This implies an uptake of anthropogenic CO<sub>2</sub>. Gruber et al. (2009) also suggested that an important part of this natural outgassing is due to the riverine contribution of organic matter, especially that stemming from the Amazon river (Louchard et al., 2021).

The subtropical South Atlantic is a sink for atmospheric CO<sub>2</sub> (Rödenbeck et al., 2015; Schuster et al., 2013), driven in almost equal parts by natural and anthropogenic CO<sub>2</sub> fluxes (Gruber et al., 2009). It has been suggested that strong upwelling events in the eastern part generate significant interannual variability (Schuster et al., 2013; see Text S1 in Supporting Information S2). However, pCO<sub>2</sub> variability in the SA STPS biome is relatively low, as shown by Rödenbeck et al. (2015). From 1990 to 2009, this region was a CO<sub>2</sub> sink of  $-0.14 \pm 0.04$  PgC yr<sup>−1</sup> on average, combining areas with a net outgassing north of the 23°C isotherm (Ito et al., 2005) with areas of absorption to the south. This region is relatively poorly sampled, with the domain north of 31°S acting as a source in spring and sinking in autumn (González-Dávila et al., 2009; Padín et al., 2010; Santana-Casiano et al., 2007). Estimates of long-term CO<sub>2</sub> flux trends in this region are highly dependent on the methodology used (Schuster et al., 2013).

The Mediterranean Sea represents 3.5% of the Atlantic Ocean area and is the only mid-latitude ocean basin in which deep convection occurs (see Text S1 in Supporting Information S2). This circulation is responsible for a relatively large inventory of C<sub>ant</sub> of 1.7 PgC in 2001 as estimated from CFCs (Schneider et al., 2010). The overturning time is fast in relation to that of the global ocean (60–220 years vs. more than 1,000 years; Khatiwala et al., 2013; Stöven & Tanhua, 2014) and allows a complete renewal of water in the basin on a centennial time scale. Hence, surface waters enriched in C<sub>ant</sub> transfer this signature to deep layers relatively quickly, leading to all water masses in the basin being already invaded by C<sub>ant</sub> (Hassoun et al., 2015; Touratier et al., 2016). However, surface pCO<sub>2</sub> exhibits large variability, due to the large heterogeneity of physical and trophic regimes in the two main Mediterranean sub-basins, with a marked west-to-east oligotrophy gradient and different atmospheric forcings that regulate seawater pCO<sub>2</sub> and the sea-air CO<sub>2</sub> exchanges (Coppola et al., 2018; De Carlo et al., 2013; Ingrosso et al., 2016; Kapsenberg et al., 2017; Krasakopoulos et al., 2009, 2017; Petihakis et al., 2018; Sisma-Ventura et al., 2017; Urbini et al., 2020; Wimart-Rousseau et al., 2021).

In RECCAP1, the assessment of the ocean carbon cycle relied on five global ocean biogeochemical models (GOBMs), several atmospheric and oceanic inversions, the pCO<sub>2</sub> climatology published by Takahashi et al. (2009), as well as the SOCAT (Surface Ocean CO<sub>2</sub> Atlas) database. A crucial progress since RECCAP1 is the annual update of SOCAT (Bakker et al., 2016), with over 33.7 million quality-controlled surface ocean pCO<sub>2</sub> measurements in the 2022 release (Bakker et al., 2022). The availability of these data sparked the development of time-varying reconstructions of surface ocean pCO<sub>2</sub> distributions. These pCO<sub>2</sub> products rely on advanced statistical techniques and neural networks to extrapolate sparse observations in time and space to achieve temporally resolved global coverage (e.g., Chau et al., 2022; Gregor et al., 2019; Landschützer et al., 2014; Rödenbeck et al., 2014). Similarly, advances in biogeochemical modeling since RECCAP1 led to the contribution of an increased number of GOBMs that provided output from up to four different simulations allowing to disentangle the natural carbon cycle and the anthropogenic perturbation (Friedlingstein et al., 2022; Wanninkhof et al., 2013).

Improved process understanding and increasing availability of ocean biogeochemical data have led to advances in GOBMs, particularly in simulating the large-scale features and mean state of the ocean carbon cycle (Séférian et al., 2020). When forced with atmospheric reanalysis and atmospheric CO<sub>2</sub> concentration data, these models were assessed to be suitable for quantifying the global ocean carbon fluxes, from annual mean to interannual time-scale (Hauck et al., 2020). Regionally, such models have also been shown to be capable of simulating the observed long-term pCO<sub>2</sub> trends (Tjiputra et al., 2014). Nevertheless, some GOBMs still have difficulties in representing the observed seasonal cycle in key ocean sink regions in the North Atlantic, likely owing to mismatch in the timing of deep winter mixing and/or biological bloom events (Schwinger et al., 2016; Tjiputra et al., 2012). Since RECCAP1, the number of GOBMs has increased from 6 to 11 in RECCAP2, and while not all RECCAP1 models participated in the RECCAP2 exercise, those that did have likely gone through iterations of

improvements (the readers are referred to Table S1 in DeVries et al., 2023, for individual biogeochemical model descriptions). In the Atlantic domain, recent developments in the ocean physical component have led to better representation of large scale circulation and ventilation processes (Hirschi et al., 2020), which could have strong implications on the transports of biogeochemical tracers driving the sea-air CO<sub>2</sub> fluxes and interior carbon sequestration in this basin.

This synthesis paper is structured as follows. Section 2 provides the details of the database consisting of data sets based on observations of both surface pCO<sub>2</sub> and the marine carbonate system in the ocean interior, and an ensemble of global biogeochemical models together with a regional model and an assimilation model. In Section 3, the CO<sub>2</sub> fluxes obtained for each class of products are described and analyzed considering both the mean values for the study period, trends in two periods (1985–2000 and 2000–2018), the seasonal cycle and the interannual variability of CO<sub>2</sub> fluxes. In addition, the accumulation of anthropogenic CO<sub>2</sub> in the ocean interior is evaluated. In Section 4, we discuss the results obtained in RECCAP2 in comparison to RECCAP1, as well as the consistency and discrepancies between the global biogeochemical models and different data products, and suggest ways for future improvements. Section 5 summarizes the main conclusions and lists some of the remaining challenges to be solved in future versions of RECCAP.

## 2. Methods

The Atlantic Ocean and its subdivision into biomes is defined by the RECCAP2 basin mask (Müller, 2023) that builds on the biome definition by Fay and McKinley (2014) and extends from approximately 79°N to approximately 35°S (Figure 1). The RECCAP2 ocean database used in this study is described by DeVries et al. (2023), consisting of observation-based and model-based products. Here, we use two types of observation-based products, namely surface ocean pCO<sub>2</sub> (pCO<sub>2</sub> products) and ocean interior C<sub>ant</sub> reconstructions. For model-based products, we use Global and Regional Ocean Biogeochemical Model (GOBM/ROBM) hindcast simulations and an ocean data-assimilation model. All products were re-gridded onto a common 1° × 1° horizontal grid and monthly temporal resolution by the data providers, except for ocean interior model outputs, which were submitted as annual averages. Ocean model outputs were either provided on the models' standard depth levels or re-gridded to fixed depth levels chosen by the data providers.

### 2.1. Observation-Based Products

#### 2.1.1. pCO<sub>2</sub> Products

This analysis draws on a variety of observation-based products for surface ocean pCO<sub>2</sub> and sea-air CO<sub>2</sub> fluxes (Table S1 in Supporting Information S2). These products are based on the interpolation of in situ pCO<sub>2</sub> data accessed from different releases (v2019–v2021, v5) of SOCAT (Bakker et al., 2016) to near-global coverage. Several interpolation methods are used including machine learning techniques (Chau et al., 2022; Gloege et al., 2021; Gregor & Gruber, 2021; Gregor et al., 2019; Iida et al., 2021; Landschützer et al., 2014; Watson et al., 2020; Zeng et al., 2022) and a diagnostic mixed layer scheme (Rödenbeck et al., 2013). Sea-air CO<sub>2</sub> fluxes (FCO<sub>2</sub>) are computed from reconstructed pCO<sub>2</sub> fields following:

$$FCO_2 = Kw (1 - f_{ice}) K_0 (pCO_2 - pCO_{2,air}) \quad (1)$$

where Kw is gas transfer velocity;  $f_{ice}$  is sea-ice cover fraction;  $K_0$  is CO<sub>2</sub> solubility in seawater; and pCO<sub>2</sub>, and pCO<sub>2, air</sub> are the partial pressures of CO<sub>2</sub> in seawater (nominally at 5 m depth) and in the overlying atmosphere, respectively. The gas transfer velocity is computed as a function of wind speed at 10 m, mostly assuming a quadratic relationship (Ho et al., 2006; Nightingale et al., 2000; Wanninkhof, 1992, 2014). For the set of pCO<sub>2</sub> products, the uncertainty of the mean is determined as the standard deviation of the FCO<sub>2</sub> of the nine pCO<sub>2</sub> products referenced in Table S1 of the Supporting Information S2.

The pCO<sub>2</sub> product by Watson et al. (2020), UOEX-Wat20, is different from the other products as it adjusts the underlying pCO<sub>2</sub> observations accounting for the cool-skin effect and for near-surface temperature gradients following Goddijn-Murphy et al. (2015) and Woolf et al. (2016), henceforth referred to as the surface skin effects. While it applies the SOMFFN interpolation approach also used in MPI-SOMFFN, it does so in different fashion such that the differences between the UOEX-Wat20 and other approaches are not solely attributed to adjusting the

pCO<sub>2</sub> values. While UOEX-Wat20 is included in the analysis, it is kept distinct from the other nine pCO<sub>2</sub> products, because of the difference in approach.

The pCO<sub>2</sub> products all use the bulk flux parameterization (Equation 1) and aside from UOEX-Wat20 follow the convention of reference depth for pCO<sub>2</sub> at nominally 5-m. The uncertainty estimates provided here are mainly based on differences between the different products. Uncertainties and biases in gas transfer velocities, and impacts of near-surface pCO<sub>2</sub> gradients (Bellenger et al., 2023; Dong et al., 2022) are not taken into account but are estimated to increase the uncertainty in the pCO<sub>2</sub> products by 3-fold on global scales (see Table 3, DeVries et al., 2023).

### 2.1.2. Ocean Interior C<sub>ant</sub> Reconstructions

Furthermore, we consider two ocean interior observation-based products, one based on measurements of dissolved inorganic carbon (DIC) concentrations collected over more than 30 years, and other physical and biogeochemical parameters by Gruber et al. (2019), and another one combining an inversion approach with tracer measurements by Khatiwala et al. (2009). The Gruber et al. (2019) product provides an estimate of the ocean C<sub>ant</sub> storage change ( $\Delta C_{ant}$ ) between the years 1994 and 2007. This estimate is based on the eMLR(C\*) method (Clement & Gruber, 2018) applied to the GLODAPv2 data (Olsen et al., 2016). It includes estimates from surface to 3,000 m depth for both the steady-state and non-steady-state components of  $\Delta C_{ant}$  in the ocean interior. In the North Atlantic and below 3,000 m, Gruber et al. (2019) estimated an inventory change C<sub>ant</sub> of 0.05 PgC yr<sup>-1</sup> (~8% of the accumulation above 3,000 m), which has been proportionally distributed across biomes according to the GOBM  $\Delta C_{ant}$  below 3,000 m. The product from Khatiwala et al. (2009) provides estimates of the increase of the oceanic C<sub>ant</sub> content from 1850 up to 2011 and is based on a Green's Function approach that allows a gradual increase in the CO<sub>2</sub> disequilibrium between the atmosphere and the ocean.

The C<sub>ant</sub> reconstruction product from Khatiwala et al. (2009) was pre-processed to match the RECCAP2 1° × 1° grid and depth levels of the  $\Delta C_{ant}$  reconstruction from Gruber et al. (2019). Since the product provides annual values, we calculated  $\Delta C_{ant}$  between 1994 and 2007 to allow for comparison with Gruber et al. (2019). The  $\Delta C_{ant}$  reconstruction of Gruber et al. (2019) does not cover the entire NA SPSS biome explored in our study, so we extrapolated the product to the Nordic Seas assuming the same vertical  $\Delta C_{ant}$  profile as at 65°N, resulting in a 23% increase of  $\Delta C_{ant}$  storage rate in the NA SPSS biome. The percentage of increase obtained was also applied to Khatiwala et al. (2009), as it also does not fully cover the NA SPSS biome. The uncertainty in the C<sub>ant</sub> inventory increase in each biome was estimated by surface area scaling of the uncertainties of the North and South Atlantic provided by Gruber et al. (2019). For the Khatiwala et al. (2009) product, a relative uncertainty of 17% was set following Khatiwala et al. (2013).

### 2.2. Global Ocean Biogeochemical Models

As an improvement from RECCAP1 (Wanninkhof et al., 2013), the RECCAP2 protocol provides a set-up of four simulations with four combinations of atmospheric physical and CO<sub>2</sub> concentration forcings such that the simulated total CO<sub>2</sub>-fluxes can be divided into their steady-state and non-steady state natural and anthropogenic components: (a) Simulation A: temporally varying atmospheric forcing and increasing atmospheric CO<sub>2</sub>, (b) Simulation B: climatological atmospheric forcing and constant pre-industrial atmospheric CO<sub>2</sub>, (c) Simulation C: climatological atmospheric forcing and increasing atmospheric CO<sub>2</sub>, and (d) Simulation D: temporally varying atmospheric forcing and constant pre-industrial CO<sub>2</sub>.

We used outputs from 11 GOBMs of which the majority also contributed to the Global Carbon Budget (Friedlingstein et al., 2022; Table S2 in Supporting Information S2). All GOBMs used here are general ocean circulation models with coupled ocean biogeochemistry, run in hindcast mode and hence forced by atmospheric data sets. Details of the respective model resolutions, forcings, and references are provided in an overview table in DeVries et al. (2023). All models performed four simulations (A, B, C, and D), except for MOM6-Princeton (not C and D). Additionally, we considered the output from the regional ocean biogeochemical model (ROBM) ROMS-AtlanticOcean-ETHZ (Louchard et al., 2021) that only performed Simulation A. We also included results from the ocean data-assimilation model OCIMv2021 (DeVries, 2022) that performed simulations A, B, and C. OCIMv2021 uses a climatological mean circulation but has a time-varying SST. It includes an abiotic carbon cycle model forced with atmospheric CO<sub>2</sub> to estimate the anthropogenic carbon distribution.

To determine the  $\text{FCO}_2$  for the period 1985–2018, for each GOBM and each biome, we subtracted the linear trend of the respective fluxes estimated in Simulation B from Simulation A, to correct for potential model-dependent drift. For the ensemble of GOBMs, the uncertainty of the mean was determined as the standard deviation of the 11 models referenced in Table S2 of the Supporting Information S2.

In order to be consistent with the  $\Delta C_{\text{ant}}$  reconstruction of Gruber et al. (2019), the  $C_{\text{ant}}$  accumulation rate in the GOBMs was evaluated between 1994 and 2007. Here,  $C_{\text{ant}}$  was calculated as the difference in DIC between the simulation with increasing atmospheric  $\text{CO}_2$  concentrations (Simulation A) and the one with constant pre-industrial atmospheric  $\text{CO}_2$  concentrations (Simulation D), both with time-varying atmospheric physical forcing. We considered all GOBMs that ran Simulation A and Simulation D (Table S2 in Supporting Information S2). However, MPIOM-HAMOCC was excluded from the final analysis because of its large negative  $C_{\text{ant}}$  values in the interior due to inconsistent physical forcing between its Simulations A and D. For the OCIMv2021 model,  $C_{\text{ant}}$  was determined as the difference between Simulations C (increasing atmospheric  $\text{CO}_2$ , climatological atmospheric forcing) and B (constant pre-industrial atmospheric  $\text{CO}_2$ , climatological atmospheric forcing), as this model uses a steady-state circulation and did not run Simulation D. Once we obtained the total  $C_{\text{ant}}$  concentrations from all GOBMs, we computed the  $C_{\text{ant}}$  storage changes as the difference between the concentrations in 1994 and 2007.  $C_{\text{ant}}$  concentration changes were vertically integrated to get the column inventory storage changes, as well as biome-integrated  $\Delta C_{\text{ant}}$  rates. For the GOBM ensemble, the uncertainty of the mean  $\Delta C_{\text{ant}}$  rate is determined as the standard deviation of the  $\Delta C_{\text{ant}}$  rates of the nine models referenced in Table S2 of the Supporting Information S2.

### 2.3. Area Coverage

Practically all  $\text{pCO}_2$  products considered have a spatial coverage of almost 100% of the Atlantic basin, except JMA-MLR and MPI-SOMFFN with about 91%–92% of the area coverage in the northernmost biome (NA SPSS). Here,  $\text{pCO}_2$  product fluxes were not scaled to the same ocean area, following the assumption of Hauck, Gregor, et al. (2023), that the discrepancy arising from differences in covered area are smaller than the uncertainty arising from any extrapolation to an equal area. All GOBMs cover more than 98% of the area, except MPIOM-HAMOCC (95%) and CESM-ETHZ (97%). ROMS-ETHZ (ROBM) covers 95% of the Atlantic Region, and only 93% of the NA SPSS biome and 25% of the Mediterranean Sea. Likewise, most of the missing coverage of the MPIOM-HAMOCC is located in the Mediterranean Sea, thus ROMS-ETHZ and MPIOM-HAMOCC have not been used for the evaluation of the MED biome.

### 2.4. Riverine Carbon Outgassing

The flux of natural  $\text{CO}_2$  across the sea-air interface also includes a flux balancing the input of inorganic and organic carbon at the land-sea interface minus the fraction buried in marine sediments (Regnier et al., 2013; Sarmiento & Sundquist, 1992). We refer to this flux component as preindustrial riverine  $\text{CO}_2$  outgassing (RCO). Since  $\text{pCO}_2$  products are based on real-world observations, they provide estimates of total  $\text{FCO}_2$ , including the RCO. In contrast, RCO is not at all or not adequately represented in GOBMs. Its approximation would require several thousands of years of integration with a GOBM including a sediment module. None of the GOBMs used here includes such a long pre-industrial spin-up (Terhaar et al., 2024). Though several of the GOBMs analyzed in this study include river inputs of carbon, not all processes relevant for the land-sea flux are adequately represented. In consequence, the average of the global imbalance between river input and flux to the sediment is small ( $<0.14 \text{ PgC yr}^{-1}$ ) in the GOBM ensemble (Terhaar et al., 2024) compared to the observation-based global integral of RCO recommended in the RECCAP2 protocol, that amounts to  $0.65 \pm 0.3 \text{ PgC yr}^{-1}$  (Regnier et al., 2022). Combining the spatial distribution of RCO by Lacroix et al. (2020) and the globally integrated estimate by Regnier et al. (2022) allows us, in principle, to estimate its contribution to  $\text{FCO}_2$  at biome scale, albeit with a relative uncertainty that is most likely even larger than that of the global integral ( $>50\%$  of the absolute value) and without considering the already-present land-sea fluxes of the GOBMs. However, the magnitude of RCO is a major source of uncertainty and hinders the straightforward comparison of fluxes from  $\text{pCO}_2$  products and GOBMs. In our analysis, we chose not to add the estimated RCO to the GOBMs but to present it separately, whenever this is meaningful. As the RCO spatial distribution by Lacroix et al. (2020) is uncertain, we do not evaluate it on a grid scale but only at the biome scale.



### 3. Results

#### 3.1. Sea-Air CO<sub>2</sub> Fluxes

##### 3.1.1. Long-Term Mean Fluxes From 1985 to 2018: Spatial Patterns and Regional Integrals

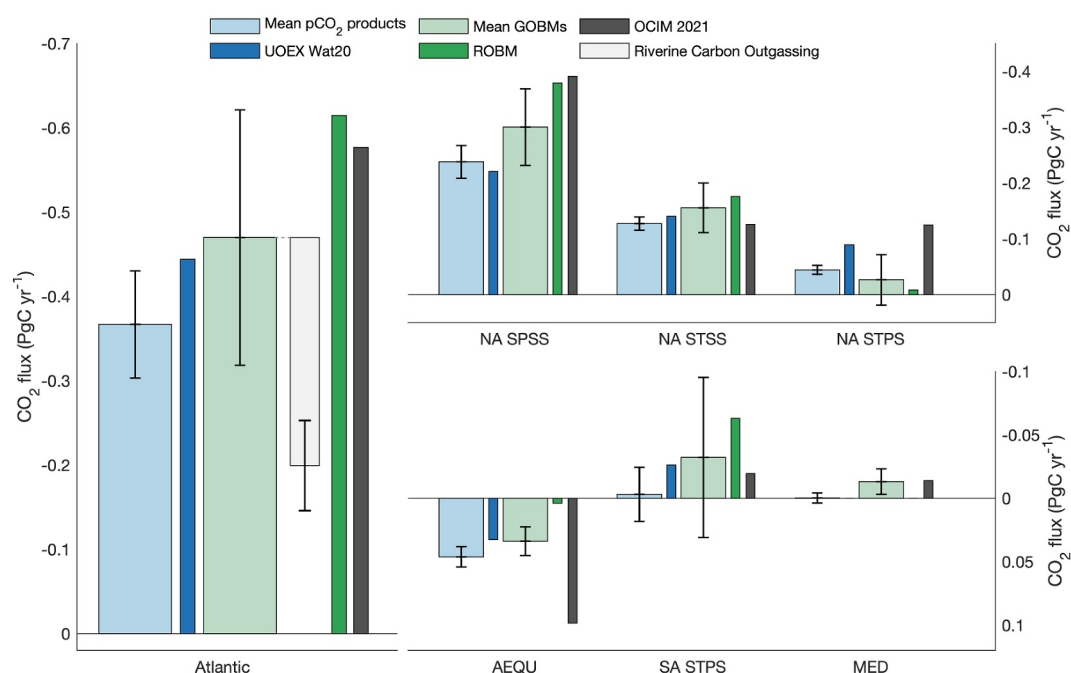
The mean sea-air CO<sub>2</sub> fluxes of the pCO<sub>2</sub> products and GOBMs have very similar spatial patterns when averaged over the 1985 to 2018 period (Figures 1c and 1d). The pCO<sub>2</sub> products show a weak CO<sub>2</sub> outgassing over large areas of the tropical regions of the South and North Atlantic, which is more intense in the western equatorial Atlantic. In comparison, the GOBMs exhibit weaker CO<sub>2</sub> fluxes in the equatorial region but more intense CO<sub>2</sub> fluxes in the Benguela and Mauritanian upwelling areas. In these upwelling regions, the ocean circulation delivers nutrients and DIC to the surface layer where they are consumed by photosynthesizing organisms. In many of these regions, the supply of DIC from below exceeds the amount of DIC being drawn down by the net balance between photosynthesis and remineralization/respiration, that is, net community production, such that an excess of DIC and nutrients remain at the surface, indicative of an inefficient biological pump (Sarmiento & Gruber, 2006). As a result, these regions act as a source of CO<sub>2</sub> to the atmosphere. Downstream of many of these regions, the remaining nutrients and the DIC get drawn down completely. This resulting large increase in the biological pump efficiency makes these regions strong uptake regions. The NA SPSS and NA STSS biomes, and the southern parts of biome SA STPS are characterized by strong CO<sub>2</sub> uptake with some differences between the spatial patterns modeled in the GOBMs and those derived from observations. In these regions, both the cooling of the warm poleward moving waters and an efficient and strong biological pump promote CO<sub>2</sub> uptake from the atmosphere (Takahashi et al., 2009; Thomas et al., 2008; Watson et al., 1995).

pCO<sub>2</sub> products and GOBMs are in good agreement with respect to their zonally integrated CO<sub>2</sub> fluxes when regarding the northern hemisphere between equator and 40°N and the southern hemisphere south of 20°S (Figure 1b). The GOBMs show a more intense ocean uptake of CO<sub>2</sub>, coinciding with the deep convection regions in the subpolar gyre (NA SPSS biome). In this region, models underestimate the transport and mixing of high subsurface DIC water to the surface during winter, underestimating the winter-time outgassing from the ocean (McKinley et al., 2018). The results obtained with the ROBM are very similar to that of the GOBMs between 35°S and 52°N, while the ROBM seems to overestimate uptake north of 52°N even more than the GOBMs. The inverse model OCIMv2021 follows the large-scale pattern of the other products, but shows more meridional variations and, similar to the ROBM, it also simulates a much stronger uptake than seen in models and observations north of 52°N (Figure 1b).

As shown in Figure S1 of the Supporting Information S1, the SOCAT gridded data of pCO<sub>2</sub> covers the NA SPSS biome with the highest number of observations among the Atlantic biomes, resulting in an average of 10.2% of the maximum possible coverage since 2003 and making it one of the regions where the pCO<sub>2</sub> products are expected to provide comparatively robust results. The UOEX data product, that adjusts the pCO<sub>2</sub> for near-surface temperature and salinity gradients, shows higher CO<sub>2</sub> uptake than the ensemble mean of the other pCO<sub>2</sub> products between 35°S and 50°N. This difference contains the expected effect of lower skin temperature on solubility, for which adjustments have been made in the UOEX product, but it also inherits the influence of different gap filling methods. Dong et al. (2022) have globally reevaluated the effect of skin temperature on FCO<sub>2</sub>, showing an impact on FCO<sub>2</sub> that is 30% lower than that previously evaluated by Watson et al. (2020). The net effect of skin SST and salinity on FCO<sub>2</sub> integrated over the whole Atlantic and its five biomes is detailed in Table S4 of the Supporting Information S2. The change in CO<sub>2</sub> uptake due to the temperature effects estimated by Dong et al. (2022) is overall similar to the difference in FCO<sub>2</sub> between UOEX and the ensemble mean of nine pCO<sub>2</sub> products except for NA SPSS and SA STPS, where the different gap-filling methodology has a greater effect.

Integrated over the whole Atlantic Ocean, the average sea-air CO<sub>2</sub> flux (FCO<sub>2</sub>) for the period 1985 to 2018 obtained from the GOBMs ( $-0.47 \pm 0.15$  PgC yr<sup>-1</sup>) is higher than that obtained from the pCO<sub>2</sub> products ( $-0.36 \pm 0.06$  PgC yr<sup>-1</sup>; Figure 2), although the difference is within the FCO<sub>2</sub> variability across the 11 GOBMs. OCIMv2021 estimates a larger uptake ( $-0.58 \pm 0.08$  PgC yr<sup>-1</sup>) than the GOBMs. The ROBM simulates an uptake of  $-0.61 \pm 0.14$  PgC yr<sup>-1</sup>, about 30% and 65% stronger than the mean of the GOBMs and pCO<sub>2</sub> products, respectively. Relative to the mean of the pCO<sub>2</sub> products, the CO<sub>2</sub> uptake in UOEX is larger by about 23%.

The NA SPSS biome, which covers only 15% of the Atlantic Ocean surface area, has the largest CO<sub>2</sub> uptake and also the largest differences between models and observational products (Figures 1 and 2, Table 1). Here, the mean FCO<sub>2</sub> of the GOBMs, the ROBM and OCIMv2021 indicate 26%, 59%, and 64% greater carbon uptake,



**Figure 2.** Spatially integrated sea-air  $\text{CO}_2$  fluxes from 1985 to 2018 for the Atlantic and each Atlantic biome as estimated by nine  $\text{pCO}_2$  products, 10 GOBMs, the UOEX  $\text{pCO}_2$ -data product, the ROB and OCIMv2021. The white bar indicates an estimate for the outgassing of riverine carbon integrated over the whole Atlantic region, which is a flux component captured by the  $\text{pCO}_2$  products but not by the GOBMs or the ROB. Whiskers stand for standard deviation around the mean of the estimates. Negative values indicate the uptake of  $\text{CO}_2$  from the atmosphere. Note that the y-axis are reversed, so that uptake is above the zero-line and outgassing is below it.

respectively, than the  $\text{pCO}_2$  products. The spread between GOBMs is three times larger than it is for the  $\text{pCO}_2$  products (Table S4 in Supporting Information S2). The uptake flux in UOEX is slightly lower ( $\sim 7\%$ ) relative to the mean of  $\text{pCO}_2$  products.

In the NA STPS biome, there is good agreement between the different  $\text{pCO}_2$  products, with a standard deviation that is less than 10% of the mean ( $-0.13 \pm 0.01 \text{ PgC yr}^{-1}$ ). Here, the UOEX product has 10% larger  $\text{CO}_2$  uptake. With an average  $\text{FCO}_2$  of  $-0.15 \pm 0.04 \text{ PgC yr}^{-1}$ , the GOBMs vary substantially more among each other, that is,  $\pm 30\%$ . In fact, one GOBM has a  $\sim 50\%$  weaker uptake than the GOBMs mean, while the GOBMs with the most intense fluxes are only 20% above the GOBMs mean. OCIMv2021 simulates  $\text{FCO}_2$  values of similar magnitude to the  $\text{pCO}_2$  products (Table S4 in Supporting Information S2).

For the NA STPS biome,  $\text{pCO}_2$  products estimate a mean  $\text{CO}_2$  uptake of  $-0.044 \pm 0.008 \text{ PgC yr}^{-1}$  with a very high homogeneity in spite of the large area of this biome. In comparison to the  $\text{pCO}_2$  products, the uptake simulated by the GOBMs is smaller ( $-0.020 \pm 0.040 \text{ PgC yr}^{-1}$ ), and with larger intermodel variations. Only three GOBMs estimated a  $\text{CO}_2$  outgassing in this biome. In contrast, OCIMv2021 reported a quite high uptake of  $\text{CO}_2$ , almost three times larger than that of the  $\text{pCO}_2$  products. The ROB simulates a near-zero net flux in this biome. In the UOEX product, the uptake was twice as large as the mean of the other  $\text{pCO}_2$  products.

All models and  $\text{pCO}_2$  products agree that the AEQU biome is a net source of  $\text{CO}_2$  to the atmosphere, consistent with the known impact of the equatorial upwelling that brings water with high DIC content to the ocean surface. The mean flux of the  $\text{pCO}_2$  products is  $0.046 \pm 0.009 \text{ PgC yr}^{-1}$ . In the UOEX product, this outgassing is 25% lower. The mean flux in the GOBMs is  $0.035 \pm 0.011 \text{ PgC yr}^{-1}$ , and has relatively small inter-model variations. The ROB simulates a very low outgassing. OCIMv2021 shows strong  $\text{FCO}_2$ , with more than double the outgassing of the mean GOBMs.

The SA STPS biome covers a large area, extending from the southern border of the equatorial region in the north toward the subtropical front of the Southern Ocean to the south. According to the mean of the  $\text{pCO}_2$  products, the integrated flux over this region is neither a sink nor a source of  $\text{CO}_2$  to the atmosphere ( $-0.003 \pm 0.023 \text{ PgC yr}^{-1}$ ).

**Table 1**

Sea-Air Surface CO<sub>2</sub> Fluxes (1985–2018) and Anthropogenic CO<sub>2</sub> Accumulation Rates (1994–2007) of All Products Used in This Study, With Their Respective Standard Deviations or Uncertainties of Individual Estimates

Period	BIOME (Area · 10 <sup>12</sup> m <sup>2</sup> )	FCO <sub>2</sub> (PgC yr <sup>-1</sup> )			
		pCO <sub>2</sub> products Ensemble mean	GOBM Ensemble mean	ROBM ROMS-ETHZ	Assimilation model OCIMv2021
1985–2018	ATLANTIC (68.7)	−0.37 ± 0.06	−0.47 ± 0.15	−0.61 ± 0.15	−0.58 ± 0.08
	NA SPSS (9.37)	−0.24 ± 0.03	−0.30 ± 0.07	−0.38 ± 0.05	−0.40 ± 0.03
	NA STSS (6.14)	−0.127 ± 0.012	−0.149 ± 0.041	−0.176 ± 0.022	−0.126 ± 0.012
	NA STPS (22.7)	−0.044 ± 0.008	−0.020 ± 0.041	−0.008 ± 0.026	−0.125 ± 0.024
	AEQU (8.69)	0.046 ± 0.008	0.035 ± 0.011	0.004 ± 0.016	0.098 ± 0.005
	SA STPS (19.6)	−0.003 ± 0.021	−0.029 ± 0.065	−0.063 ± 0.040	−0.020 ± 0.022
	MED (2.26)	0.000 ± 0.005	−0.015 ± 0.009	–	−0.014 ± 0.003
Period	BIOME (Area · 10 <sup>12</sup> m <sup>2</sup> )	ΔC <sub>ant</sub> (PgC yr <sup>-1</sup> )			
		C <sub>ant</sub> reconstruction Gruber et al., 2019	C <sub>ant</sub> reconstruction Khattiwala et al., 2009	GOBM Ensemble mean	Assimilation model OCIMv2021
1994–2007	ATLANTIC (68.7)	0.72 ± 0.08	0.63 ± 0.11	0.52 ± 0.11	0.68 ± 0.01
	NA SPSS (9.37)	0.087 ± 0.007	0.149 ± 0.027	0.087 ± 0.033	0.127 ± 0.001
	NA STSS (6.14)	0.098 ± 0.005	0.105 ± 0.018	0.080 ± 0.031	0.107 ± 0.001
	NA STPS (22.7)	0.254 ± 0.017	0.199 ± 0.036	0.175 ± 0.045	0.236 ± 0.002
	AEQU (8.69)	0.058 ± 0.018	0.040 ± 0.007	0.037 ± 0.006	0.054 ± 0.001
	SA STPS (19.6)	0.216 ± 0.041	0.137 ± 0.025	0.127 ± 0.018	0.156 ± 0.001
	MED (2.26)	–	–	0.0176 ± 0.0068	0.0186 ± 0.0001

Note. For ΔC<sub>ant</sub> the MED biome is not included in the total Atlantic estimate to facilitate direct comparison with Gruber et al. (2019). In each biome, the graduate yellow color background sorts the estimates from lowest flux into the ocean (light yellow) to largest flux into the ocean (dark yellow), as well as the estimates from lowest ΔC<sub>ant</sub> (light yellow) to largest ΔC<sub>ant</sub> (dark yellow).

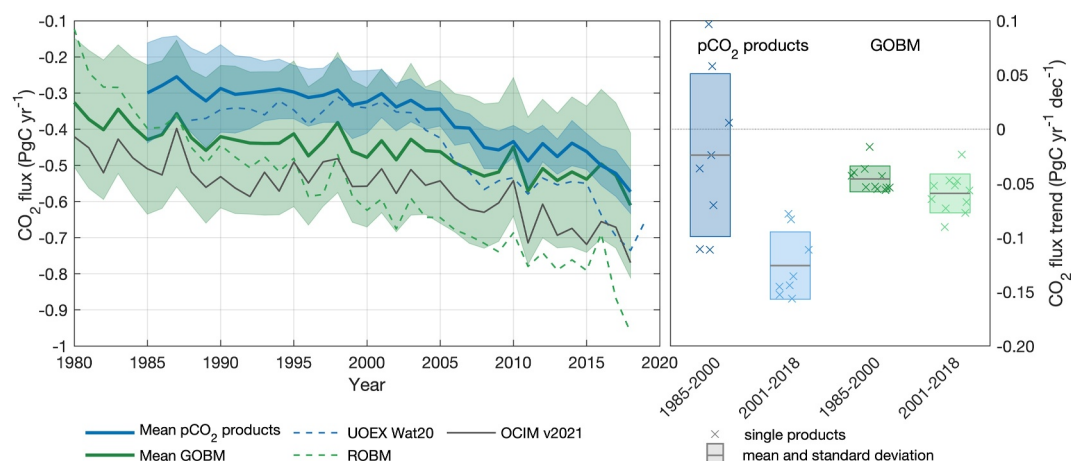
But, the spread across the pCO<sub>2</sub> products is relatively large in this region, second only to the spread in the NA SPSS, in part because of the large area of the SA STPS biome. On average, the GOBMs indicate that this region is a CO<sub>2</sub> sink with an estimated integrated flux of  $-0.029 \pm 0.076$  PgC yr<sup>-1</sup>. However, an integrated outgassing is simulated by 1/3 of the GOBMs. The FCO<sub>2</sub> in the ROBM is nearly twice as large as the mean of the GOBMs, while the OCIMv2021 suggested that the region behaves as a weaker CO<sub>2</sub> sink.

In the Mediterranean Sea, only five of the nine pCO<sub>2</sub> products have a regional coverage better than 95%. Of these, four pCO<sub>2</sub> products agree that the biome does not present significant sea-air CO<sub>2</sub> fluxes (Figure 2; Table S4 in Supporting Information S2). Most of the GOBMs have a coverage better than 95% and they broadly agree that the Mediterranean Sea represents a very weak CO<sub>2</sub> sink ( $-0.015 \pm 0.010$  PgC yr<sup>-1</sup>). The flux in the OCIMv2021 is very similar. The ROBM has insufficient regional coverage for an assessment in this biome.

In summary, for the Atlantic, the GOBMs predict a  $28\% \pm 14\%$  larger CO<sub>2</sub> uptake than pCO<sub>2</sub> products (Table 1). The regional and data-assimilation models simulate a stronger Atlantic CO<sub>2</sub> sink than pCO<sub>2</sub> products by 67% and 57%, respectively (Table S4 in Supporting Information S2). The same is the case for the UOEX product, where the CO<sub>2</sub> uptake is 25% larger than that of the mean pCO<sub>2</sub> products, as a consequence of its adjustment for near surface temperature gradients.

### 3.1.2. FCO<sub>2</sub> Trends

The temporal evolution of the annual mean sea air fluxes in the pCO<sub>2</sub> products shows a change of rate around the year 2000 (Figure 3). In agreement with the recommended core analysis in RECCAP2, we thus analyzed changes in FCO<sub>2</sub> during two periods: between 1985 and 2000, and between 2001 and 2018. Over these two periods, the atmospheric CO<sub>2</sub> concentration increased on average by 1.5 and 2.1 ppm yr<sup>-1</sup>, respectively, representing an acceleration in the atmospheric growth rate of 43% from the first to the second period. Integrated over the Atlantic

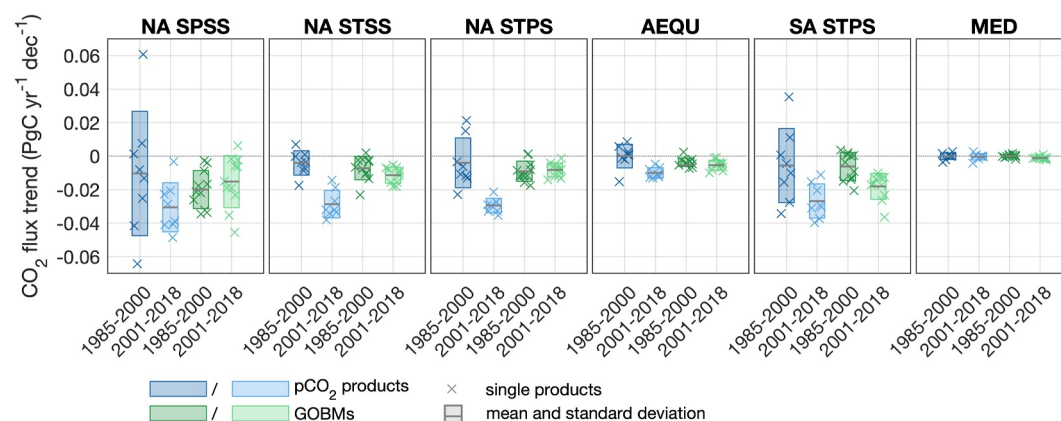


**Figure 3.** Trends in the sea-air  $\text{CO}_2$ -fluxes of the Atlantic Ocean. Time-series of the annual mean sea-air  $\text{CO}_2$  fluxes ( $\text{PgC yr}^{-1}$ ). Boxplots of the ensemble mean trends in sea-air  $\text{CO}_2$  fluxes ( $\text{PgC yr}^{-1} \text{dec}^{-1}$ ) and their  $1\sigma$  spread before and after 2000. Shown are the ensemble of the  $\text{pCO}_2$  products (blue) and GOBMs (green) on both panels, and UOEX, OCIM and one ROBM on the left panel.

as a whole, the  $\text{pCO}_2$  products indicate a 5-fold increase in the growth rate of the ocean carbon sink from  $-0.024 \pm 0.075 \text{ PgC yr}^{-1} \text{dec}^{-1}$  between 1985 and 2000 to  $-0.126 \pm 0.031 \text{ PgC yr}^{-1} \text{dec}^{-1}$  between 2001 and 2018 (Figure 3). In contrast, GOBMs simulate only a 33% increase in the growth rate between the two periods, that is, from  $-0.045 \pm 0.012 \text{ PgC yr}^{-1} \text{dec}^{-1}$  between 1985 and 2000 to  $-0.060 \pm 0.017 \text{ PgC yr}^{-1} \text{dec}^{-1}$  between 2001 and 2018 (Figure 3). This is only slightly below the observed acceleration in the atmospheric  $\text{CO}_2$  growth rate. The two products also differ strongly with regard to their spreads (Figure 3b). While the  $\text{pCO}_2$  products exhibit a relatively low spread for the 1985–2018 mean flux, they differ considerably with regard to their  $\text{FCO}_2$  trends. Conversely, GOBMs show a large spread in the 1985–2018 mean flux, but have a low spread in their  $\text{FCO}_2$  trends in both periods, reflecting that the trends in the GOBMs are more strongly governed by the rate of change in atmospheric  $\text{CO}_2$ .

The  $\text{CO}_2$  uptake trend increased in OCIMv2021 from  $-0.045 \pm 0.016 \text{ PgC yr}^{-1} \text{dec}^{-1}$  during the first period to  $-0.111 \pm 0.018 \text{ PgC yr}^{-1} \text{dec}^{-1}$  for the second period. Its estimate is thus similar to that of the GOBMs in the first period but almost twice as large in the second. The ROBM simulates a much stronger growth than the GOBMs in both periods ( $-0.19 \pm 0.02$  and  $-0.14 \pm 0.02 \text{ PgC yr}^{-1} \text{dec}^{-1}$ ), but no significant change in trend. On the other hand, the UOEX  $\text{pCO}_2$  product reveals an even greater contrast between the growth rates before 2000 ( $0.048 \pm 0.014 \text{ PgC yr}^{-1} \text{dec}^{-1}$ ) and after 2000 ( $-0.188 \pm 0.012 \text{ PgC yr}^{-1} \text{dec}^{-1}$ ) than the ensemble mean of the  $\text{pCO}_2$  products. The trends obtained by the UOEX product showed a weakening of  $\text{CO}_2$  uptake in the Atlantic Ocean before 2000, and an increase of about  $0.35 \text{ PgC yr}^{-1}$  in the second period, which is higher than in any of the other eight  $\text{pCO}_2$  products (range: 0.14 to  $0.28 \text{ PgC yr}^{-1}$ ). Three of the other  $\text{pCO}_2$  products also suggest a weakening of the  $\text{CO}_2$  uptake in the Atlantic before 2000, while four other products suggest increasing trends in  $\text{CO}_2$  uptake by the Atlantic. Possibly the sharp contrast in observational coverage before and after the year 2000 (Figure S1 in Supporting Information S1; Bakker et al., 2022), as well as the availability of observed predictor data affected in a noticeable way some of the products. Indeed, the agreement among  $\text{pCO}_2$  products significantly improved throughout the 1985–2000 period. This underscores a notable distinction from the GOBMs, as the observation-based trends in the initial period are markedly influenced by early-year  $\text{FCO}_2$  estimates (Figure S2 in Supporting Information S1). During this period, only limited  $\text{pCO}_2$  observations and predictor variables are available, and most products rely on climatology of specific predictors, such as chlorophyll and mixed layer depth, due to a scarcity of observational data.

Temporal trends in the individual biomes of the Atlantic are variable and highly dependent on the products used to estimate them (see Figure 4; Table S5 in Supporting Information S2). Between 2001 and 2018, the  $\text{pCO}_2$  products show that the  $\text{CO}_2$  uptake rate grows with values close to  $-0.03 \text{ PgC yr}^{-1} \text{dec}^{-1}$  in the NA SPSS, NA STSS, NA STPS, and SA STPS biomes (which present very different areas) and  $-0.01 \text{ PgC yr}^{-1} \text{dec}^{-1}$  in the AEQU biome. During this period, all  $\text{pCO}_2$  products agreed on the sign of the trend in all biomes, with the exception of the MED



**Figure 4.** Trends in sea-air CO<sub>2</sub>-fluxes for each Atlantic biome and two different time-periods as estimated by pCO<sub>2</sub> products (blue) and GOBMs (green). Shown are individual products (crosses), together with the ensemble mean and 1 $\sigma$  spread.

biome where the trend was consistently close to zero. However, for the period 1985–2000, trends in CO<sub>2</sub> uptake estimated by the pCO<sub>2</sub> products are more variable across the different products and biomes, with non-significant trends in FCO<sub>2</sub> in NA STPS ( $-0.004 \pm 0.015$  PgC yr<sup>-1</sup> dec<sup>-1</sup>), AEQU ( $0.000 \pm 0.001$  PgC yr<sup>-1</sup> dec<sup>-1</sup>), and SA STPS ( $-0.006 \pm 0.022$  PgC yr<sup>-1</sup> dec<sup>-1</sup>) and mark a notable contrast between the two periods.

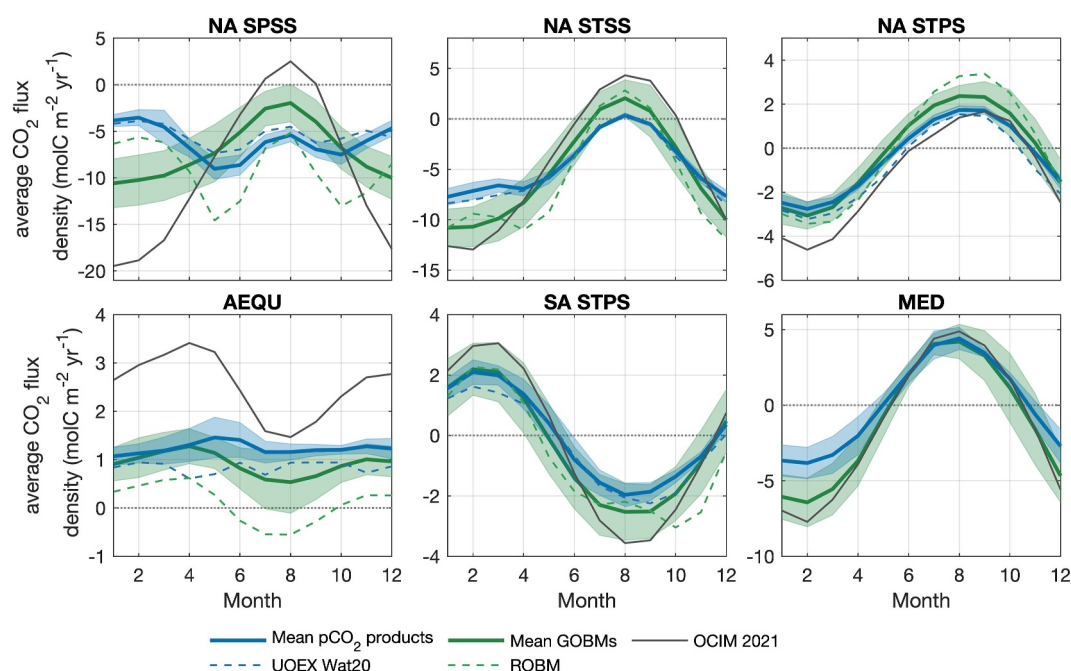
The biome-level trends are more consistent across the GOBMs than across the pCO<sub>2</sub> products, and also more similar in the two periods. In three biomes, NA STSS, AEQU and SA STPS, the GOBMs simulate on average an increase in fluxes to the ocean between the first and the second periods. The disagreement among the GOBMs and between GOBMs and pCO<sub>2</sub> products is largest in the NA SPSS biome. The ROBM shows rates of increase of CO<sub>2</sub> uptake higher than  $-0.03$  PgC yr<sup>-1</sup> dec<sup>-1</sup> in practically all biomes and in both periods except in NA STPS in the second one, and AEQU biomes in both (see Table S5 in Supporting Information S2). OCIMv2021 shows similar rates of increase to those observed in pCO<sub>2</sub> products for the second period, in line with the ROBM (Table S5 in Supporting Information S2).

### 3.1.3. Seasonal Cycle

The Atlantic Ocean sea-air CO<sub>2</sub> flux varies seasonally in a pronounced manner in all biomes, except for the equatorial (Figure 5). The Mediterranean Sea and the subtropical biomes (in their respective hemispheres) are CO<sub>2</sub> sinks in winter and sources in summer. Here, the impact of biological DIC drawdown on pCO<sub>2</sub> is relatively weak and seasonal warming and cooling dominate the seasonal cycle such that it peaks and reaches supersaturation in summer while minimum and undersaturated values occur in winter (Figures S3 and S4 in Supporting Information S1; Rodgers et al., 2023). The seasonal amplitude in the flux in these regions is slightly larger in the GOBMs than in the pCO<sub>2</sub> products. This has been attributed to a likely underestimation of seasonal mixed layer depth changes and seasonal drawdown of DIC by net primary production, such that the thermal component on the seasonal pCO<sub>2</sub> and sea-air CO<sub>2</sub> flux cycle is too strong in these models (Rodgers et al., 2023). The OCIMv2021 is an abiotic model and shows the largest seasonal pCO<sub>2</sub> (Figure S3 in Supporting Information S1) and flux variations because of the complete absence of biological processes. The difference in the seasonal cycle as modeled by the OCIMv2021 and the other GOBMs can be taken as a rough estimate of the importance of biology.

In the NA SPSS biome, the GOBMs' seasonal CO<sub>2</sub> flux cycle is similar to that in the subtropical biomes (and of the abiotic OCIM model), while that of the pCO<sub>2</sub> products is broadly reversed, apart from the summertime intermediate minimum in CO<sub>2</sub> uptake (Figure 5). The pCO<sub>2</sub> products have the highest pCO<sub>2</sub> values in winter, as a consequence of the supply of remineralized DIC into the surface layer through deep mixing (Figure S3 in Supporting Information S1). Seasonal stratification and increased light availability trigger spring blooms that cause a sharp pCO<sub>2</sub> decrease from March to June, after which the pCO<sub>2</sub> steadily increases back to its winter maximum. The existence of these patterns is well known from the many direct observations in this region (Becker et al., 2018; Fröb et al., 2019; Olsen et al., 2008; Takahashi et al., 1993). The opposite seasonal pCO<sub>2</sub> cycle in the GOBMs is likely due to the fact that their seasonal variations in mixed layer depths are too small (Rodgers et al., 2023), such that too few nutrients are upwelled during winter, likely resulting in an underestimation of





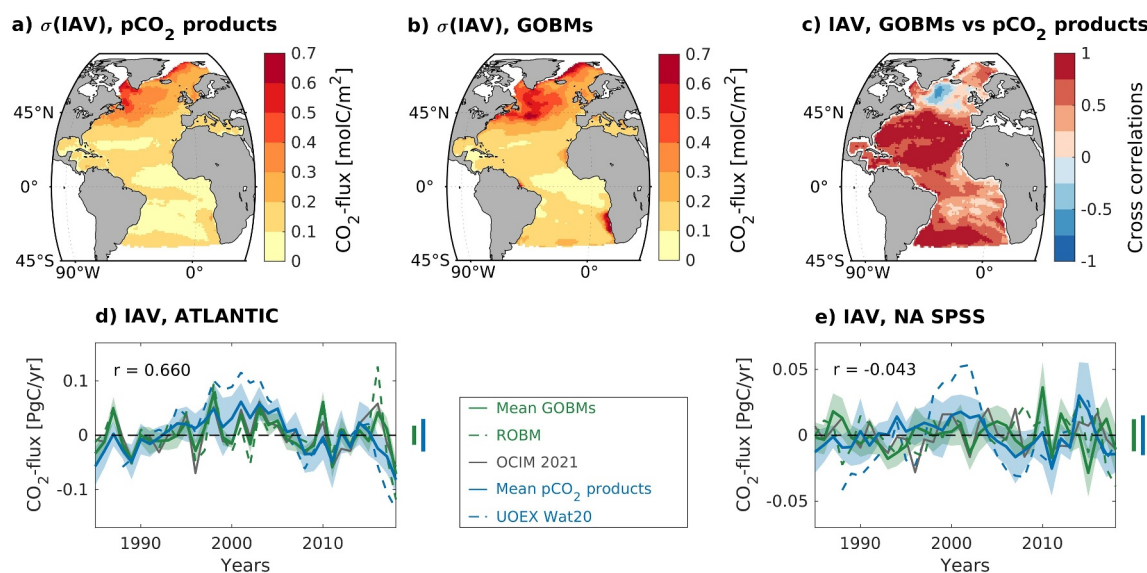
**Figure 5.** Seasonal cycle of the sea-air CO<sub>2</sub>-fluxes for each Atlantic biome as estimated by pCO<sub>2</sub> products (blue) and GOBMs (green), both shown as the ensemble mean (thick lines) and 1 $\sigma$  spread (shadings) (1985–2018 average). Additional lines represent UOEX, OCIM and one ROBM. OCIM is an abiotic model and thus does not include the effect of the seasonality of net community production.

summer biological drawdown of DIC in the GOBMs (Rodgers et al., 2023; also shown for Earth System Models in Goris et al. (2018)). Since the opposing seasonal cycle of the GOBMs leads to a lower pCO<sub>2</sub> at the time of the strongest wind speeds, the GOBM ensemble shows higher annual net NA SPSS CO<sub>2</sub> uptake (Figures 1 and 2), that is, the GOBMs tend to simulate too strong uptake. Again, the OCIMv2021, as an abiotic model, is an extreme example of these model-effects. The ROBM appears more consistent with the pCO<sub>2</sub> products in this regard, but it overall appears to overestimate the NA SPSS CO<sub>2</sub> uptake as the modeled pCO<sub>2</sub> values are too low (Figure S3 in Supporting Information S1). The summertime intermediate minimum in CO<sub>2</sub> uptake in the pCO<sub>2</sub> products is a consequence of the minimum in wind speeds in that season. More quantitative analyses of the seasonal cycle including their drivers and differences between GOBMs and pCO<sub>2</sub> products are presented by Rodgers et al. (2023).

### 3.1.4. Interannual Variability of the Sea-Air CO<sub>2</sub> Fluxes

We further analyzed the interannual variability (IAV) of sea-air CO<sub>2</sub> fluxes, determined as the annual anomaly of the detrended sea-air CO<sub>2</sub> fluxes with respect to their mean values. Here, the removed linear trends and means are considered over the period 1985–2018 for pCO<sub>2</sub> products and GOBMs. When referencing the amplitude of IAV, we refer to the standard deviation of the so-derived detrended sea-air CO<sub>2</sub> flux anomalies. We find that, over the whole Atlantic basin, the IAV time-series of the sea-air CO<sub>2</sub> fluxes of GOBMs and pCO<sub>2</sub> products correlate relatively well (Figure 6d). Furthermore, both pCO<sub>2</sub> products and GOBMs show a high IAV amplitude in the northern parts and low IAV amplitude in the equatorial region (Figures 6a and 6b). This general spatial pattern of the IAV amplitude of net sea-air CO<sub>2</sub> fluxes has also been found in other studies (Brady et al., 2019; Park et al., 2010). However, the GOBMs show a larger IAV amplitude than the pCO<sub>2</sub> products in the interior subpolar gyre as well as in the eastern boundary upwelling regions (Figures 6a and 6b), while showing a smaller IAV amplitude for the NA SPSS biome as a whole (Figure 6e).

The pCO<sub>2</sub> products and GOBMs agree on the phasing of the IAV in net sea-air CO<sub>2</sub> fluxes, apart from in the subpolar region where correlations are small and negative (Figures 6c and 6e; Figure S5 in Supporting Information S1). We note that there is also little agreement in the IAV of this biome between pCO<sub>2</sub> products (Figure S6 in Supporting Information S1), while the GOBMs agree relatively well (Figure S6 in Supporting Information S1).



**Figure 6.** Interannual Variability (IAV) of observation-based and simulated sea-air CO<sub>2</sub> fluxes in the Atlantic. Panels (a and b) show the spatial distribution of the amplitude of IAV (calculated as standard deviation of the time-series of detrended annual sea-air CO<sub>2</sub> flux anomalies per grid point) for both (a) pCO<sub>2</sub> products and (b) GOBMs. Correlations between time-series of ensemble-averaged detrended sea-air CO<sub>2</sub> flux anomalies of GOBMs and pCO<sub>2</sub> products are shown in panel (c). Time-series of detrended annual sea-air CO<sub>2</sub> flux anomalies for both GOBMs (mean: green line; std: green shading) and pCO<sub>2</sub> products (mean: blue line; std: blue shading) are illustrated in panel (d) for the whole Atlantic and (e) for the NA SPSS biome, including the correlation coefficients between the time-series of GOBMs and pCO<sub>2</sub> products (denoted in the upper left corner of each panel d and e) and the amplitude of the IAV (illustrated as colored lines on the right side of each plot d and e).

GOBMs and pCO<sub>2</sub> products agree that the total sea-air CO<sub>2</sub> fluxes of the biomes NA STSS, NA STPS, and SA STPS are characterized by a moderate IAV amplitude (Figure S7 in Supporting Information S1), and that biomes AEQU and MED have only a weak IAV amplitude (see Figure S5 in Supporting Information S1). For these five biomes, GOBMs and pCO<sub>2</sub> products correlate reasonably well with respect to the temporal variability of the IAV with correlation coefficients ranging from  $r = 0.57$  to  $r = 0.73$  (see also Figure S5 in Supporting Information S1).

For the GOBM ensemble, the IAV of net sea-air CO<sub>2</sub> fluxes is strongly positively correlated to the IAV in SST (higher FCO<sub>2</sub> in anomalously warm years) over large parts of the Atlantic basin, most notably for both permanently stratified biomes (SA STPS and NA STPS) and the northwestern subpolar gyre (Figure S8b in Supporting Information S1). Along the Gulf Stream and the North Atlantic Current as well as regions of equatorial upwelling, the IAV in net sea-air CO<sub>2</sub> fluxes of the GOBM ensemble is weakly negatively correlated to the IAV in SST (higher FCO<sub>2</sub> in anomalously cold years). Due to the known dynamics of net sea-air CO<sub>2</sub> fluxes, these negative correlations imply that SST-variations are not the main driver of the IAV in net sea-air CO<sub>2</sub> fluxes but that the anomalous cold years are likely accompanied by stronger mixing and hence more DIC upwelling. As the thermodynamic boundary conditions used to force the GOBMs result in SSTs that have relatively strong fidelity to observations when averaged over biome scales, it is plausible that the relatively small model spread around the IAV in the Atlantic is related to the fact that most of the simulated IAV is driven by SST-variations (areas with positive correlations in Figure S8b of the Supporting Information S1) and that variations in DIC play a less important role. The strong relationship to SST is also the plausible cause for high correlations between IAV in net sea-air CO<sub>2</sub> fluxes of pCO<sub>2</sub> products and GOBMs in SA STPS and NA STPS. Indeed, when correlating the IAV in net sea-air CO<sub>2</sub> fluxes of pCO<sub>2</sub> products to the IAV in SST (Figure S8a in Supporting Information S1), we find strong correlations in SA STPS and NA STPS biomes. However, in the NA SPSS, the pCO<sub>2</sub> products appear to be more negatively correlated to the IAV in SST (likely driven by DIC variations), in contrast to the GOBMs. This difference in mechanisms over the subpolar gyre is one possible explanation for the disagreement in the IAV in net sea-air CO<sub>2</sub> fluxes between pCO<sub>2</sub> products and GOBMs in the NA SPSS biome.

In the North Atlantic, one of the most prominent climate variability modes at interannual time scales is the NAO. In a study about the influences of NAO on the IAV of North Atlantic CO<sub>2</sub> fluxes, Jing et al. (2019) noted that, in summer, SST is important for the IAV in pCO<sub>2</sub> in the subtropical North Atlantic, while biogeochemical variables probably control the pCO<sub>2</sub> IAV in the subpolar North Atlantic. When relating the IAV of the GOBMs to NAO, we

find significant but weak correlations for the NA SPSS and the AEQU biomes ( $r = -0.43$ ,  $p = 0.01$  and  $r = -0.48$ ,  $p = 0.004$ ), whereas all other biomes show no significant correlation to the NAO index. However, the  $p\text{CO}_2$  products show a similar correlation between NAO and IAV for the AEQU biome ( $r = -0.41$ ,  $p = 0.02$ ), but no significant correlation in the NA SPSS biome (neither for the average nor for single products). The similar correlation in the AEQU is consistent with the fact that in this region, the temperature-driven Atlantic Niño climatic mode plays an important role in modulating the IAV of  $\text{CO}_2$  fluxes (Koseki et al., 2023), and that the GOBM - simulated SST variability is well constrained by the observations. The absent correlation in the NA SPSS could be due to the opposing imprints of NAO on  $p\text{CO}_2$  in the western and eastern domains of the NA SPSS. In a modeling study, Tjiputra et al. (2012) demonstrated that during positive NAO, SST cooling induces a negative  $p\text{CO}_2$  anomaly in the western subpolar gyre, whereas in the eastern part (in the proximity of the Irminger Sea) anomalously deep winter mixing upwells DIC-rich watermasses and induces a positive  $p\text{CO}_2$  anomaly (e.g., Fröb et al., 2019). The opposite mechanism is suggested during negative NAO.

We note that despite relatively high correlations between IAV of GOBMs and  $p\text{CO}_2$  products in all biomes apart from the NA SPSS (Figure 6c), the amplitude of the IAV of the GOBMs is smaller than that of  $p\text{CO}_2$  products in all biomes except the NA STSS (Figure S5 in Supporting Information S1). The amplitude of the IAV of the total sea-air  $\text{CO}_2$  fluxes in the Atlantic basin is  $0.029 \pm 0.01 \text{ PgC yr}^{-1}$  ( $p\text{CO}_2$  products) and  $0.018 \pm 0.005 \text{ PgC yr}^{-1}$  (GOBMs). These results are significantly different but of similar magnitude to the linear trends of the sea-air  $\text{CO}_2$  fluxes of the Atlantic basin (Figure 3). For a better estimate of the sea-air  $\text{CO}_2$  fluxes in the Atlantic basin, it is therefore important to have an accurate estimate of both temporal variability and amplitude of the IAV, which is currently not adequately represented. Moreover, the temporal disagreement of IAV of  $p\text{CO}_2$  products in the NA SPSS makes it clear that a closer examination of the gap filling methods and their dynamic realism is urgently needed here (Gloege et al., 2021; Hauck, Nissen, et al., 2023).

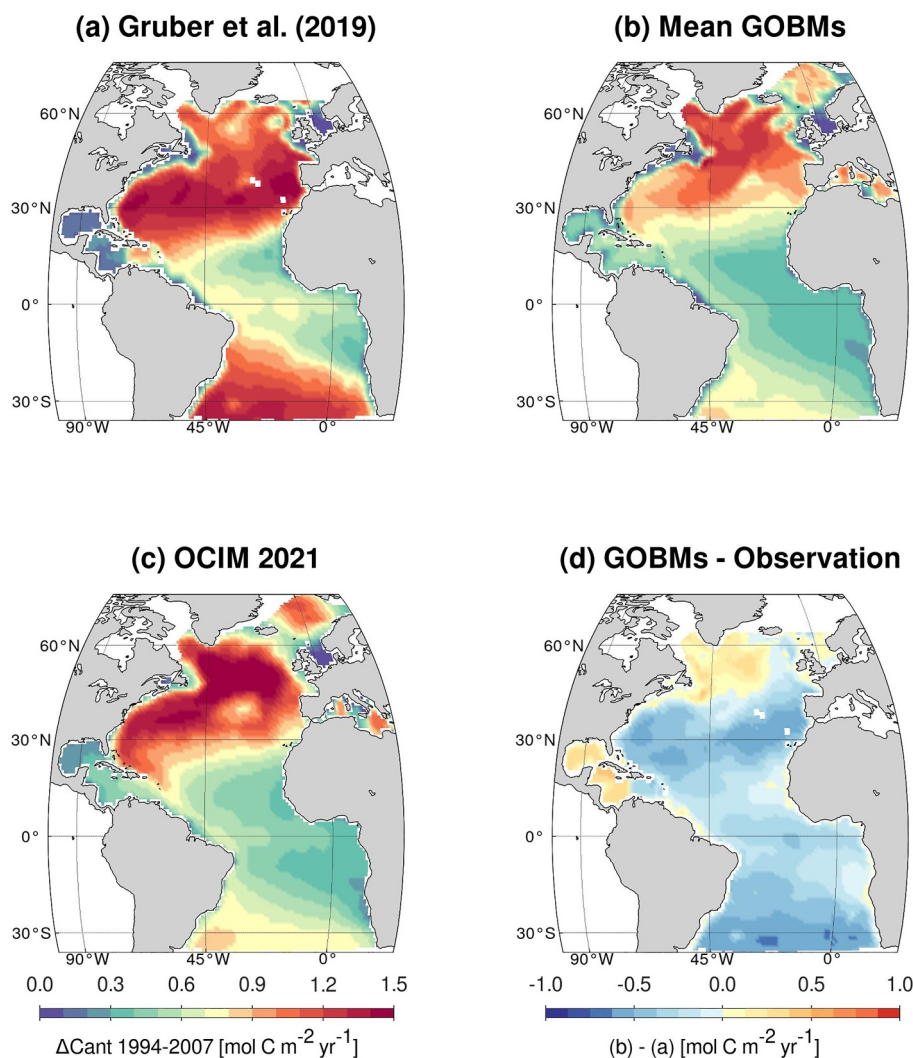
### 3.2. Ocean Interior $C_{\text{ant}}$ Accumulation From 1994 to 2007

The change in the oceanic storage of anthropogenic carbon ( $\Delta C_{\text{ant}}$ ) was evaluated for the period 1994–2007 for comparison between GOBMs, the data-assimilation model OCIMv2021 and two observation-based  $\Delta C_{\text{ant}}$  reconstruction products. All nine considered GOBMs simulated an increase in the basin-wide  $C_{\text{ant}}$  inventory that is broadly consistent among themselves and with observations (Table S6 in Supporting Information S2). Seven models show high column inventory changes of  $C_{\text{ant}}$  in the NA SPSS biome and in the NA STSS, consistent with the observation-based  $\Delta C_{\text{ant}}$  reconstructions (Figure S9 in Supporting Information S1), while the other two (PlankTOM12 and CESM-ETHZ) show high  $\Delta C_{\text{ant}}$  column inventories in the vicinity of 35°S, but very weak accumulation in the North Atlantic.

The spatial distribution of the change in column-integrated  $\Delta C_{\text{ant}}$  averaged across the ensemble of nine GOBMs is shown in Figure 7b. Spatial patterns in the  $\Delta C_{\text{ant}}$  column inventory distribution obtained by the OCIMv2021 inverse-model (Figure 7c) are very similar to the GOBM ensemble mean but with higher values throughout the Atlantic, except in the region of the Brazil Current and in the vicinity of the Azores Islands. In addition, OCIMv2021 produces a similar pattern to that obtained from the DIC-based product from Gruber et al. (2019) (Figure 7a), but with stronger (weaker)  $\Delta C_{\text{ant}}$  in the northernmost regions (south of the equator). The GOBM ensemble mean reveals slightly higher  $\Delta C_{\text{ant}}$  column inventories in the subpolar North Atlantic than the observation-based product from Gruber et al. (2019) (Figure 7d). In contrast, over the tropical and South Atlantic, the  $\Delta C_{\text{ant}}$  column inventory of the GOBM ensemble is only about half as high as the reconstruction of Gruber et al. (2019), representing the main discrepancy between both products.

Integrated over the whole Atlantic Ocean, the  $\Delta C_{\text{ant}}$  inventory simulated by the GOBM ensemble (Table 1) is about  $28\% \pm 20\%$  lower than the inventory estimate obtained with the observation-based eMLR( $C^*$ ) method (Gruber et al., 2019), 17% lower than the age-tracer based method (Khatriwala et al., 2009), and  $28\% \pm 15\%$  lower than the inverse model OCIMv2021. By contrast, the OCIMv2021  $\Delta C_{\text{ant}}$  inventory is very similar to the estimate from Gruber et al. (2019), while 8% higher compared to that of Khatriwala et al. (2009).

Integrated over the individual biomes of the Atlantic, we found the best agreement between the GOBM ensemble and the estimate from Gruber et al. (2019) in the northern biomes. In the NA SPSS and NA STSS biomes,  $C_{\text{ant}}$  accumulation rates are very similar between GOBMs, and only two models show extraordinarily low values (50% lower than the GOBMs average; Table S6 in Supporting Information S2). In contrast, OCIMv2021 simulates a  $\Delta C_{\text{ant}}$  inventory that is about 40% higher than the observation-based estimate and the GOBM ensemble mean, and



**Figure 7.** Column inventories of anthropogenic carbon storage changes ( $\Delta C_{ant}$ ), integrated from the surface to 3,000 m from 1994 to 2007. Shown are  $\Delta C_{ant}$  column inventories for (a) an observation-based reconstruction with the eMLR(C\*) method by Gruber et al. (2019); (b) multi-model GOBM ensemble mean; and (c) OCIMv2021. Panel (d) illustrates the difference between the estimates from the GOBM ensemble mean and Gruber et al. (2019).

therefore closer to the values obtained with the Green's Function (Khatiwala et al., 2009). Further south, discrepancies between the GOBM-based and the observation-based  $\Delta C_{ant}$  inventories increase, bringing the GOBM inventories closer to the age-tracer based product, while OCIMv2021 resembles the eMLR(C\*)-based estimates in two of the three remaining biomes. In the NA STPS biome, characterized by the largest inter-GOBM spread, the  $\Delta C_{ant}$  inventory of the GOBMs is about 25% lower than the observation-based product, while the OCIMv2021 inventory reveals a similar rate of change as the observation-based product. Likewise, in the AEQU biome, the GOBMs' ensemble mean  $\Delta C_{ant}$  inventory is approximately 30% lower than that from Gruber et al. (2019) and OCIMv2021. The AEQU biome further revealed the lowest  $\Delta C_{ant}$  inventories with a very narrow inter-GOBM spread. The largest  $\Delta C_{ant}$  inventory difference between the GOBMs and the observation-based product exists in the SA STPS biome, with a GOBM  $\Delta C_{ant}$  storage rate nearly 50% lower than that of Gruber et al. (2019). In addition, in the SA STPS biome, the OCIMv2021  $\Delta C_{ant}$  inventory is also 30% lower than that of Gruber et al. (2019). No comparison is done for the MED biome because of the lack of data in  $\Delta C_{ant}$  reconstruction products.

Average  $\Delta C_{ant}$  vertical profiles of each biome and for the whole Atlantic (Figure S10 in Supporting Information S1) reveal that the maximum  $\Delta C_{ant}$  occurs near the surface, while the accumulation rates decrease rapidly





**Figure 8.** Anthropogenic  $\text{CO}_2$  budget in the Atlantic using Simulation A-Simulation D of the GOBMs from 1994 to 2007. The blue arrows indicates sea-air  $\text{CO}_2$  fluxes and red arrows indicate lateral transport of  $\text{C}_{\text{ant}}$ .  $\Delta\text{C}_{\text{ant}}$  storage changes and sea-air  $\text{C}_{\text{ant}}$  fluxes (Table S7 in Supporting Information S2) are given in red bold numbers. Net sea-air  $\text{FCO}_2$  fluxes from GOBMs are given in blue. Red cursive numbers indicate northward  $\text{C}_{\text{ant}}$  transport inferred by the difference between  $\text{C}_{\text{ant}}$  accumulation rate and sea-air uptake. Gray numbers are the  $\Delta\text{C}_{\text{ant}}$  estimates from Gruber et al. (2019) and the  $\text{F}_{\text{ant}}$  sea-air flux estimated using the  $\text{C}_{\text{ant}}$  transport of  $0.19 \pm 0.020 \text{ PgC yr}^{-1}$  from Cainzos et al. (2022) at  $30^\circ\text{S}$ .

with depth. In general, GOBM simulations and the estimates from Gruber et al. (2019) agree with regard to this vertical distribution, both in the Atlantic and at the biome-level. However, in the NA STPS, AEQU, and SA STPS biomes, the observation-based reconstruction presents a second  $\Delta\text{C}_{\text{ant}}$  maximum between 1,400 and 3,000 m depths that is only reproduced by the GOBMs in the NA STPS. Such depth range is associated with waters with moderate values of  $\text{C}_{\text{ant}}$  transported by the DWBC circulating southward below the Antarctic Intermediate Water  $\Delta\text{C}_{\text{ant}}$  minimum (Fajar et al., 2015; Rhein et al., 2015; Rios et al., 2003, 2012), mainly North Atlantic Deep Water (NADW). The fact that the GOBMs do not agree with the observations in the southernmost biomes (AEQU and SA STPS) could be indicative of how the GOBMs ventilate the ocean interior below 1,400 m during 1994–2007 period. Updated reconstructions with the eMLR(C\*) method by Müller et al. (2023) detect these deep-water accumulations only for the period from 1994 to 2004 but not from 2004 to 2014. These findings could either indicate that (a)  $\Delta\text{C}_{\text{ant}}$  in the NADW is subject to larger decadal scale variability than simulated in GOBMs, that (b) the observational data from the 1990s used for the reconstructions from Gruber et al. (2019) as well as the first decade of the

reconstruction by Müller et al. (2023) contribute to unidentified biases in the observation-based estimates, or that (c) the statistical gap-filling with the eMLR method approaches its limits in reconstructing the low  $\text{C}_{\text{ant}}$  accumulation rates in these water masses.

### 3.3. Anthropogenic $\text{CO}_2$ Uptake and Lateral Transport

In terms of recent storage changes in  $\text{C}_{\text{ant}}$ , GOBMs tend to simulate lower accumulation rates than observation-based estimates (Section 3.2), whereas we have previously described that the net  $\text{CO}_2$  uptake is larger in GOBMs than  $\text{pCO}_2$  products (Section 3.1.1). To assess this apparent inconsistency, the anthropogenic component of the  $\text{CO}_2$  fluxes in GOBMs is assessed from the differences of Simulation A minus Simulation D in each GOBM, allowing us to determine the sea-air fluxes caused solely by increased  $\text{CO}_2$  in the atmosphere. The anthropogenic  $\text{FCO}_2$  averaged across 9 GOBMs ( $\text{F}_{\text{ant}}$ ) in the Atlantic as a whole is shown in Figure 8 and for each of the biomes in Figure S11 of the Supporting Information S1. Integrated over the entire Atlantic Ocean, the  $\text{F}_{\text{ant}}$  fluxes are lower in magnitude than the net flux  $\text{FCO}_2$  because the natural flux component contributes an additional  $\text{CO}_2$  uptake in the NA SPSS and NA STSS biomes. In the other biomes, the natural contributions are lower or even represent positive fluxes (outgassing) (Figure S11 in Supporting Information S1). However, the net result for the Atlantic Ocean is an uptake of natural  $\text{CO}_2$  (approx  $\sim 0.1 \text{ PgC yr}^{-1}$  obtained from the Simulation B) being transported to the Southern Ocean. In terms of  $\text{C}_{\text{ant}}$ , the biome with the highest uptake is also the NA SPSS, although the latitudinal variability is by far not as marked as in the natural component of  $\text{FCO}_2$ .

Knowing the  $\text{C}_{\text{ant}}$  accumulation rate in the ocean interior and the flux entering from the atmosphere, we can infer horizontal transport rates (Figure 8; Figure S11 in Supporting Information S1). Given the enclosed bathymetry of the Mediterranean Sea, the GOBM ensemble simulates a mean net export of  $\text{C}_{\text{ant}}$  from the Atlantic to the Mediterranean of  $0.0055 \pm 0.0050 \text{ PgC yr}^{-1}$ , inferred as the residual between an accumulation rate of  $0.018 \text{ PgC yr}^{-1}$  and  $\text{C}_{\text{ant}}$  uptake from the atmosphere at a rate of  $-0.012 \text{ PgC yr}^{-1}$  (Figure S11 in Supporting Information S1). This inferred  $\text{C}_{\text{ant}}$  import to the Mediterranean Sea is consistent with the observation-based transport estimates in the Strait of Gibraltar of  $0.0042 \pm 0.0010 \text{ PgC yr}^{-1}$  (Huertas et al., 2009).

From estimates of the exchange between the Nordic Seas and the Arctic (Jeansson et al., 2011) and the exchange from the Nares Strait, a net flux to the Arctic of  $0.02 \pm 0.01 \text{ PgC yr}^{-1}$  was estimated. Thus, the remaining lateral transport rates between the different biomes were estimated for each GOBM as the difference between surface flux, interior accumulation and the boundary fluxes. From the average of the GOBM results (lateral  $\text{C}_{\text{ant}}$  transport and  $\text{F}_{\text{ant}}$ , Figure S11 in Supporting Information S1), the average transport from the Southern Ocean is obtained (Figure 8).

For the Atlantic, the northward transport of  $\text{C}_{\text{ant}}$  from the Southern Ocean decreases northward to almost zero (Figure S11 in Supporting Information S1) or reverses sign at the boundary between the NA STSS and NA SPSS biomes. These  $\text{C}_{\text{ant}}$  transports are fully compatible with the AMOC, in which the upper branch transports more



$C_{\text{ant}}$  northward than the southward lower branch, and also with the decrease of the vertical gradient of  $C_{\text{ant}}$  northward such that in the NA SPSS biome the vertical gradient of  $C_{\text{ant}}$  is small (Figure S10 in Supporting Information S1). With these results, the net transports of  $0.163 \pm 0.057 \text{ PgC yr}^{-1}$  at the South Atlantic boundary obtained from the GOBM results are consistent with recent transports estimated from ocean sections at  $30^{\circ}\text{S}$  of  $0.186 \pm 0.019 \text{ PgC yr}^{-1}$  (Cainzos et al., 2022). This suggests that the weak anthropogenic sea-air  $\text{CO}_2$  fluxes are the primary cause of low  $\Delta C_{\text{ant}}$  in the South Atlantic. The lower  $\Delta C_{\text{ant}}$  in the interior ocean and in particular in the NA STPS and SA STPS biomes suggest that the anthropogenic contribution to the total  $\text{FCO}_2$  in GOBMs is 30% weaker than expected from  $C_{\text{ant}}$  observations in the interior ocean. Although the total  $\text{FCO}_2$  obtained from GOBMs is stronger in the Atlantic than those derived from  $\text{pCO}_2$  products, we note that the total  $\text{FCO}_2$  from the GOBMs contains no RCO correction here. While our estimate of the anthropogenic contribution is unaffected by RCO, this is not true for the  $\text{FCO}_2$  estimates of the GOBMs. If we were to apply the RCO-values based on Regnier et al. (2022) and Lacroix et al. (2020) to the  $\text{FCO}_2$  estimates of the GOBMs, then their total  $\text{FCO}_2$  estimate would be weaker than that derived by the  $\text{pCO}_2$  products.

The inferred northward  $C_{\text{ant}}$  transport at the southern boundary between Atlantic and Southern Oceans obtained for each of the nine GOBMs with Simulations A and D shows a high correlation ( $r^2 = 0.61$ ;  $p\text{-level} < 0.01$ ) with the maximum AMOC values at  $26^{\circ}\text{N}$  of each of these GOBMs (Figure S12 in Supporting Information S1), indicating that the northward physical transport is the main driver of the northward  $C_{\text{ant}}$  transport. We note additionally that, in comparison with observations, the GOBMs tend to underestimate the maximum AMOC values at  $26^{\circ}\text{N}$  (Figure S12 in Supporting Information S1) and hence the inferred northward  $C_{\text{ant}}$  transport (Terhaar et al., 2024).

## 4. Discussion

### 4.1. Progress Since RECCAP1

Over the last decade, the nature of the  $\text{pCO}_2$  products has changed significantly, not only because of a significant growth in the number of observations (Bakker et al., 2016) but also because of the implementation of new data interpolation methodologies (e.g., Denvil-Sommer et al., 2019; Gregor et al., 2019; Rödenbeck et al., 2015), and improvements in the fidelity of predictor variables. This led to RECCAP2 being able to use nine different  $\text{pCO}_2$  products with time-varying  $\text{FCO}_2$  estimates. RECCAP1 only had 2 products available for time-varying Atlantic Ocean  $\text{FCO}_2$  estimates: a multi-parameter regression based on the gridded product of SOCATv1.5 (Pfeil et al., 2013) as well as regional scale  $\text{FCO}_2$  estimates based on the  $\text{pCO}_2$  database analysis of McKinley et al. (2011). Additionally, the climatology of Takahashi et al. (2009) formed a cornerstone of the RECCAP1 studies and has proven to be a very robust product, with estimates close to the climatology obtained from the new  $\text{pCO}_2$  products. On the modeling side, there have also been many relevant advances between RECCAP1 and RECCAP2. In RECCAP1, only six models were used, while RECCAP2 employs almost twice the number of models. A subset of the available GOBMs in RECCAP2 also participated in RECCAP1 and have been improved in both physical and biological processes (e.g., Aumont et al., 2015; Schwinger et al., 2016; Wright et al., 2021), with an enhanced spatial resolution (e.g., from  $\sim 2^{\circ}$  to  $\sim 1^{\circ}$ ), though they remain too coarse to resolve mesoscale processes.

A direct comparison between RECCAP1 and RECCAP2 estimates of the Atlantic cannot be performed due to the appreciable improvements in methods and data coverage. Moreover, the estimates made in RECCAP1 have a different regional domain (spanning from  $44^{\circ}\text{S}$  to  $79^{\circ}\text{N}$ ). The zonal region from  $35^{\circ}\text{S}$  to  $44^{\circ}\text{S}$  is no longer part of the Atlantic Ocean mask in RECCAP2, but instead considered to be part of the Southern Ocean (Hauck, Gregor, et al., 2023). In addition, the time period covered in RECCAP1 spans the years 1990–2009, while RECCAP2 covers the years from 1985 to 2018 and considers not only the whole time-period but also two sub-periods (before and after 2000). In RECCAP1, an estimate of RCO flux component ( $0.17 \pm 0.04 \text{ PgC yr}^{-1}$ ) was subtracted from the  $\text{FCO}_2$  values obtained for GOBMs (see Table S8 in Supporting Information S2 for details), which was not performed here. However, to allow a direct comparison with RECCAP1 (Table S8 in Supporting Information S2), we additionally calculated  $\text{FCO}_2$  averages for the region from  $44^{\circ}\text{S}$  to  $79^{\circ}\text{N}$  for the RECCAP1 time period. To compare the GOBM estimates, we additionally re-added the RCO-flux to the RECCAP1 estimate. Our inferred RECCAP2-results for the  $\text{FCO}_2$  averages are similar to those published in RECCAP1 (Schuster et al., 2013) as the  $\text{FCO}_2$  estimates are within the uncertainty of each other (Table S8 in Supporting Information S2). Yet, the mean  $\text{FCO}_2$ -values increased in both the GOBM ensemble mean (21% increase) and  $\text{pCO}_2$  product ensemble average (9% increase). Both the RECCAP1 as well as the RECCAP2 estimate show a higher average  $\text{FCO}_2$ -value for the GOBMs, yet the mean difference between GOBMs and  $\text{pCO}_2$  products is larger in RECCAP2.

It is difficult to compare estimates of decadal trends between RECCAP1 and RECCAP2, as RECCAP1 only provided upper-bound estimates for the trends based on the pCO<sub>2</sub>-database. The GOBMs of RECCAP1 estimated the largest trends in FCO<sub>2</sub> for the North Subtropics and estimated the trends for the other Atlantic regions to be negligible or very small (Schuster et al., 2013). We cannot confirm this in RECCAP2 (Figure 4) because our trend estimates consider different time-periods and that the IAV could additionally influence the trend-estimates substantially (e.g., Figure 4).

For the IAV in FCO<sub>2</sub>, Schuster et al. (2013) found in RECCAP1 significant but weak correlations to the NAO for the Equatorial biome with opposing signs between GOBMs ( $r = -0.43$ ) and pCO<sub>2</sub> products ( $r = 0.35$ ), potentially relating to their IAV being driven by SST-variations and DIC-variations, respectively. Here, the new definition of the Equatorial biome in RECCAP2 helps to confine the upwelling region such that the IAV in this region is DIC driven in both GOBMs and pCO<sub>2</sub> products, with correlations of  $r = -0.40$  and  $r = -0.47$  with the NAO, respectively. In our case, negative correlations indicate DIC-driven variations as we correlate the NAO with the sea-to-air flux, while Schuster et al. (2013) correlated it with the air-to-sea flux (i.e., flux of opposite sign). However, the issue of the GOBMs being more SST-driven remains also within the IAV of RECCAP2, most notably in the North Atlantic subpolar gyre.

In terms of FCO<sub>2</sub> seasonality, the southern subtropical regions, the equatorial region and the northern subtropics studied in RECCAP1 followed the seasonal increase and decrease of pCO<sub>2</sub> driven mainly by warming and cooling in both GOBMs and observation-based estimates. These general results remain consistent in our analysis. In the NA SPSS, the RECCAP1 pCO<sub>2</sub> products showed that the seasonal cycle is reversed with a minimum during summer and outgassing in winter (Schuster et al., 2013), conforming to direct observations (Olsen et al., 2008), whereas the seasonal cycle of GOBMs was dominated by the temperature component. As the Atlantic regions in RECCAP1 were defined simply via latitudinal boundaries, Schuster et al. (2013) denoted that the temperature controlled seasonal cycle of the GOBMs is likely due to the inclusion of the northern reaches of the subtropical gyre. The refinement of the Atlantic regions in RECCAP2, however, shows that biogeochemical boundaries with a clearer exclusion of the subtropical gyre do not change the temperature control of the seasonal cycle of the GOBMs in the NA SPSS.

Even though RECCAP2 benefits from a substantial increase in observations and improvements in modeling (complexity and resolution), the mean difference between GOBMs and pCO<sub>2</sub> products is larger in RECCAP2 and the disagreements between pCO<sub>2</sub> products and GOBMs in the NA SPSS remain in terms of IAV and seasonal cycle. Potential mechanisms for this are further discussed in Section 4.3.

#### 4.2. The Influence of the Riverine CO<sub>2</sub> Outgassing on Comparisons of the CO<sub>2</sub> Sink in RECCAP2 Models and Observation-Based Products

When averaged over the 1985 to 2018 period, the mean FCO<sub>2</sub> of pCO<sub>2</sub> products and GOBM ensemble agree within the ranges of their ensemble spread for most of the biomes and the Atlantic basin (Table 1). The related spatial distribution of FCO<sub>2</sub> also agrees with respect to the large-scale, basin-wide patterns (Figure 1), although some discrepancies are detected in the NA SPSS biome. The average Atlantic FCO<sub>2</sub> estimated by the GOBM ensemble is 30% lower than the estimate from pCO<sub>2</sub> products.

The riverine carbon outgassing (RCO, see Section 2.4) hampers the comparison of the FCO<sub>2</sub> estimates from the GOBMs with those of pCO<sub>2</sub> products, since the input of riverine carbon and the burial of carbon is treated in various ways across the ensemble of GOBMs. Furthermore, relevant output from the GOBMs is missing to properly assess the contribution of carbon, alkalinity and nutrient input from land and their burial in sediments, resulting in a situation where only a rough approximation of the RCO in the GOBMs is possible (Terhaar et al., 2024). In the RECCAP2 protocol, it was recommended to apply the spatial distribution of the RCO of Lacroix et al. (2020), scaled to a globally integrated RCO value of  $0.65 \pm 0.3 \text{ PgC yr}^{-1}$  (Regnier et al., 2022). This procedure results in a large adjustment of  $0.27 \pm 0.06 \text{ PgC yr}^{-1}$  ( $3.9 \pm 1.0 \text{ mol C m}^{-2} \text{ yr}^{-1}$ ) for the Atlantic sea-air CO<sub>2</sub> flux, which is more than half of the FCO<sub>2</sub> derived from the set of GOBMs and 70% of that estimated from pCO<sub>2</sub> products (in absolute numbers). Although other estimates of the RCO reported by Aumont et al. (2001) and Jacobson et al. (2007) reduce the RCO in the Atlantic by 1/3, the relative magnitude of the RCO compared to the FCO<sub>2</sub> from GOBMs and pCO<sub>2</sub> products remains substantial.

In the Atlantic Ocean, the difference between the  $\text{FCO}_2$  obtained from the ensemble  $\text{pCO}_2$  products and the ensemble of GOBMs is  $0.10 \pm 0.11 \text{ PgC yr}^{-1}$ . The RCO derived from Aumont et al. (2001), Jacobson et al. (2007), and Lacroix et al. (2020) scaled up to Regnier et al. (2022) are  $0.16 \pm 0.05$ ,  $0.16 \pm 0.04$ , and  $0.27 \pm 0.06 \text{ PgC yr}^{-1}$ , respectively (Table S3 in Supporting Information S2), yielding an ensemble average of  $0.20 \pm 0.05 \text{ PgC yr}^{-1}$ . All four of these values are higher than the average difference between the  $\text{pCO}_2$  products and GOBMs, although within the combined uncertainty of all estimates. Importantly, for four of the five biomes (NA SPSS, NA STSS, AEQU, and SA STPS), the ensemble RCO-estimates agree well with the  $\text{FCO}_2$  differences ( $\text{pCO}_2$  products minus GOBMs) with a mean difference of only  $-0.001 \pm 0.019 \text{ PgC yr}^{-1}$  when the ensemble of RCO is added to the GOBMs estimate (last column in Table S3 of the Supporting Information S2).

The biome with the largest discrepancy between  $\text{FCO}_2$  in the GOBMs and in the  $\text{pCO}_2$  products is the NA STPS. Likewise, the three estimates of the RCO diverge most in this biome, indicating a high RCO-uncertainty. The  $\text{FCO}_2$  difference between  $\text{pCO}_2$  products and GOBMs would require an RCO of  $-0.024 \pm 0.013 \text{ PgC yr}^{-1}$  to be balanced, that is, an additional  $\text{CO}_2$  uptake rather than outgassing due to riverine input of carbon. However, this difference is of reversed sign and much lower than the ensemble mean of the direct RCO estimates ( $+0.073 \pm 0.048 \text{ PgC yr}^{-1}$ , Table S3 in Supporting Information S2). This discrepancy would be even larger when the RCO estimate recommended in RECCAP2 ( $+0.126 \pm 0.010 \text{ PgC yr}^{-1}$ ) was used. At the same time, the  $\Delta C_{\text{ant}}$  (Table 1) and  $F_{\text{ant}}$  rates of the GOBMs in the NA STPS biome are lower than an observation-based estimate from Zunino et al. (2015), who used DIC measurements along the  $26.5^\circ\text{N}$  and  $7.5^\circ\text{N}$  sections from 1992/93 and 2010/11, and inferred a  $F_{\text{ant}}$  of  $-0.23 \pm 0.02 \text{ PgC yr}^{-1}$  over an area of  $15.3 \times 10^{12} \text{ m}^2$  (70% of NA STPS), which is more than twice the estimated  $F_{\text{ant}}$  in the GOBMs ( $-0.084 \pm 0.010 \text{ PgC yr}^{-1}$ , Figure S11 in Supporting Information S1). The  $F_{\text{ant}}$  difference between the GOBMs and the observation-based estimate from Zunino et al. (2015) is very similar to the difference between the direct RCO estimate ( $+0.126 \pm 0.040 \text{ PgC yr}^{-1}$ ) and the residual between the  $\text{FCO}_2$  from  $\text{pCO}_2$  products and GOBMs ( $-0.024 \pm 0.013 \text{ PgC yr}^{-1}$ ). This agreement in the differences suggests that the GOBMs indeed underestimate the  $C_{\text{ant}}$  uptake in the NA STPS biome. If the GOBMs simulated a substantially stronger  $C_{\text{ant}}$  uptake (by about  $-0.1 \text{ PgC yr}^{-1}$ ), then the direct RCO estimate would plausibly explain the  $\text{FCO}_2$  difference between the GOBMs and  $\text{pCO}_2$  products, albeit with large uncertainty. The likely underestimation of  $F_{\text{ant}}$  by the GOBMs in the NA STPS biome is further supported by their  $\Delta C_{\text{ant}}$  that is only about half as large as the observation-based estimate from Gruber et al. (2019) (Table 1), as well as the lower northward  $C_{\text{ant}}$  transport compared to two observation-based estimates (Brown et al., 2021; Cainzos et al., 2022).

### 4.3. Temporal Variability in Sea-Air $\text{CO}_2$ Fluxes in Models and $\text{pCO}_2$ Products

In the results section, differences between models and  $\text{pCO}_2$  products in sea-air  $\text{CO}_2$ -flux dynamics are described in terms of trends, seasonality and interannual variability. The region where the GOBMs and  $\text{pCO}_2$  products show the largest discrepancies is the NA SPSS: the biome with the highest  $\text{CO}_2$  uptake rates. When looking at the seasonal decomposition of surface  $\text{pCO}_2$ , it becomes clear that the seasonality is primarily temperature-driven in the GOBMs so that their  $\text{CO}_2$  uptake is larger in winter than in summer because of the seasonal SST changes. The seasonal cycle of the  $\text{pCO}_2$  products is driven by DIC variations (for more information see Figure S4 in Supporting Information S1; Rodgers et al., 2023). In the North Atlantic subpolar gyre, direct observations of interannual variability in winter  $\text{pCO}_2$  have shown that this is associated with variations in mixed layer depths in this season (Fröb et al., 2019). That means that more intense mixing during colder winters leads to higher surface DIC and consequently higher  $\text{pCO}_2$ , and thus a reduced flux of  $\text{CO}_2$  into the ocean. A DIC-driven dynamic is supported by the seasonal cycle of the  $\text{pCO}_2$  products and the strong, negative correlation of the  $\text{pCO}_2$  product between the IAV of  $\text{CO}_2$  flux and SST in this region (Figure S8a in Supporting Information S1). On the other hand, the GOBMs simulate positive correlations between the IAV of  $\text{CO}_2$  flux and SST. Hence the disagreement between  $\text{pCO}_2$  products and GOBMs in IAV and seasonal cycle is interconnected and driven by the same cause: SST-driven temporal variations in the GOBMs versus DIC-driven temporal variations in the  $\text{pCO}_2$  products. We note that the NA SPSS is also the region in which  $\text{pCO}_2$  products and GOBMs have the largest disagreement in their mean  $\text{CO}_2$ -fluxes and the largest uncertainty in their  $\text{CO}_2$ -trends (see Figures 1 and 4).

When looking for the underlying causes for the disagreement in seasonal driving forces and IAV between  $\text{pCO}_2$  products and GOBMs in the NA SPSS, we find that most of the GOBMs for which the simulated AMOC is available show significant correlations between their IAV of  $\text{CO}_2$  fluxes in the NA SPSS and AMOC-variations with correlation between 0.37 and 0.62. Further, using Earth System Models, Goris et al. (2023) showed that the AMOC-strength drives the simulated seasonal variability in the North Atlantic. Altogether, this suggests that the

underestimation of the AMOC in the GOBMs (Terhaar et al., 2024) could be an underlying cause for the underestimation of the role of biogeochemical variability for both IAV and seasonality by the GOBMs in the NA SPSS.

Furthermore, we identify that the comparatively small DIC variations (as seen in both seasonal cycle and IAV) in the GOBMs might also be a consequence of their current simplified set-up, or the total lack, of riverine carbon fluxes (Terhaar et al., 2024). According to Aumont et al. (2001) and Gao et al. (2023), the contribution of RCO weakens the CO<sub>2</sub> uptake in the NA subpolar gyre and in the Southern Ocean. In fact, applying the predicted riverine carbon outgassing of Aumont et al. (2001) to the NA SPSS biome removes the difference in FCO<sub>2</sub> mean fluxes (1985–2018) between pCO<sub>2</sub> products and GOBMs (Table S3 in Supporting Information S2). The RCO modeled by Aumont et al. (2001) also shows a similarity (in numbers) to the mean FCO<sub>2</sub> differences (1985–2018) between GOBMs and pCO<sub>2</sub> products in the NA STSS, AEQU, and SA STPS biomes (Table S3 in Supporting Information S2). The study of Aumont et al. (2001) highlights the importance of the slow reactivity of dissolved organic carbon (DOC) supplied by rivers to the regional distribution of RCO, which hence might also contribute significantly to seasonal and interannual variability.

Finally, the different strengths of drivers and the resulting large disagreements in IAV between GOBMs and pCO<sub>2</sub> products may leave an imprint on the calculated trends of the sea-air CO<sub>2</sub>-fluxes of the NA SPSS biome for the period 2001–2018 (Figure 4). Here, the pCO<sub>2</sub> products show an accelerated trend for the period 2001–2018 which is not simulated by the GOBMs. Similarly, the IAV of the pCO<sub>2</sub> products is in a positive phase in the year 2000 and in a negative phase in the year 2018 in the NA SPSS (Figure 6), which is not the case for the GOBMs. While this behavior is especially pronounced in the NA SPSS, the NA STPS biome shows a similar phasing in its IAV when comparing GOBMs and pCO<sub>2</sub> products. In a previous study (McKinley et al., 2020), it was found that the IAV is a potential driver of differences in trends between observational products and GOBMs.

While the IAV has an influence on the decadal trends, it cannot solely explain that the calculated trends of sea-air CO<sub>2</sub> fluxes before and after the year 2000 are similar across our ensemble of GOBMs, while the trends obtained from surface CO<sub>2</sub> observations show a sharp increase between the trends of the pre- and post-2000. We advise caution when comparing the CO<sub>2</sub> trends before the year 2000 between GOBMs and pCO<sub>2</sub> products, as the trends of the pCO<sub>2</sub> products are strongly conditioned by the FCO<sub>2</sub> estimates in the early years (Figure S2 in Supporting Information S1), where the available observations (pCO<sub>2</sub> data and predictors) to generate the pCO<sub>2</sub> products are far less, such that the estimates of the pCO<sub>2</sub> products agree less than in later years (Figure 3; Figure S2 in Supporting Information S1). In fact, the pCO<sub>2</sub> products do not agree on the CO<sub>2</sub> trends before the year 2000 ( $-0.024 \pm 0.075$  PgC-yr<sup>-1</sup> dec<sup>-1</sup>) with three pCO<sub>2</sub> products suggesting a weakening of the CO<sub>2</sub> uptake in the Atlantic before 2000 and four pCO<sub>2</sub> products a strengthening (Table S5 in Supporting Information S2). For the trends after the year 2000, the agreement of the pCO<sub>2</sub> products allows for a more confident estimate of a strengthening CO<sub>2</sub> sink in the Atlantic with a trend of  $-0.126 \pm 0.031$  PgC-yr<sup>-1</sup> dec<sup>-1</sup>, which is twice the trend estimated by the GOBMs, of  $-0.060 \pm 0.017$  PgC-yr<sup>-1</sup> dec<sup>-1</sup>. Nevertheless, by using one of the pCO<sub>2</sub> products (MPI-SOM-FFN) in a model, it has been shown that a bias in sampling locations influences the trends and an optimal sampling strategy reduces the negative trend estimate in the northern hemisphere for the years 2000–2018 (Hauck, Nissen, et al., 2023). Hence, a skewed sampling strategy could potentially influence the 2000–2018 trend estimate of the pCO<sub>2</sub> products. For the GOBMs, we want to note that their simulated seasonal cycle might lead to a trend estimate that is too low, as it has been shown for an ensemble of Earth System Models that a more SST-driven seasonal cycle is related to a shallower MLD and a less vivid AMOC (Goris et al., 2018, 2023). Earth System Models with a weaker AMOC simulate more warming and less future carbon uptake in the North Atlantic. Contrarily, a biology-driven seasonal cycle will lead to enhanced carbon uptake due to the increasing sensitivity of pCO<sub>2</sub> to DIC variations with declining buffer capacity of the ocean (Hauck & Völker, 2015).

#### 4.4. C<sub>ant</sub> Storage and Transport

In the Atlantic, the GOBM ensemble C<sub>ant</sub> accumulation rate (1994–2007) is  $28\% \pm 20\%$  lower than the observation-based estimate of Gruber et al. (2019). In general, both GOBMs and the Gruber et al. (2019) product show maximum C<sub>ant</sub> concentrations near the surface with a rapid decrease toward depth. Nevertheless, surface GOBM estimates are in general slightly lower than the observation-based product, which might be related to biases in the Revelle factor caused by too high pre-industrial CO<sub>2</sub> values in a couple of GOBMs with a late starting date past 1765 (Terhaar et al., 2024). The highest agreement between GOBMs and the observation-based product in ΔC<sub>ant</sub> is found north of 30°N, while the GOBMs simulate systematically lower accumulation rates in the South



Atlantic (Figure 7, Table 1). In the upper ocean layer, where the upper limb of the AMOC is located, the differences in  $\Delta C_{\text{ant}}$  are not particularly evident (Figure S10 in Supporting Information S1). However, between 1,400 and 3,000 m depths, GOBMs do not reproduce the  $C_{\text{ant}}$  peak estimated by the observation-based product (Fajar et al., 2015; Gruber et al., 2019; Rhein et al., 2015; Rios et al., 2012) for the Atlantic (Figure S10 in Supporting Information S1) and, more specifically, for the AEQU and SA STPS biomes. This depth interval, with lower  $\Delta C_{\text{ant}}$  in GOBMs compared to the observation-based estimate, coincides with the depth at which the NADW is located. This result suggests that over the 1994–2007 period, the GOBMs simulated too little  $C_{\text{ant}}$  advection into the South Atlantic within the Deep Western Boundary Current that carries the  $C_{\text{ant}}$ -rich NADW toward the Southern Hemisphere (Goris et al., 2023). This interpretation would be consistent with the fact that most of the RECCAP2 GOBMs simulate too weak AMOC strengths (Terhaar et al., 2024). In addition, we note that biased low  $C_{\text{ant}}$  uptake in the Southern Ocean (Hauck, Gregor, et al., 2023), and the subsequent northward transport to the Atlantic, could also contribute to the too-low  $\Delta C_{\text{ant}}$  in the South Atlantic by GOBMs. However, the transport of  $C_{\text{ant}}$  from the Southern Ocean to the Atlantic is in accordance with the observations (Cainzos et al., 2022). We also note that the GOBMs may underestimate the temporal variability of the ocean interior transport, since the  $\Delta C_{\text{ant}}$  of the GOBMs in the South Atlantic are more similar to the estimates by Khatiwala et al. (2009), which assumes a quasi-stationary ocean circulation (see Table 1 for SA STPS biome). In contrast, the GOBMs show a lower decadal variability of the  $\Delta C_{\text{ant}}$  than observation-based products (Gruber et al., 2019; Müller et al., 2023). The interannual variability of the  $\Delta C_{\text{ant}}$ , derived from the linear regressions, is typically  $1.5\% \pm 1.0\%$  of the absolute increase rates across all biomes and the whole Atlantic Ocean, indicating that the  $\Delta C_{\text{ant}}$  in the GOBMs occurs as a rather steady process.

The assessment of  $C_{\text{ant}}$  accumulation and transport in the Atlantic conducted in RECCAP1 (Khatiwala et al., 2013) revealed that the largest anthropogenic  $\text{CO}_2$  uptake occurs in the Southern Ocean, with much of this uptake being transported equatorward through the Antarctic Intermediate Water and Subantarctic Mode Water. Most of this  $C_{\text{ant}}$  is stored in the SA STPS (Mikaloff Fletcher et al., 2006). There is also a significant  $C_{\text{ant}}$  uptake in the tropical Atlantic that is partially transported southward, but most of it is stored in the tropics or transported northward. The  $C_{\text{ant}}$  taken up in the North Atlantic is transported northward in the upper limb of the AMOC and subsequently entrained to the NADW and transported southward in the lower limb of the AMOC. The GOBMs analyzed here confirm these spatial patterns of  $\Delta C_{\text{ant}}$  (though accumulation is low in the South Atlantic below 1,500 m, Figure 7; Figures S9 and S10 in Supporting Information S1) and of meridional transport (dominated by inflow from the Southern Ocean, Figure 8; Figure S11 in Supporting Information S1).

Khatiwala et al. (2013) stated that the  $C_{\text{ant}}$  transports estimated from GO-SHIP sections using hydrographic data and observation-based  $C_{\text{ant}}$  estimates (Álvarez et al., 2003; Holfort et al., 1998; Macdonald et al., 2003; Pérez et al., 2013; Rosón et al., 2003) represent  $C_{\text{ant}}$  transport at a single time point. Such  $C_{\text{ant}}$  transport estimates may be biased because seasonal variability is not resolved (Wilkin et al., 1995). However, recent estimates cover long time series (Brown et al., 2021), or aim to provide decadal climatological estimates (Cainzos et al., 2022). In RECCAP1, Khatiwala et al. (2013) showed that  $C_{\text{ant}}$  transports, obtained based on GOBMs and from hydrographic sections, exhibit similar  $C_{\text{ant}}$  transports between  $44^\circ\text{S}$  and the Equator with a northward transport of  $0.15\text{--}0.20 \text{ PgC yr}^{-1}$ , but, in contrast, in the North Atlantic, the GOBMs simulated a gradual northward decrease of the  $C_{\text{ant}}$  transport, reaching zero horizontal net transport between  $35^\circ$  and  $60^\circ\text{N}$ . This pattern is confirmed in RECCAP2 (Figure S11 in Supporting Information S1) with a larger number of GOBMs involved. Estimates of  $C_{\text{ant}}$  transport at  $26^\circ\text{N}$  along transoceanic sections (Brown et al., 2021; Cainzos et al., 2022; Macdonald et al., 2003; Pérez et al., 2013; Rosón et al., 2003; Zunino et al., 2015) showed larger values than those of the oceanic inversion or GOBMs. These discrepancies remained uncertain in RECCAP1 due to the uncertainties in the hydrographic estimates and the difficulties in directly comparing the two techniques. However, one must also consider the difficulties that inverse models and GOBMs have in representing mesoscale processes, mainly in regions of very intense currents such as the Florida Current, Gulf Stream, and DWBC (Bower et al., 2019; Hirschi et al., 2020; Khatiwala et al., 2013; Ma et al., 2016).

Recent estimates by Brown et al. (2021) using the RAPID long time series (2004–2012), with an assessment of  $C_{\text{ant}}$  transports at 10-day timescale, confirm a strong  $C_{\text{ant}}$  transport at  $26.5^\circ\text{N}$  of  $0.191 \pm 0.013 \text{ PgC yr}^{-1}$ , which is in the middle of the range ( $0.128 \pm 0.032$  to  $0.25 \pm 0.05 \text{ PgC yr}^{-1}$ ) of the eight estimates obtained from five sections between 1992 and 2011 (collected in Cainzos et al. (2022)). The ensemble average  $C_{\text{ant}}$  transport over  $26^\circ\text{N}$  obtained for the nine GOBMs used here is  $0.053 \pm 0.037 \text{ PgC yr}^{-1}$ , which is almost four times lower than the  $C_{\text{ant}}$  transport of Brown et al. (2021). Racapé et al. (2018), using a global NEMO-PISCES model with a



finer spatial resolution ( $0.5^\circ \times 0.5^\circ$ ), obtained a northward transport of  $0.092 \pm 0.04 \text{ PgC yr}^{-1}$  somewhat closer to observation-based estimates, suggesting that the spatial resolution of the GOBMs is relevant for the simulation of ocean interior transport. Observational-based evaluations of  $C_{\text{ant}}$  transport indicate the dynamical difficulties that CMIP5/6 climate models in certain regions have in achieving realistic simulations of the AMOC and DWBC, when run at relatively coarse resolutions on the order of  $1^\circ$  (Hirschi et al., 2020; Ma et al., 2016), which does not allow to correctly simulate vertical structures nor to resolve mesoscale ocean eddies (Bower et al., 2019). For the RECCAP2 GOBMs, it was shown that the AMOC is, on average, underestimated by  $3.1 \pm 5.2 \text{ Sv}$  at  $26.5^\circ\text{N}$ , which can partly explain this discrepancy between GOBMs and observation-based estimates (Terhaar et al., 2024).

The weak  $C_{\text{ant}}$  northward transport in the subtropical region as shown by GOBMs might also be connected to a possible mismatch in  $C_{\text{ant}}$  uptake in the NA STPS biomes (Zunino et al., 2015) described above. Despite the agreement in mean  $\text{FCO}_2$  between  $\text{pCO}_2$  products and GOBMs in the NA STPS, the mismatch between the potentially strong RCO (Table S3 in Supporting Information S2) and the “residual RCO” (difference between GOBMs and  $\text{pCO}_2$  products) further supports that the GOBMs simulate a too low  $C_{\text{ant}}$  uptake (Table S3 in Supporting Information S2) despite the apparent agreement in the net flux. The reduced  $C_{\text{ant}}$  uptake would be conveyed both northward and downward to the ocean interior. In fact, Cainzos et al. (2022) show that the contribution of vertical mixing is somewhat larger than the southward horizontal advection of  $\Delta C_{\text{ant}}$  in the lower limb of AMOC. Therefore, the insufficient incorporation of the RCO in the GOBMs may also result in a lower  $\text{CO}_2$  uptake, and at the same time also generates an excess  $\text{CO}_2$  uptake in the NA SPSS (Aumont et al., 2001; Gao et al., 2023).

#### 4.5. Future Recommendations

Observations of  $\text{pCO}_2$  in the Atlantic Ocean have greatly improved over the past two decades, making it one of the most densely sampled oceans temporally and spatially. However, the surface  $\text{pCO}_2$  observations are highly skewed in space and time, potentially inducing spurious results in the gap-filling algorithms used for estimating  $\text{CO}_2$  fluxes. In fact, even in the well sampled Atlantic, the observations cover less than 10% of all  $1^\circ \times 1^\circ$  by 1-month grid points, requiring the gap filling methods to fill more than 90% of the grid cells. Recent studies with synthetic model data using similar resolution and parameterizations to observations (Gloege et al., 2021; Hauck, Nissen, et al., 2023) indicate that gap-filling methods may be prone to a possible overestimation of the decadal rates of increase in  $\text{CO}_2$  uptake when data are sparse, partially explaining the discrepancy between these products and GOBMs. We also note that in the data-sparse period 1985–2000, the trends generated by the various observation-based products were highly correlated with their flux estimate in 1985. This shows that with reduced observational coverage, the trend in the products tends to drift apart. Therefore, data-coverage as well as gap-filling methods need to be improved to reduce uncertainties in the trends. It is now quite worrisome that key Atlantic ship of opportunity lines for surface ocean  $\text{pCO}_2$  observations have been lost or operated with reduced capacity in the past years—this tendency must be reversed if we want to retain our ability to accurately constrain the Atlantic Ocean  $\text{CO}_2$  sink and its variability. Another aspect is the lack of funding in SOCAT itself, resulting in a longer time lag before collected data gets included in the database ([https://www.ioccp.org/images/Gnews/2023\\_A\\_Case\\_for\\_SOCAT.pdf](https://www.ioccp.org/images/Gnews/2023_A_Case_for_SOCAT.pdf)).

This assessment relies on simple bulk flux formulations used in  $\text{pCO}_2$ -based products and GOBMs to determine  $\text{FCO}_2$  from  $\Delta\text{fCO}_2$  fields with little regard to interfacial processes controlling gas fluxes. Gas transfer is based on a global parameterization with wind speed. Recent advances in direct flux estimates provide the opportunity to use regionally resolved gas transfer estimates (Blomquist et al., 2017; Butterworth & Miller, 2016). Yang et al. (2022) show clear regional variation in the K660-wind speed relationship, which can explain some of the regional differences observed between GOBMs and  $\text{pCO}_2$  products. Near-surface  $\text{CO}_2$  concentration gradient impact fluxes as well as shown herein by applying a cool skin effect. Further improvements in characterization of these gradients will improve the quantification of  $\text{CO}_2$  fluxes (Dong et al., 2022). Of note is that the effect of gas transfer and near-surface gradients will be less in GOBMs than  $\text{pCO}_2$  products because of the inherent feedback between fluxes and concentration gradients in GOBMs (Bellenger et al., 2023).

The Atlantic Ocean is characterized by high temporal dynamics not only in the surface layer but also in the deep layers connecting the North Atlantic to the Southern Ocean through the deep western boundary current. This involves strong mesoscale and sub-mesoscale dynamic currents and structures. The effectiveness of GOBMs in representing dynamic climate change processes is highly dependent on their spatial and temporal resolutions.

Current spatial resolution can barely reproduce the dynamics of strong CO<sub>2</sub> transport in the Atlantic, as well as ocean-coastal interactions.

A number of future model improvements could further address or minimize the discrepancies in the interior C<sub>ant</sub> inventory estimates. As simulations of the ocean biogeochemistry are strongly constrained by the performance of the physical model (Doney et al., 2004), more detailed assessments should be carried out of key physical dynamics that govern the surface to deep carbon transport, such as the representation of mode water and intermediate and deep waters in the North Atlantic (Racape et al., 2018). Assessment of GOBMs' ability to simulate observed episodic ventilation events and their impact on interior C<sub>ant</sub>, for example, as documented in Rhein et al. (2017) and Fröb et al. (2016), could shed additional light on their validity. Through winter convective mixing, biases in the interior carbon chemistry can influence the upper ocean carbon uptake capacity in models due to biases in the buffering capacity of the ocean (Terhaar et al., 2022; Vaithinada Ayar et al., 2022). Improvements in the representation of mixing by the models would likely also alleviate the issues with the simulated amplitude and timing of spring bloom and winter remineralization in the subpolar region (that we identified as key deficiencies in GOBMs) and further improve their FCO<sub>2</sub> seasonal cycle. Better observational constraints and improvement in mixing parameterizations are needed to alleviate this issue. Higher spatial resolution is likely necessary to improve key upper ocean physical features in the Atlantic Ocean, such as the Gulf Stream (Chassignet et al., 2020), which has been shown to play a significant role in constraining the seasonality and trends of North Atlantic carbon fluxes and interior sequestration (Goris et al., 2023). Results from the high-resolution regional model (ROMS-ETHZ) indicate a better representation of the FCO<sub>2</sub> seasonal cycle in the NA SPSS and a better representation of the trends for 2001–2018 in NA SPSS, NA STSS and SA STPS, while we see no improvement or even a worse representation in other regions. A detailed and overarching investigation of the benefits of higher resolution for the carbon cycle would be desirable. Further, as the number of observations continues to increase, improvements in biogeochemical parameterizations can be achieved through data assimilation, for example, to address the regionally heterogeneous biological processes (Gharamti et al., 2017; Tjiputra et al., 2007). In addition, improvement in biological model complexity may be needed to optimally reproduce the observed biogeochemical dynamics across spatially varying regimes such as the Atlantic basin (Gehlen et al., 2015). The interior lateral transport of C<sub>ant</sub> is projected to play an increasing role in the future (Tjiputra et al., 2010). Better constraints of the northward (and southward) transport of anthropogenic CO<sub>2</sub> in the ocean, through the upper (and lower) limb of the AMOC should be considered to improve estimates of fluxes further north (Cainzos et al., 2022). Finally, an improved model experiment protocol that includes a multi-centennial pre-industrial spin up (Séférian et al., 2020), common initialization procedure, and implementation of the river carbon loop should be considered (see also Terhaar et al., 2024).

In the North Atlantic, Fontela et al. (2020) showed that semi-refractory DOC mineralization in the lower limb of AMOC represents a significant contribution to DIC of the same order of magnitude as CO<sub>2</sub> exchange with the atmosphere, resulting in a possible CO<sub>2</sub> source that could explain the differences observed between the observed FCO<sub>2</sub> (Takahashi et al., 2009) and those estimated by inverse methods (Gerber et al., 2009; Gruber et al., 2009; Mikaloff Fletcher et al., 2007). In RECCAP, the role of DOC has not been evaluated, nor the double impact of its seasonal cycle, that is, diverting DIC which reduces pCO<sub>2</sub> in summer or by DOC deep mineralization, increasing DIC transport. Semi-refractory DOC is exported to the mesopelagic zone and even deeper depths in the North Atlantic, as documented by Hansell (2013), who estimated ~0.34 PgC yr<sup>-1</sup> DOC export, with a mineralization time scale to CO<sub>2</sub> of decades. In the North Atlantic, the coupling between DOC production and export is revealed in the export of locally produced DOC (Fernandez-Castro et al., 2019; Roshan & DeVries, 2017). In fact, the carbon sequestration mediated by DOC has been shown to represent around a third of the North Atlantic CO<sub>2</sub> sink (Fontela et al., 2016). It has been demonstrated in DOC enrichment along the AMOC and its coupling with intense overturning in the NA SPSS leads to downward transport of 0.07 PgC yr<sup>-1</sup> associated mainly with water masses transported by the DWBC (Fontela et al., 2020). In addition, 0.09 PgC yr<sup>-1</sup> of DOC exported northward from the subtropics is mineralized in the deep layers of the AMOC. Inverse models do not include the DOC divergence, which is assumed to be small (Mikaloff Fletcher et al., 2007). This carbon cycle component has not been evaluated neither in RECCAP1 nor in RECCAP2, and considering the importance of its magnitude relative to FCO<sub>2</sub>, it is relevant to consider it in future biogeochemical modeling experiments together with other modeling improvements proposed here. Articles highlighting the importance of DOC in the carbon balance are relatively recent (Fontela et al., 2016, 2020), with global non-seasonal climatology (Roshan & DeVries, 2017) and the compilation of a global DOC database (Hansell et al., 2021) being very recent, making it difficult to assess DOC modeling in GOBMs.

## 5. Conclusions

We provide here the current “best estimate” of surface CO<sub>2</sub> fluxes as well as the accumulation and transport of C<sub>ant</sub> in the Atlantic, including the Mediterranean Sea for the RECCAP2 period, 1985–2018. For this estimate, we have compared different types of ocean biogeochemical models (GOBMs, ROBM, data-assimilated models) with various observation-based products. Our analysis includes several time-scales of variability.

We find that the *mean* net sea-air CO<sub>2</sub> flux of the GOBM ensemble is 27% stronger than estimates from observation-based pCO<sub>2</sub> products. This difference is within the uncertainties of the GOBMs and pCO<sub>2</sub> products and can be explained, in part, by known discrepancies between pCO<sub>2</sub> products and GOBMs. Specifically, this includes the oceanic CO<sub>2</sub> outgassing due to the impact of riverine discharge that is not explicitly represented in most GOBMs. The pCO<sub>2</sub> products may also be biased by not including near surface pCO<sub>2</sub> gradients. Adjusting for these effects mostly leads to higher fluxes into the ocean, which—if applied to all pCO<sub>2</sub> products—would lead to better agreement between GOBMs and pCO<sub>2</sub> products for the time period considered here.

The *trends* of sea-air CO<sub>2</sub> fluxes before and after the year 2000 are similar across our ensemble of GOBMs (from  $-0.045 \pm 0.012$  to  $-0.060 \pm 0.017$  PgC yr<sup>-1</sup> dec<sup>-1</sup>) and are consistent with the 43% increase in the atmospheric CO<sub>2</sub> growth rate between the pre-2000 period and the post-2000 periods. In contrast, the trends obtained from surface CO<sub>2</sub> observations show a sharp increase from the trend of the pre-2000 of  $-0.024 \pm 0.075$  PgC yr<sup>-1</sup> dec<sup>-1</sup> to a trend of  $-0.126 \pm 0.031$  PgC yr<sup>-1</sup> dec<sup>-1</sup> in the post-2000 period.

All biomes apart from the subpolar North Atlantic show a high correlation between GOBMs and pCO<sub>2</sub> products in terms of FCO<sub>2</sub> seasonality. In the North Atlantic subpolar biome, the GOBMs simulate a seasonal cycle driven predominantly by temperature variation, which the pCO<sub>2</sub> products do not show.

Averaged over the Atlantic, the ensemble of GOBMs shows lower interannual variability (IAV) in FCO<sub>2</sub> than the pCO<sub>2</sub> products. Spatially and temporally, IAV in pCO<sub>2</sub> products and GOBMs agree well in most of the Atlantic biomes but disagree quite substantially in the subpolar North Atlantic. Here, the variability of the GOBMs is mostly driven by SST variations, which is not the case for the pCO<sub>2</sub> products.

The mean C<sub>ant</sub> storage change between 1994 and 2007 simulated by the GOBM ensemble was found to be 28% lower than that estimated from DIC observations in the ocean interior and 25% lower than the data-assimilated model. These differences are higher than the standard deviation of the GOBM estimates (17%). In contrast to the results described for the surface CO<sub>2</sub> fluxes, there is a high agreement in anthropogenic CO<sub>2</sub> storage rates between GOBMs and those based on DIC observations in the NA SPSS and NA STSS biomes, whereas there are significant differences in the NA STPS, AEQU, and SA STPS biomes, where the GOBM estimates are on average 36% lower than observation-based estimates. The GOBMs indicate that 32% of the C<sub>ant</sub> accumulated in the Atlantic comes from the Southern Ocean, in line with previous estimates from the literature. The Mediterranean Sea revealed an almost balanced net sea-air flux of CO<sub>2</sub>; however, it presented a C<sub>ant</sub> accumulation of 0.018 PgC yr<sup>-1</sup>, of which 70% are taken up from the atmosphere and 30% are imported from the Atlantic.

Estimates of the land-to-ocean transport of carbon and nutrients indicate a significant and large net CO<sub>2</sub> outgassing due to the input of this terrestrially derived matter. The protocol of RECCAP2 recommended the use of the updated estimate of 0.65 PgC yr<sup>-1</sup> of Regnier et al. (2022) at the global level. For the Atlantic Ocean, the outgassing rates per square meter are twice the global rates when considering the spatial distribution of the riverine carbon outgassing (RCO) simulated by Lacroix et al. (2020). This RCO is especially significant in the NA STPS biome and hampers the comparison of GOBM and observation-based estimates of CO<sub>2</sub> fluxes, transport and accumulation. Therefore, it is essential to have more realistic models to better understand the influences of land-sea fluxes in the Atlantic Ocean and to be able to use observational-estimates with confidence when determining the accumulation of C<sub>ant</sub>. This also requires better spatial and seasonal coverage of biogeochemical observations such as CO<sub>2</sub>, nutrients and DOC to allow for improved model evaluation or even generate new emergent constraints.

## Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

## Data Availability Statement

The RECCAP2 ocean data collection can be found in Müller (2023).

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