

1 **Recent trends and variability in the oceanic storage of**
2 **dissolved inorganic carbon**

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11 **Key Points:**

- 12 • From 2004 through 2019, the global oceanic dissolved inorganic carbon (DIC)
13 pool increased at an average rate of 3.2 ± 0.7 Pg C yr⁻¹
14 • Most of this increase is associated with the uptake of anthropogenic CO₂,
15 while natural CO₂ is mostly redistributed within the ocean
16 • The interannual variability of DIC is largest in the tropical Pacific Ocean

Abstract

Several methods have been developed to quantify the oceanic accumulation of anthropogenic carbon dioxide (CO₂) in response to rising atmospheric CO₂. Yet, we still lack a corresponding estimate of the changes in the total oceanic dissolved inorganic carbon (DIC). In addition to the increase in anthropogenic CO₂, changes in DIC also include alterations of natural CO₂. Once integrated globally, changes in DIC reflect the net oceanic sink for atmospheric CO₂, complementary to estimates of the air-sea CO₂ exchange based on surface measurements. Here, we extend the MOBO-DIC machine learning approach by Keppler et al. (2020a) to estimate global monthly fields of DIC at 1° resolution over the top 1500 m from 2004 through 2019. We find that over these 16 years and extrapolated to cover the whole global ocean down to 4000 m, the oceanic DIC pool increased close to linearly at an average rate of 3.2±0.7 Pg C yr⁻¹. This trend is statistically indistinguishable from current estimates of the oceanic uptake of anthropogenic CO₂ over the same period. Thus, our study implies no detectable net loss or gain of natural CO₂ by the ocean, albeit the large uncertainties could be masking it. Our reconstructions suggest substantial internal redistributions of natural oceanic CO₂, with a shift from the mid-latitudes to the tropics and from the surface to below ~200 m. Such redistributions correspond with the Pacific Decadal Oscillation and the Atlantic Multidecadal Oscillation. The interannual variability of DIC is strongest in the tropical Western Pacific, consistent with the El Niño Southern Oscillation.

1 Introduction

The global oceanic dissolved inorganic carbon (DIC) pool is a powerful recorder of changes in the net exchange of carbon dioxide (CO₂) across the air-sea interface, i.e., the strength of the net oceanic carbon sink. This net sink is the sum of a flux of natural carbon that reflects the exchange driven by changes in solubility, ocean circulation, mixing, and biological processes, and the flux of anthropogenic carbon that corresponds to the anomalous flux of CO₂ driven by the human-induced rise in atmospheric CO₂ (McNeil & Matear, 2013; Gruber et al., 2023). When integrated globally, the sources and sinks of natural CO₂ fluxes cancel each other out in a steady state as the ocean strives towards equilibrium with the overlaying atmosphere (Landschützer et al., 2022). On the contrary, the observed increase in the net air-sea CO₂ exchange is caused by anthropogenic CO₂ emissions, (Friedlingstein et al., 2022). An important exception is residual outgassing that reflects the balance between the input of carbon by rivers and the deposition of carbon on the seafloor (Sarmiento & Sundquist, 1992; Regnier et al., 2022). As long as this balance is maintained, this latter (i.e., natural) component does not leave an imprint on changes in DIC, so that changes in this pool are then directly attributable to the ocean interior accumulation or loss of both natural and anthropogenic CO₂.

Knowing the magnitude of the net oceanic sink for CO₂ is crucial for closing the global carbon budget and its anthropogenic perturbation (Sarmiento & Gruber, 2002; Friedlingstein et al., 2022). The need is heightened by efforts such as the United Nations' global stocktake efforts (<https://unfccc.int/topics/science/workstreams/global-stocktake>), which require a more refined estimate of the changing ocean carbon content, connecting the surface and interior ocean, and demonstrating the total changes in DIC, as well as its spatial distribution. Finally, better global-scale constraints on the changes in oceanic DIC are of great interest to better document the progression of ocean acidification and better establish the connection between changes in seawater chemistry and biological impacts (Doney et al., 2009; Orr et al., 2005; Feely et al., 2004).

In terms of observations, the net oceanic CO₂ sink is at present primarily determined using observations of the surface ocean partial pressure of CO₂ (pCO₂), which are mapped

67 to the globe using various data interpolation methods (Landschützer et al., 2014; Rödenbeck
68 et al., 2015; Fay et al., 2021; Gregor & Gruber, 2021). The mapped $p\text{CO}_2$ is then used,
69 in combination with the atmospheric $p\text{CO}_2$ and the gas transfer velocity, to estimate the
70 air-sea CO_2 flux. However, this approach is subject to various uncertainties, such as data
71 sparsity (Fay & McKinley, 2013), an ill-constrained gas transfer coefficient (Wanninkhof
72 et al., 2009; Roobaert et al., 2019), and a potential offset in the $p\text{CO}_2$ measurements as
73 they are not directly taken at the cool skin surface (Watson et al., 2020). Furthermore,
74 the steady-state outgassing of river-derived carbon needs to be subtracted from the inferred
75 flux to obtain the anthropogenic flux relevant to the global carbon budget. Estimates for
76 this riverine flux range from $0.23 \text{ Pg C yr}^{-1}$ (Lacroix et al., 2020) and $0.45 \pm 0.18 \text{ Pg C yr}^{-1}$
77 (Jacobson et al., 2007) to $0.78 \pm 0.41 \text{ Pg C yr}^{-1}$ (Resplandy et al., 2018), with the most
78 recent review by Regnier et al. (2022) suggesting a value of $0.65 \pm 0.3 \text{ Pg C yr}^{-1}$. This range
79 and the associated uncertainties add further uncertainty to the $p\text{CO}_2$ derived estimates of
80 the net carbon uptake by the global ocean.

81 Confidence in quantifying this net uptake could be strengthened if constrained
82 independently through the direct determination of changes in the global ocean DIC
83 content. Nevertheless, this is a challenging task, owing to the sparsity of observations, the
84 substantial background DIC pool of $\sim 37,000 \text{ Pg C}$ (Keppler et al., 2020b), and the many
85 physical and biological processes that govern the distribution of DIC in the ocean
86 (Sarmiento & Gruber, 2006). A very successful approach to overcome this challenge has
87 been to only focus on the interior ocean's accumulation of the anthropogenic CO_2 (C_{ant})
88 component (Wallace, 1995; Tanhua et al., 2007). Under the assumption that either the
89 ocean is in a near steady state or that the contribution of natural carbon (C_{nat}) to
90 global-scale changes in DIC is small, several methods have been developed to determine
91 the changes in C_{ant} either from single surveys of DIC (Brewer, 1978; Chen & Millero,
92 1979; Gruber et al., 1996), or from repeat hydrography programs (Friis et al., 2005;
93 Clement & Gruber, 2018; Carter et al., 2019). The application of these methods has
94 permitted the oceanographic community to quantify the increase in the C_{ant} inventory,
95 both since preindustrial times (Gruber, 1998; Sabine et al., 1999) and for the past few
96 decades (Friis et al., 2005; Wanninkhof et al., 2010; Carter et al., 2019), with the global
97 studies providing invaluable constraints for the global budget of C_{ant} (Sabine et al., 2004;
98 Gruber et al., 2019).

99 Although these global C_{ant} estimates have proven to be extremely valuable for
100 constraining the global carbon budget and hence the fate of the emitted anthropogenic
101 CO_2 , they have not been able to fully address whether the steady-state assumption or the
102 assumption of a small natural CO_2 signal is justified. Questions were raised early on,
103 especially in the context of ocean warming (Keeling, 2005; Sabine & Gruber, 2005), which
104 many models suggest will lead to a loss of CO_2 from the ocean (Joos et al., 1999;
105 Sarmiento et al., 1998; Matear & Hirst, 1999). Later, using a combination of different
106 model and observation-based methods, McNeil and Matear (2013) invoked the presence of
107 a decadal-scale outgassing signal of natural CO_2 , but without being able to support this
108 conjecture with direct observations. Dedicated modeling studies also suggest that the
109 ocean might have lost natural CO_2 in recent decades, e.g., in response to the trends in the
110 Southern Annular Mode (Le Quéré et al., 2007; Lenton & Matear, 2007; Zickfeld et al.,
111 2007; Hauck et al., 2013; Lovenduski et al., 2008, 2007). In their global study on the
112 increase in anthropogenic CO_2 between 1994 and 2007, Gruber et al. (2019) speculated
113 that perhaps as much as 5 Pg C of natural CO_2 might have been lost from the ocean over
114 this period. Conversely, enhanced lateral transport of natural carbon from the land could
115 yield a gain of C_{nat} in the ocean (Regnier et al., 2022). Similarly, changes in the
116 circulation or biological productivity could cause an anomalous uptake or release of CO_2
117 from the atmosphere, altering the total stock of C_{nat} .

118 As the arguments for potential changes in C_{nat} accumulate, the need to constrain
119 the changes in the total DIC pool increases, as this permits to assess the changes in both

120 natural and anthropogenic CO₂. When doing so, one needs to also consider that even if
121 the global-scale changes in the natural CO₂ pool might be small, this pool is subject to
122 strong redistributions within the ocean, associated with changes in circulation, shifts in
123 ocean fronts, and changes in biological productivity, causing locally large changes in DIC
124 (Clement & Gruber, 2018). Such changes are commonly seen when comparing the DIC
125 distributions between two occupations of a particular hydrographic section (Wanninkhof
126 et al., 2010; Carter et al., 2019). They are also expected in the context of interannual
127 variability, especially in regions with large vertical undulations of the thermocline, and
128 hence also the “carbocline,” i.e., the strong vertical gradient in DIC. Such redistributions
129 of DIC within the ocean not only pose a challenge for the detection of global-scale changes
130 in the DIC pool, but they also potentially threaten organisms, as spatial redistributions of
131 DIC might cause more rapid local changes in ocean acidification and, perhaps, a more rapid
132 reaching of critical thresholds (McNeil & Sasse, 2016).

133 Currently, no sensor technology exists that can operationally measure DIC in situ.
134 Thus, we must rely on physical seawater samples collected and analyzed during ship-based
135 surveys and programs (Talley et al., 2016; Bates et al., 2014), strongly limiting the coverage
136 and the sampling frequency. Most of these DIC measurements and the associated ancillary
137 data are compiled and subjected to secondary quality control by the Global Ocean Data
138 Analysis Project (GLODAP; Olsen et al. (2016); Key et al. (2004)). A recent version
139 (GLODAPv2.2021) contains over one million measurements from across the global ocean,
140 spanning measurements from 1972 to 2020 (Lauvset et al., 2021). Most of the measurements
141 contained within GLODAP stem from repeat hydrography programs, where the same set
142 of stations along long lines are revisited at roughly decadal intervals (Talley et al., 2016).
143 In addition to GLODAP, some long-term time-series stations provide information on the
144 temporal variability in the interior ocean at a few locations, including the Hawaii Ocean
145 Timeseries (HOT; Dore et al. (2009)) and the Bermuda Atlantic Timeseries Study (BATS;
146 Bates et al. (2014)). More recently, Argo floats equipped with biogeochemical (BGC) sensors
147 that measure pH, salinity, and other variables, supplement the ship data. Using these float
148 measurements and some empirical relationships to infer alkalinity, DIC can be estimated
149 (Carter et al., 2018; van Heuven et al., 2011). However, this method has much larger
150 uncertainties than the ship data (Bittig et al., 2018), and to date, the available BGC-Argo
151 float data are largely limited to the Southern Ocean, as part of the Southern Ocean Carbon
152 and Climate Observations and Modelling project (SOCCOM, Talley et al. (2019)), while
153 the global ocean BGC-Argo array is still in its early stages (Bittig et al., 2019).

154 In parallel to the efforts in combining and unifying carbon cycle observations (Olsen
155 et al., 2016; Bakker et al., 2016), a second branch related to big data analysis based on
156 machine learning has emerged. Keppler et al. (2020b) adopted a cluster-regression approach
157 previously applied to reconstruct the air-sea CO₂ exchange (Landschützer et al., 2013, 2014)
158 and extended it to map a monthly climatology of DIC in the upper 2000 m of the near-global
159 ocean, i.e., Mapped Observation-Based Oceanic DIC (MOBO-DIC, Keppler et al. (2020a)).
160 Similarly, Broullón et al. (2020) developed a single-step machine learning approach to map
161 the monthly climatology of interior DIC at a global scale. In addition, a recent study has
162 mapped out the temporal evolution of DIC globally (Gregor & Gruber, 2021), but this
163 approach was limited to the documentation of variations at the sea surface. These studies
164 revealed the feasibility of reconstructing the DIC content from observations at the global
165 scale. In addition, using CMIP6 models and synthetic Argo data, Turner et al. (2022)
166 demonstrated very recently that interior temperature and salinity data are well suited to
167 reconstruct interior DIC fields and their variability. However, they have not yet mapped
168 the interior ocean DIC with real-world Argo observations. Further, Sharp et al. (2022)
169 successfully mapped monthly fields of interior ocean dissolved oxygen at a global scale,
170 using a machine learning approach. However, mapped estimates of interior observation-
171 based DIC remain limited to seasonal climatologies (Keppler et al., 2020b; Broullón et
172 al., 2020) or the surface (Gregor & Gruber, 2021), and reconstructions of the trend and

173 interannual variability of the upper ocean total DIC at the global scale based on direct
174 observations are still lacking.

175 To fill this gap, we use the MOBO-DIC approach and extend the monthly climatology of
176 DIC by Keppler et al. (2020b) to resolve monthly global DIC fields from 2004 through 2019
177 (i.e., January 2004 through December 2019). The temporal extent of our reconstructions
178 is primarily determined by the availability of temperature and salinity fields from the Argo
179 program that we use as key predictors. Our new DIC product is mapped at a monthly
180 resolution on a $1^\circ \times 1^\circ$ grid, from 65°N to 65°S , and reaching 80°N in the Atlantic (see
181 Supporting Information Fig. S1), extending from 2.5 m to 1500 m depth. Subsequently,
182 we investigate the trend and interannual variability of the interior oceanic DIC at a global
183 scale and put these changes into the context of the ongoing accumulation of anthropogenic
184 CO_2 in the ocean's interior and from this, infer the changes in the natural CO_2 pool.

185 2 Data and Methods

186 2.1 Cluster-regression

187 We adopt the two-step neural network MOBO-DIC approach introduced by Keppler
188 et al. (2020b) to map the sparse DIC observations to the (near) global ocean at monthly
189 resolution for the period January 2004 through December 2019. Here, we present a
190 summary of the most important features and the main changes compared to the
191 climatological approach taken by Keppler et al. (2020b). Our approach first clusters the
192 ocean into regions of similar properties using self-organizing maps (SOM) and then applies
193 a feed-forward neural network (FFN) in each cluster to reconstruct a physical relationship
194 between a set of driver variables and the target DIC data. This cluster-regression approach
195 does not require information about the measurement location, a feature that separates it
196 from many other mapping approaches (Sasse et al., 2013; Gregor et al., 2017; Bittig et al.,
197 2018; Broullón et al., 2019, 2020). Thus, our regression method is solely based on the
198 physical and biogeochemical relations between the predictor and target variables. Not
199 using the measurement location as a predictor permits our method to benefit from
200 information obtained in other places within each cluster, where predictor and target data
201 are similarly related. Due to data availability and the presence of different processes near
202 the surface and below, we run the method separately for two depth slabs: from 2.5 m to
203 500 m and from 500 m to 1500 m. We take the mean of the two estimates at 500 m to
204 minimize boundary problems between the two depth slabs. This approach does not
205 eliminate all discontinuities, but they are well within the uncertainty limits of the method.

206 In the first step, we use a SOM, i.e., a type of unsupervised machine learning, to
207 determine clusters. Following Keppler et al. (2020b), we use six clusters in the upper 500
208 m and four between 500 m and 1500 m. This number of clusters leads to the smallest
209 overall error in the DIC reconstruction. To avoid boundary problems inherent in cluster-
210 regression approaches, we adjust the original method by creating an ensemble of SOM
211 clusters, following the approach introduced by Gregor and Gruber (2021). To this end, we
212 performed the SOM-step three times, where the DIC input has a different weight ranging
213 from 2 to 4 in each run. The resulting SOM clusters vary mostly around the boundaries (see
214 Supporting Information Fig. S2). In the second step, we run an FFN for each SOM cluster.
215 Our FFN network architecture consists of 8 neurons in the hidden layer of the FFN, as this
216 setup results in the most robust output based on a comparison between the mapped output
217 and the original training data.

218 To avoid overfitting, we use 80% of the input data to train the network and the
219 remaining 20% for internal cross-validation. As the training and validation data are
220 separated randomly, the output from the FFN is slightly different each time it is run. For
221 each SOM setup, we run the FFN five times, where each time, the data is separated
222 differently into training and validation data to create an ensemble of outputs. Thus, our

223 ensemble comprises 15 members (three SOM setups, each with five FFN runs). The final
224 reported data are the mean across the ensemble, and the standard deviation across the
225 ensemble (hereafter ensemble spread) represents the uncertainty linked to the weighting of
226 the SOM clusters and the random assignment of training and validation data. We smooth
227 the ensemble mean fields at each depth level by taking the running mean with a window
228 size of three grid cells in each horizontal direction (latitude and longitude) and in the
229 temporal dimension.

230 Some runs produced outputs with unlikely values, e.g., considerably larger or smaller
231 than the measured variables in GLODAP. We attribute this to the random assignment of
232 training and validation data, where some data subsets are unsuitable for training. Such runs
233 with unlikely values occurred both with the GLODAP training data and with synthetic data,
234 so it cannot be attributed to noise in the observations. We have tried many different setups
235 of the network to eliminate this issue. However, with the current training data, we were
236 unable to resolve it. Thus, when an output results in values that are more than 5 standard
237 deviations larger or smaller than the observed data in GLODAP (i.e., outside of the range
238 1639 to 2575 $\mu\text{mol kg}^{-1}$ and 1898 to 2629 $\mu\text{mol kg}^{-1}$, for the upper and lower depth slab,
239 respectively), the entire ensemble member was discarded and re-run with the same setup,
240 but with a different sub-set of training data. We trust that removing the runs with unlikely
241 values, in addition to the bootstrapping approach, yields a robust estimate.

242 2.2 Data and Domain

243 As input to the SOM, we use monthly mapped fields of temperature and salinity based
244 on Argo float measurements (Roemmich & Gilson, 2009) and an annual-mean climatology
245 of DIC (Lauvset et al., 2016). We weigh the DIC input stronger than the physical predictors
246 so that the clusters largely represent the climatological mean DIC and, to a lesser extent,
247 the physical water masses, following Landschützer et al. (2013).

248 For the FFN step, we use the ship measurements of DIC from GLODAPv2.2021 between
249 January 2004 and December 2019 (Lauvset et al., 2021) as the target data. We only retain
250 GLODAP data with a WOCE quality control of 0 or 2 and a secondary quality control flag
251 of 1. As predictors, we use the same Argo-based temperature and salinity fields that we
252 used during the SOM step. In addition, we use monthly climatologies of mapped dissolved
253 oxygen, nitrate, and silicic acid from the World Ocean Atlas 2018 (WOA18; Boyer et al.
254 (2018)). These climatologies are based on ship measurements from 1955 through 2017 and
255 were interpolated to the global ocean using optimal interpolation. As the nitrate and silicic
256 acid from WOA only extend until 500 m, they were not used as predictors in the deeper
257 slab, while dissolved oxygen extends to 1500 m in WOA and is thus a predictor variable in
258 both depth slabs. Deviating from the approach taken to produce the monthly climatology
259 of MOBO-DIC (Keppler et al., 2020b), we use atmospheric pCO_2 as an additional predictor
260 in the upper depth slab (0 to 500 m) to be able to represent the long-term trend in the
261 atmospheric CO_2 concentration. Atmospheric pCO_2 at each grid cell was computed from the
262 GlobalView marine boundary layer product of the mole fraction of CO_2 ($x\text{CO}_2$; GlobalView-
263 CO_2 (2011)) and converted to pCO_2 following Landschützer et al. (2013). In the deeper
264 slab below 500 m, we use Julian days as a predictor to represent any long-term trend in
265 the data. Thus, the predictors between the surface and 500 m are temperature, salinity,
266 dissolved oxygen, nitrate, silicic acid, and atmospheric pCO_2 . Between 500 m and 1500
267 m, we use temperature, salinity, dissolved oxygen, and Julian day as predictors. A more
268 detailed discussion on the choice of predictors can be found in Keppler et al. (2020b).

269 Note that we use the mapped monthly mean fields as predictors, as opposed to the
270 co-measured data from GLODAP during the training step of the FFN. We tested both
271 approaches but found the results were very noisy when using the co-measured data as
272 predictors. This noisy output may be partially caused by the WOA monthly gridded fields
273 being smoother than the point measurements in GLODAP. Furthermore, using the co-

274 measured predictors leads to a substantial loss of training data, as in $\sim 60,000$ data points out
 275 of $\sim 440,000$ (i.e., $\sim 14\%$), and the training data do not have usable co-measured predictors.

276 The availability of the data limits the domain and resolution of our mapping approach.
 277 For example, we limit the vertical extent of the multi-year product here to 1500 m (as
 278 opposed to 2000 m used for the MOBO-DIC climatology) as the DIC observations are
 279 very sparse below 1500 m and only temperature and salinity are available as physical or
 280 biogeochemical predictors there. This lack of predictors below 1500 m prevents a robust
 281 estimate of the DIC variations and trends at these depths. Temporally and spatially, the
 282 limits tend to be set by the predictor data. The Argo-based data products used here extend
 283 from 65°N to 65°S globally, to 80°N in the Atlantic Ocean, with shallow coastal regions
 284 being masked, marking the horizontal extent of our domain. As the mapped Argo-dataset
 285 starts in 2004, and GLODAPv2.2021 includes cruise data until January 2020, the temporal
 286 extent of MOBO-DIC is from January 2004 through December 2019.

287 All predictors have a monthly resolution on $1^\circ \times 1^\circ$ grids, and we interpolate them onto
 288 28 uneven depth levels between 2.5 m and 1500 m. Note that due to an update to the Argo
 289 data, the domain of this study is slightly larger than in the monthly climatology of MOBO-
 290 DIC (Keppeler et al., 2020b): it extends further north in the Atlantic (until 80°N instead of
 291 65°N), and some more coastal and shallow regions are included (see Supporting Information
 292 Fig. S1). As the domain covers most of the global ocean, we refer to our domain as global
 293 in-text but want to note that it is technically only near-global.

294 **2.3 Calculation of the trend and inventory changes**

295 We estimate the trend in DIC over our period based on the slope of a linear regression
 296 of the deseasonalized DIC at each grid cell. The data were deseasonalized by applying a
 297 12-month running mean at each grid cell. To calculate the trends in the inventories, we first
 298 normalize DIC for salinity (hereafter sDIC) to remove any effects from potential changes in
 299 the salinity, following Friis et al. (2003). For this normalization, we use the same monthly
 300 Argo-based salinity product as above (Roemmich & Gilson, 2009), using the temporal mean
 301 salinity from 2004 through 2019 at each grid cell as reference salinity. We convert sDIC from
 302 gravimetric (unit: $\mu\text{mol kg}^{-1}$) to volumetric (unit: $\mu\text{mol m}^{-3}$), and then vertically integrate
 303 the volumetric trend in the whole domain (upper 1500 m). To estimate the uncertainty in the
 304 trend, we calculate it with each of the 15 ensemble members and take the standard deviation
 305 range as the uncertainty range. Note that the uncertainty of the trend only includes the
 306 ensemble spread (i.e., the prediction uncertainty) and does not consider other sources of
 307 error, for example, those associated with measurements or representation uncertainty. We
 308 trust that there should not be a trend in measurement or representation uncertainty in
 309 the data, yielding a robust estimate of the overall trend uncertainty. We then conduct an
 310 upscaling to estimate the global changes in sDIC that includes regions beyond our domain,
 311 i.e., the high latitudes, coastal regions, and below 1500 m (see Supporting Information S3).

312 **2.4 Comparison with C_{ant}**

313 We compare the trend in MOBO-DIC with an estimate of the change in anthropogenic
 314 CO_2 (ΔC_{ant}). For this comparison, we use two estimates of C_{ant} and scale them to our
 315 study period. The two estimates are (i) the total change in C_{ant} between 1800 and 2007
 316 and (ii) the change in C_{ant} between 1994 and 2007. The former is estimated by adding the
 317 total C_{ant} up to 1994 estimated by Sabine et al. (2004) to the change in C_{ant} between 1994
 318 and 2007 estimated by Gruber et al. (2019). For the latter, we use the ΔC_{ant} by Gruber et
 319 al. (2019).

320 To scale C_{ant} to our period, we apply the transient steady-state approach described by
 321 Mikaloff Fletcher et al. (2006) and Gruber et al. (2019), which relies on the assumption that
 322 the change in C_{ant} scales with the change in atmospheric CO_2 :

$$\Delta C_{ant}^{t_3-t_2} = \alpha(t_0, t_1, t_2, t_3) \cdot \Delta C_{ant}^{t_1-t_0} \quad (1)$$

where t_0 and t_1 are the bounds of the periods used to determine ΔC_{ant} (either 1800 through 2006 or 1994 through 2006) and t_2 and t_3 bound the period to which the scaling should be applied to (here: 2004 through 2019). The scaling factor α is specific for each pair of periods, i.e., is a function of t_0, t_1, t_2 , and t_3 , and can be estimated from the relative changes in atmospheric CO₂, also considering changes in the Revelle factor (Sarmiento et al., 1995) and the changes in the air-sea disequilibrium (Gruber et al., 1996; Matsumoto & Gruber, 2005):

$$\alpha(t_0, t_1, t_2, t_3) = \frac{\Delta_t pCO_2^{atm}(t_3 - t_2)}{\Delta_t pCO_2^{atm}(t_1 - t_0)} \cdot \frac{\xi(t_2..t_3)}{\xi(t_0..t_1)} \cdot \frac{\gamma(t_2..t_3)}{\gamma(t_0..t_1)} \quad (2)$$

where the first factor on the right-hand side is the relative change in atmospheric CO₂, the second factor is the relative change in the disequilibrium ξ , and the third factor is the relative change in the Revelle factor γ .

In the first case, i.e., for the base period 1800 through 2006, inserting the observed values in atmospheric CO₂ in the respective years ($t_0 = 1800$, 280 ppm; $t_1 = 2006$, 381 ppm; $t_2 = 2004$; 377 ppm, $t_3 = 2019$, 410 ppm) gives a value of 0.33 for the first factor on the right-hand side of Eq. 2. For the disequilibrium, we take the same estimate Gruber et al. (2019) used when scaling from 1800 through 1993 to 1994 through 2006. They estimated a change in the disequilibrium of about 6 μatm between 1800 and 1994, and about 3 μatm between 1994 and 2007, yielding a ratio of 0.94. Similarly, we also take the estimate by Gruber et al. (2019) of 0.94 for the third factor, i.e., the ratio of the Revelle factors. Entering these three ratios, we obtain an overall scaling factor α of 0.29 ($0.33 \cdot 0.94 \cdot 0.94$) when comparing the period 1800 to 2007 with the period from 2004 through 2019.

In the second case, the base period for the scaling factor goes from 1994 (t_0 , 358 ppm) through 2006 (t_1 , 381 ppm), yielding a relative change in atmospheric CO₂ of 1.45 compared to the period 2004 (t_2 , 377 ppm) through 2019 (t_3 , 410 ppm). As the two periods are largely overlapping in this case, we assume that the ratio of the disequilibrium and the ratio of the Revelle factors are very close to 1 and thus set their values to 1, yielding an overall scaling factor α of 1.45 when comparing the period 1994 to 2007 with the period from 2004 through 2019.

This scaling is based on many assumptions, especially the assumption of a transient steady-state. While the large-scale distribution of C_{ant} has been demonstrated to follow this prediction rather closely, Gruber et al. (2019) pointed out that the reconstructed distribution of the change in C_{ant} between 1994 and 2007 differs in a few places considerably from that reconstructed for the period up to 1994. In particular, they found a meridional shift in the accumulation within the Atlantic Ocean, with a reduction in the North Atlantic storage being compensated by an increase in the temperate latitudes of the South Atlantic. Using two different base periods, we attempt to quantify the potential impact of such changes on our conclusions. Direct estimates of the accumulation of C_{ant} over the same period as analyzed here would be preferable to our scaling approach but are not published to date.

3 Uncertainty Assessment

3.1 Calculation of the overall uncertainty

We identify three main sources of uncertainties that contribute to the total uncertainty of our DIC estimate, following Gregor and Gruber (2021): the uncertainties linked to the measurements (M), the representation (R), and the prediction (P). The overall uncertainty of our DIC estimate (DIC_{err}) can then be estimated with standard error propagation:

$$DIC_{err} = \sqrt{M^2 + R^2 + P^2} \quad (3)$$

366 The uncertainty M linked to the measurements stems from sampling errors and
 367 imprecisions in the measurement system. While GLODAP currently does not report
 368 measurement uncertainties, they include a measure of spatial consistency based on
 369 inter-cruise comparisons, which amounts to $2.4 \mu\text{mol kg}^{-1}$ for DIC (Lauvset et al., 2021).
 370 We assume that this uncertainty is the same at all grid points.

371 The representation uncertainty R results from the fact that the discrete measurements
 372 in GLODAPv2.2021 that are used as target data to train the network are taken at one point
 373 in time and space and thus might not represent the true monthly mean of the $1^\circ \times 1^\circ$ grid cell
 374 and the depth bin it falls in. Especially problematic are regions where the spatiotemporal
 375 variability is high and the number of observations in a grid cell and depth bin is low. It is
 376 not straightforward to quantify the representation error as this requires full knowledge of the
 377 spatiotemporal variability of DIC. Gregor and Gruber (2021) estimated the representation
 378 error of total alkalinity of about $16 \mu\text{mol kg}^{-1}$ at the sea surface of the open ocean. As
 379 the density and spatial distribution of total alkalinity and DIC measurements in GLODAP
 380 are similar, and regions with high spatiotemporal variability in total alkalinity tend to be
 381 regions of high variability in DIC as well, we adopt this estimate for all grid cells and depth
 382 bins. The representation error is expected to be larger near the coast than in the open ocean
 383 due to more variability near the coasts and is also expected to decrease with depth (Torres
 384 et al., 2021), adding some uncertainty to our uncertainty estimate.

385 The prediction uncertainty P represents how well our method can map DIC in time and
 386 space. We take the standard deviation across the 15-member ensemble of our bootstrapping
 387 approach as our estimate of the prediction error. The differences in the ensemble members
 388 are linked to both the ensemble of SOM clusters and the different subsets of training and
 389 validation data, as described in Section 2.1. The global mean prediction uncertainty is 7
 390 $\mu\text{mol kg}^{-1}$, but with a large spread. We find the highest prediction error in the northern
 391 Indian Ocean (up to $\sim 80 \mu\text{mol kg}^{-1}$), where the observations are particularly sparse and
 392 where our estimate is heavily extrapolated (Supporting Information Fig. S3). Such large
 393 local uncertainties illustrate that our approach can reconstruct global fields, but care must
 394 be taken when evaluating regional changes, as the uncertainties on a regional level are quite
 395 high. Combining the three uncertainty contributions (Eq. 3) yields an overall global mean
 396 uncertainty of $18 \mu\text{mol kg}^{-1}$.

397 3.2 Quality of fit

398 During our mapping approach, we estimate the target data at all grid points. Thus,
 399 unlike in an interpolation, there is a difference between the target data (i.e.,
 400 GLODAPv2.2021) and the mapped estimate (i.e., MOBO-DIC). In Supporting
 401 Information Fig. S4, we present these residuals to get a better handle on the quality of our
 402 fits. This analysis intends to examine if there are any systematic offsets in different regions
 403 of depth levels. It also highlights the magnitude of the differences between the training
 404 data and MOBO-DIC. We find that there is no systematic under- or overestimation
 405 compared to the training data, and the global mean bias cancels out to be 0, while the
 406 global mean root mean square difference (RMSD) is $16 \mu\text{mol kg}^{-1}$ (see Table 1 and
 407 Supporting Information Fig. S4), slightly less than our global mean uncertainty of $18 \mu\text{mol}$
 408 kg^{-1} .

409 4 Evaluation

410 We evaluate the quality of the mapped MOBO-DIC product with various independent
 411 observations and using a synthetic dataset derived from a model for which we know the

412 true value. Independence means here that these data were not used for the training of
 413 MOBO-DIC.

414 To evaluate our method with the synthetic data, we subsample the simulated DIC in the
 415 biogeochemical component of the Ocean General Circulation Model HAMOCC (Ilyina et al.,
 416 2013; Mauritsen et al., 2019) when and where we have observations in GLODAPv2.2021. We
 417 then run our MOBO-DIC method with these synthetic data to reconstruct the simulated DIC
 418 fields. We can then compare our reconstructed fields with the actual DIC in HAMOCC. For a
 419 more detailed description of the evaluation with synthetic data, see Supporting Information
 420 S6.1.

421 For the observations, we use three different sources: First, we use a suite of mapped
 422 DIC climatologies, all of which are based on GLODAP data (Lauvset et al., 2016; Keppler et
 423 al., 2020b; Broullón et al., 2020). Second, we compare MOBO-DIC with observations from
 424 time-series stations and biogeochemical Argo floats. Third, we use the mapped surface DIC
 425 product contained in OceanSODA-ETHZ (Gregor & Gruber, 2021), allowing us to compare
 426 the monthly $1^\circ \times 1^\circ$ fields at the surface when and where the two datasets overlap (January
 427 2004 to December 2018).

428 We first evaluate the climatological mean, then the trend, and the interannual
 429 variability, for each of these evaluation data where the temporal resolution allows. Note
 430 that we use DIC and not sDIC in the evaluation with observations, as salinity is not
 431 always co-measured, and using monthly 1-degree gridded salinity fields could introduce
 432 errors. In contrast, our comparison of synthetic MOBO-DIC and the HAMOCC model
 433 uses sDIC, as here we have both salinity and DIC as monthly 1-degree gridded fields. For
 434 a more in-depth analysis of the evaluation, see Supporting Information S6, and for a
 435 summary, see Table 1.

436 4.1 Evaluation of climatological mean

437 The evaluation of the MOBO-DIC method with the synthetic data from HAMOCC
 438 illustrates that the method is well-equipped to reconstruct the mean DIC fields in HAMOCC
 439 well (see Table 1). MOBO-DIC reconstructs the simulated climatological mean DIC fields
 440 with a negligible bias of $-1 \mu\text{mol kg}^{-1}$ and with an RMSD of $12 \mu\text{mol kg}^{-1}$.

441 The evaluation with the observational climatological constraints also reveals good
 442 performance of MOBO-DIC. The mean differences relative to the MOBO-DIC, Lauvset,
 443 and Broullón climatologies, are between 7 and $11 \mu\text{mol kg}^{-1}$, with an RMSD of 17 to 20
 444 $\mu\text{mol kg}^{-1}$. This is within the combined uncertainties of the MOBO-DIC and the
 445 comparison data sets (see Table 1 and Supporting Information Fig. S5 and S6). It also
 446 needs to be noted that the Lauvset climatology is normalized to the year 2002, while the
 447 Broullón monthly climatology is normalized to 1995, and the MOBO-DIC monthly
 448 climatology by Keppler et al. (2020b) is centered around the years 2010/2011. In
 449 comparison, this study is centered around 2012. Thus, the positive biases compared to
 450 these climatologies may largely stem from the differences in the period and the increase in
 451 anthropogenic carbon in the ocean.

452 The bias between MOBO-DIC and the comparison data sets from time-series stations
 453 and floats ranges from -5 to $16 \mu\text{mol kg}^{-1}$. As the biases are both positive and negative,
 454 there is no indication of MOBO-DIC having a systematic bias towards over/underestimating
 455 the global carbon content (see Table 1 and Supporting Information Fig. S9 and S10). The
 456 RMSD between MOBO-DIC and these data range from $14 \mu\text{mol kg}^{-1}$ for the SOCCOM
 457 floats to $42 \mu\text{mol kg}^{-1}$ for Drake Passage but are mostly between 15 and $20 \mu\text{mol kg}^{-1}$,
 458 i.e., comparable to the mean global uncertainty of MOBO-DIC ($18 \mu\text{mol kg}^{-1}$). In all cases
 459 except for Drake Passage, the RMSD is within the combined uncertainties of MOBO-DIC at
 460 the location of the compared data set and the uncertainty of the compared data set, using
 461 standard error propagation. The disagreement at Drake Passage, a well-observed time-series

Table 1. Summary of the bias and RMSD between MOBO-DIC and the comparison data sets. Also displayed are the mean uncertainty of MOBO-DIC at the time and location of the compared data set and the uncertainty of the comparison data sets.

Compared data set	Type of data	Bias ($\mu\text{mol kg}^{-1}$)	RMSD ($\mu\text{mol kg}^{-1}$)	MOBO-DIC uncertainty ($\mu\text{mol kg}^{-1}$)	Comparison uncertainty ($\mu\text{mol kg}^{-1}$)
GLODAPv2.2021	Ship data, without interpolation or mapping (used to train the network)	0	16	18	2
Lauvset climatology	Global climatology (optimal interpolation)	7	17	18	7
Broullón climatology	Global monthly climatology (single-step neural network)	10	17	18	N/A
MOBO-DIC _{clim}	Global monthly climatology (cluster-regression)	11	20	18	9
HAMOCC	Synthetic data	-1	12	18	N/A
BATS	Time-series station	12	17	17	2
HOT	Time-series station	-4	15	17	2
Drake Passage	Time-series station (surface)	16	42	18	1
SOCCOM floats	Calculated DIC from BGC floats (pH) with LIAR algorithm	-5	14	17	6
OceanSODA-ETHZ	Global surface estimate (cluster-regression)	4	15	18	21

462 station, is associated with large local variabilities that are not captured in MOBO-DIC and
 463 are further discussed in Section 4.3.

464 Comparing MOBO-DIC at the surface with the surface DIC from OceanSODA-ETHZ,
 465 we find that the magnitude and spatial patterns of the mean DIC agree very well, considering
 466 they are based on independent data (SOCAT pCO₂ vs. GLODAP DIC; Bakker et al. (2016);
 467 Lauvset et al. (2021)). The global mean RMSD between the two data sets is 15 $\mu\text{mol kg}^{-1}$,
 468 and there is a mean bias of approximately 4 $\mu\text{mol kg}^{-1}$, which is well within the uncertainties
 469 (see Table 1 and Supporting Information Fig. S11 a-c).

470 4.2 Evaluation of trends

471 Our synthetic MOBO-DIC generally reconstructs both the spatial distribution and
 472 magnitude of the trend of sDIC in HAMOCC well, with no indication of a systemic over-
 473 or underestimation of the trend (see Supporting Information Fig. S7). An exception is the
 474 deep eastern equatorial Pacific, where MOBO-DIC overestimates the trend. This could be
 475 the result of overfitting or of challenges of MOBO-DIC to fit the trends in a region with
 476 very large lateral gradients and where data coverage is intermittent. We do not see similarly
 477 large trends in the reconstructions with observations, possibly because the observed lateral
 478 gradients are smaller than those in the model. Still, this mismatch in the synthetic data
 479 suggests that the MOBO-DIC reconstructed trends are likely somewhat less robust than the
 480 climatologies and that care must be taken to avoid an overinterpretation of the results.

481 The sDIC trends at the time-series stations are comparable to MOBO-DIC at the times
 482 and locations of these independent observations (see Table 2 and Supporting Information
 483 Fig. S9). For example, we observe a mean trend in the water column at BATS of 7 $\mu\text{mol kg}^{-1}$
 484 decade⁻¹, while the mean trend in the water column in MOBO-DIC at the grid cell closest
 485 to BATS is $5 \pm 2 \mu\text{mol kg}^{-1} \text{ decade}^{-1}$. However, some quantitative differences exist, with
 486 the largest difference in the trend found at depths between 600 and 800 m at BATS. There,
 487 MOBO-DIC, with an estimated trend of only $5 \pm 2 \mu\text{mol kg}^{-1} \text{ decade}^{-1}$ underestimates the

488 observed trend of $16 \mu\text{mol kg}^{-1} \text{ decade}^{-1}$ quite substantially. With BATS being one of
 489 the best-constrained time-series sites, the observed trend is very robust. The trend is much
 490 better reconstructed in the shallower waters at BATS, indicating that this is not a general
 491 issue but an issue specifically associated with the intermediate water masses in the North
 492 Atlantic.

493 MOBO-DIC also underestimates the trend seen in the biogeochemical Argo floats in
 494 the Southern Ocean (SOCCOM floats) between 20 and 40 m (see Table 2 and Supporting
 495 Information Fig. S10). The observed trend is $-20 \mu\text{mol kg}^{-1} \text{ decade}^{-1}$, while the trend
 496 estimate in MOBO-DIC at the same grid cells is only $-9 \pm 2 \mu\text{mol kg}^{-1} \text{ decade}^{-1}$. There is
 497 a known difference between ship-based DIC measurements and DIC derived from float pH
 498 measurements (Gray et al., 2018). However, this offset is thought to be relatively steady and
 499 should not affect the trend. Nevertheless, the time series is short and spatially sparse, so
 500 it is not entirely clear whether the issue is with MOBO-DIC or with the SOCCOM-derived
 501 DIC trend.

502 The trend of MOBO-DIC at the surface has a similar spatial distribution but is
 503 slightly smaller than the trend of the mapped surface DIC from Gregor and Gruber
 504 (2021), with a global mean trend between January 2004 and December 2018 of 0.6 ± 0.1
 505 $\mu\text{mol kg}^{-1} \text{ yr}^{-1}$ and $0.8 \mu\text{mol kg}^{-1} \text{ yr}^{-1}$, respectively (see Supporting Information Fig.
 506 S11 d-f). As OceanSODA-ETHZ is based on considerably more surface measurements
 507 than MOBO-DIC, it is likely that MOBO-DIC slightly underestimates the trend of the
 508 surface DIC.

509 4.3 Evaluation of Interannual variability

510 Similar to the trend, our synthetic MOBO-DIC reconstructs the spatial distribution
 511 and magnitude of the interannual variability, defined here as the standard deviation across
 512 the ensemble, of sDIC in HAMOCC well (see Supporting Information Fig. S8). However,
 513 we also find an artifact in the deep eastern equatorial Pacific, i.e., the same region where
 514 we had difficulties with the trend. There, the interannual variability is too large in the
 515 synthetic MOBO-DIC reconstruction. Again, no such artifact exists in the MOBO-DIC
 516 reconstructions with observations, but smaller artifacts cannot be ruled out.

517 MOBO-DIC tends to underestimate the observed interannual variability of sDIC at the
 518 time-series stations and the locations of the SOCCOM floats (see Table 2 and Supporting
 519 Information Figs. S9 and S10). The biggest difference in the interannual variability is
 520 between 20 and 40 m at HOT, where MOBO-DIC estimates a variability of only $4 \mu\text{mol}$
 521 kg^{-1} , while the observations suggest a value of $11 \mu\text{mol kg}^{-1}$. As above, such differences
 522 can be at least partially explained by the observations containing a lot of noise and not
 523 necessarily being representative of the mean monthly 1° fields. At Drake Passage, the
 524 comparison data displays considerably more variability than our gridded product and may
 525 include outliers. Thus, there are instances where the discrepancies between MOBO-DIC and
 526 the comparison data sets are beyond the uncertainty limits. We expect that this is mostly
 527 due to large local variabilities that are smoothed out in the monthly mean $1^\circ \times 1^\circ$ fields in
 528 MOBO-DIC.

529 The interannual variability of MOBO-DIC at the surface also has a similar distribution
 530 and is slightly smaller than the interannual variability of the mapped surface DIC from
 531 Gregor and Gruber (2021) (see Table 2 and Supporting Information Fig. S11 g-i). Here, we
 532 observe global mean standard deviations of 3 and $4 \mu\text{mol kg}^{-1}$, respectively (see Supporting
 533 Information Fig. S11 g-i). An explanation for their slightly higher variability could lie
 534 in the fact that OceanSODA-ETHZ uses satellite-based sea surface temperature (SST) as a
 535 predictor while we use float data for temperature and salinity. Satellite-based SST estimates
 536 are known to display more variability than float-based estimates (Roemmich & Gilson, 2009).
 537 Further, OceanSODA-ETHZ has less interannual variability in pCO_2 than other surface

Table 2. Comparison of the trends (in $\mu\text{mol kg}^{-1} \text{ decade}^{-1}$) and interannual variability (IAV, in $\mu\text{mol kg}^{-1}$), defined as the standard deviation in time (seasonal cycle and trend removed), from independent DIC estimates, and from MOBO-DIC at the time and locations of the independent data. Due to data sparsity in the observational data, we average the fields over depth slabs (20 to 40 m, 100 to 150 m, 600 to 800 m). The locations of the Stations are illustrated in Supporting Information Fig. S1.

	Compared data set →	BATS	MOBO-DIC at BATS	HOT	MOBO-DIC at HOT	Drake Passage (surface)	MOBO-DIC at Drake Passage	SOCCOM floats	MOBO-DIC at SOCCOM floats
Trend	Depth ↓								
	20 – 40 m	1	7	5	2	8	1	-20	-9
	100 – 150 m	3	8	13	6	N/A	N/A	3	1
	600 -800 m	16	5	4	5	N/A	N/A	19	26
IAV	20 – 40 m	5	2	11	4	9	5	4	3
	100 – 150 m	4	2	6	2	N/A	N/A	2	2
	600 -800 m	4	1	3	1	N/A	N/A	3	3

538 products such as SOM-FFN by Landschützer et al. (2016). Thus, the available evidence
 539 suggests that MOBO-DIC tends to underestimate the interannual variability.

540 5 Results and Discussion

541 5.1 Global changes in the DIC inventory

542 The reconstructed (near) global sDIC inventory between 0 and 1500 m increased steadily
 543 from 2004 through 2019, with a total increase of 42 ± 5 Pg C over this period (Fig. 1). All
 544 depth ranges contribute to this trend, with $\sim 16\%$ of the increase in sDIC having occurred
 545 in the upper 150 m, 18% between 150 and 300 m, 38% between 300 and 700 m, and 28%
 546 between 700 and 1500 m. Superimposed onto this strong positive trend, we observe the
 547 effect of the seasonal cycle on the total inventory (order of ~ 2 Pg C), some interannual
 548 variations, and a weakening of the trend in the second half of the record, most strongly
 549 visible in the deepest depth slice analyzed, i.e., below 700 m.

550 By adding an estimate of the sDIC changes in the shallow coastal regions and the
 551 high latitudes (3 ± 0.4 Pg C) and in the ocean below 1500 m (6 ± 6 Pg C; see Supporting
 552 Information S3), we arrive at a global sDIC inventory change of 51 ± 11 Pg C. Over the
 553 16 years of our analysis, this corresponds to a rate of increase of 3.2 ± 0.7 Pg C yr^{-1} . We
 554 interpret this increase in sDIC to be mostly of atmospheric origin, i.e., reflecting a net uptake
 555 of CO_2 from the atmosphere, although we cannot exclude a small contribution coming from
 556 other sources, such as a trend in the input from rivers and sediment sources, or an imbalance
 557 with the marine organic carbon pool.

558 Our interior ocean data-based net ocean uptake estimate of 3.2 ± 0.7 Pg C yr^{-1} is
 559 comparable with surface pCO_2 observation-based estimates of the net carbon flux from
 560 the atmosphere into the ocean. The latest update of the net air-sea CO_2 flux estimate
 561 by Landschützer et al. (2016), which includes both the open and coastal ocean, suggests a
 562 global uptake of 2.1 ± 0.5 Pg C yr^{-1} from 2004 through 2019. Adding a riverine outgassing
 563 of CO_2 of 0.6 ± 0.4 Pg C yr^{-1} (Friedlingstein et al., 2022; Regnier et al., 2022), these surface
 564 ocean data suggest a net uptake of 2.7 ± 0.6 Pg C yr^{-1} . This is 0.5 Pg C yr^{-1} less than our
 565 estimate based on the increase in ocean interior sDIC but within the uncertainty bounds.
 566 Our estimate would be closer to the surface-based estimates if we used the higher-end

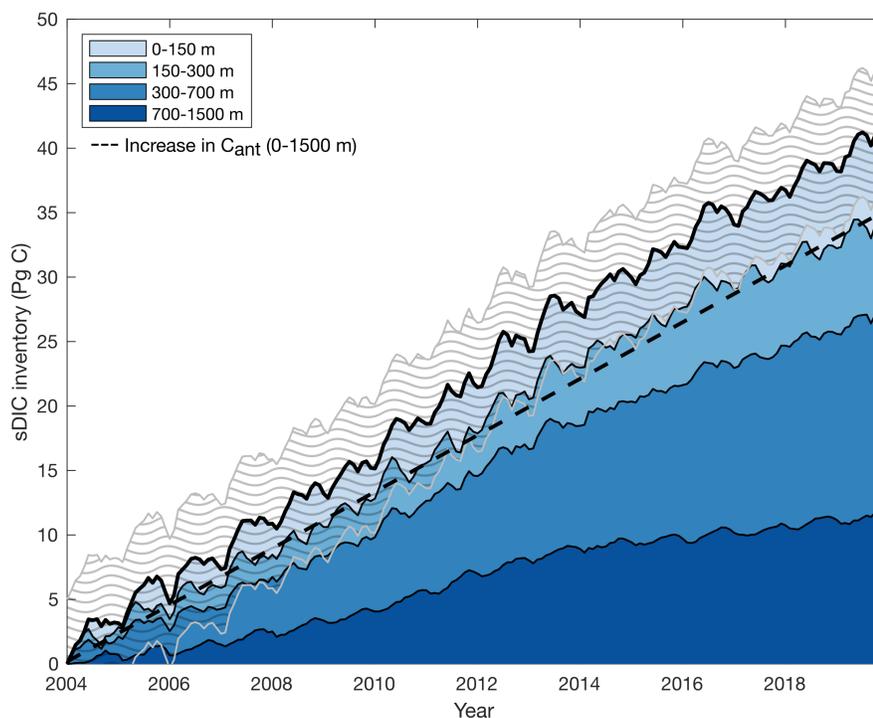


Figure 1. a) Temporal change in the global sDIC inventory derived from MOBO-DIC from 2004 through 2019, relative to January 2004, for different depth slabs: 0 to 150 m, 150 to 300 m, 300 m to 700 m, 700 m to 1500 m (from light blue to dark blue). The gray shading marks the uncertainty around the upper 1500 m. The dashed black line illustrates the estimated increase in C_{ant} based on ΔC_{ant} from 1800 to 2007 scaled to the same period, using a scaling factor α of 0.29.

567 riverine carbon flux estimate of 0.8 ± 0.4 Pg C yr⁻¹ by Resplandy et al. (2018). An even
 568 better agreement would be achieved if the recently proposed temperature corrections were
 569 applied to the surface-based estimates (Dong et al., 2022; Watson et al., 2020). Specifically,
 570 Dong et al. (2022) suggested that the proper accounting of all temperature-related issues
 571 (e.g., skin correction) would increase the ocean uptake of the commonly used surface pCO₂
 572 based products by +35% (0.6 Pg C yr⁻¹) for the period 1982 to 2020.

573 Our estimate of the total increase in ocean sDIC of 51 ± 11 Pg C implies that from
 574 2004 through 2019, the ocean sink accounted for $\sim 31 \pm 7\%$ of the total anthropogenic CO₂
 575 emissions (here: from fossil fuel emissions and land-use change, Friedlingstein et al. (2022)).
 576 This uptake fraction is larger but within the uncertainties compared to the fraction reported
 577 by the Global Carbon Project based on ocean models and surface ocean pCO₂ products
 578 during the decade 2011 to 2020 ($26 \pm 4\%$, Friedlingstein et al. (2022)). As pointed out by
 579 Friedlingstein et al. (2022), within the Global Carbon Budget estimates, it is particularly
 580 the hindcast model-based estimates that indicate a smaller uptake. Similarly, Terhaar et
 581 al. (2022) used an emergent constraint approach to demonstrate that most CMIP models
 582 tend to take up too little CO₂ from the atmosphere. Although CMIP models differ from
 583 the hind-cast models used in the Global Carbon Budget, these findings further indicate
 584 that the models underestimate the oceanic carbon uptake, as also discussed by Hauck et al.
 585 (2020). Our interior ocean-based estimate thus supports the higher-end (mostly observation-

586 based) estimates of the ocean carbon sink in the Global Carbon Budget, albeit within large
 587 uncertainties.

588 Another reference point is the oceanic accumulation of C_{ant} between 2004 and 2020.
 589 Lacking an estimate of the C_{ant} accumulation over the same period, we scale the estimates
 590 of Sabine et al. (2004) and Gruber et al. (2019) to this period, assuming a transient steady-
 591 state (see Section 2.4). We obtain a global increase of 44 ± 6 Pg C (2.8 ± 0.4 Pg C yr⁻¹) in
 592 C_{ant} (1800-2007 scaled to 2004-2019 with a scaling factor of 0.29) and 49 ± 6 Pg C (3.1 ± 0.4
 593 Pg C yr⁻¹) in C_{ant} (1994-2007, scaled to 2004-2019 with a scaling factor of 1.45). The
 594 estimates are remarkably close to our estimate of the increase in total sDIC (51 ± 11 Pg
 595 C, i.e., 3.2 ± 0.7 Pg C yr⁻¹). This suggests that we can largely attribute the reconstructed
 596 increase in the sDIC to the uptake of anthropogenic CO₂ from the atmosphere. Similarly,
 597 when only considering the domain of MOBO-DIC and without upscaling, we also find that
 598 the trend in MOBO-DIC (40 ± 5 Pg C, i.e., 2.5 ± 0.3 Pg C yr⁻¹) is close to the increase in
 599 C_{ant} over the same period and domain (35 ± 4 Pg C, i.e., 2.2 ± 0.2 Pg C yr⁻¹, dashed line
 600 in Fig. 1), and also well within the uncertainties. Considering the proposed outgassing
 601 signal of C_{nat} , this would have been reflected in a weaker trend in total sDIC than in C_{ant} ;
 602 however, we do not observe this during our study period.

603 Superimposed onto this positive long-term trend of sDIC, the reconstructions reveal
 604 substantial interannual variations and a weakening of the trend after ~ 2012 , especially in the
 605 deeper waters. The following sections will further discuss these variations and the weakening
 606 trend. We also dive deeper into the differences between the anthropogenic component and
 607 the total sDIC in the water column, revealing changes in the natural DIC pool. Additionally,
 608 we find a strong seasonal signal, most pronounced near the surface. We do not discuss the
 609 seasonal variations near the surface, as the seasonal cycle of DIC was explored in Keppeler
 610 et al. (2020b).

611 5.2 Regional distribution of trends in sDIC

612 The rate of the depth-integrated accumulation of sDIC is regionally strongly
 613 structured (Fig. 2a), with the highest rates of accumulation found in the North Atlantic
 614 south of Iceland, i.e., the Subpolar Gyre. There, the linear trend exceeds 1.5 mol m⁻²
 615 yr⁻¹. An additional region with elevated rates of increase can be identified in the southern
 616 hemisphere between about 20°S and 45°S with typical accumulation rates of ~ 1 mol m⁻²
 617 yr⁻¹. The higher latitudes of the Southern Ocean, the tropical regions, the northern
 618 Indian, and particularly the North Pacific have considerably weaker depth-integrated
 619 changes in sDIC, typically 0.5 mol m⁻² yr⁻¹ or less. In some regions of the North Pacific,
 620 the depth-integrated sDIC even decreases over our study period. This vertical integral
 621 turns out to be a robust feature of our analyses as it is only weakly changing when
 622 removing trends within the water column that are not significant (compare Supporting
 623 Information Fig. S12 with Fig. 2a).

624 At each depth level, most of the trends in sDIC are statistically significant (95%
 625 confidence interval, see Supporting Information S7). In addition, the vertical integral does
 626 not change considerably when removing trends that are not significant (compare
 627 Supporting Information Fig. S12 with Fig. 2a). This is also the case for the negative
 628 trends in the North Pacific. Further support comes from the existence of a comparable
 629 negative trend in the surface DIC reconstructions of the OceanSODA-ETHZ product
 630 (Gregor & Gruber, 2021), as demonstrated in Section 4.2 and Supporting Information Fig.
 631 S11. Thus, this negative signal in the North Pacific appears robust within our period and
 632 is not an artifact of our method.

633 The similarity between the rate of depth-integrated accumulation of sDIC and C_{ant}
 634 becomes even more evident when they are put side by side, irrespective of how we estimated
 635 the expected change in C_{ant} from 2004 through 2019. The patterns and magnitude of the
 636 depth-integrated accumulation of sDIC (Fig. 2a) and the two different estimates of C_{ant}

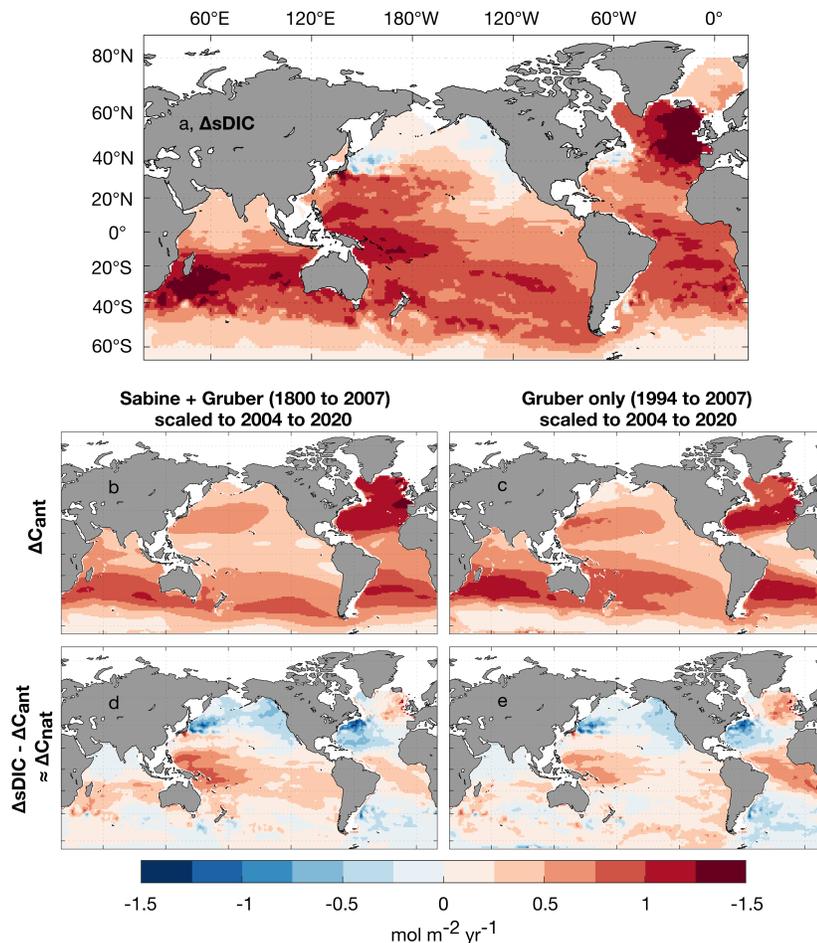


Figure 2. Maps of the column-integrated (upper 1500 m) (a) change in sDIC from 2004 through 2019 based on the linear trend, (b,c) change in C_{ant} scaled to the same period (2004 through 2019), and (d,e) the anomalous change (i.e., approximately the change in C_{nat}) from 2004 through 2019, estimated from the difference between the change in the MOBO-DIC inventory [illustrated in (a)], and the change in the C_{ant} inventory, scaled to the same period as (a) [illustrated in (b and c)]. For the combined estimate of C_{ant} by Sabine et al. (2004) (1800 to 1994) and Gruber et al. (2009) (1994 to 2007) (b,d) and for the estimate of C_{ant} by Gruber et al. (2009) (1994 to 2007; c,e). Scaling on the basis of the transient steady-state model ($anom = \Delta MOBO-DIC - \alpha \cdot \Delta C_{ant}$, $\alpha = 0.29$ for the period 1800-2007 and $\alpha = 1.45$ for the period 1994-2007). See Supporting Information Fig. S12 for the trends in MOBO-DIC on individual depth levels.

637 (Fig. 2b,c) are to the first order approximately the same, as also evidenced by their high
 638 pattern correlation coefficient $c = 0.56$ and 0.63 , between the trend in MOBO-DIC and the
 639 scaled ΔC_{ant} from the combined estimate by Sabine et al. (2004) and Gruber et al. (2019),
 640 and the estimate by Gruber et al. (2019), respectively. For example, we observe in all fields
 641 a large increase in the North Atlantic and a broad band of enhanced accumulation in the
 642 mid-latitudes of the Southern hemisphere. Also present in all fields is the weaker signal in
 643 the mid-latitude Southern Ocean. This further supports the conclusion that most of the
 644 column-integrated change in sDIC can be attributed to the increase in C_{ant} during this
 645 period.

646 However, there are also several notable differences, discernible when we subtract the
 647 estimated anthropogenic component (i.e., ΔC_{ant}) from the reconstructed change in sDIC
 648 (Fig. 2d,e). This difference can be interpreted as the change in the natural oceanic CO₂
 649 component of DIC, i.e., ΔC_{nat} , although given our steady-state assumption when estimating
 650 ΔC_{ant} , this difference can also contain an element of the non-steady-state, i.e., climate
 651 variability induced, component of ΔC_{ant} . The North Pacific stands out as the region with
 652 the biggest loss in C_{nat} . In addition, C_{nat} is lost in the upwelling region of the Atlantic
 653 sector of the Southern Ocean and the Subtropical Gyre of the North Atlantic. These losses
 654 of C_{nat} are counter-balanced by gains of C_{nat} in the tropics and the Indo-Pacific sector
 655 of the Southern Ocean. Furthermore, in the North Atlantic Subpolar Gyre, a region of
 656 strong uptake of C_{ant} , we also observe an increase in C_{nat} . Integrating ΔC_{nat} yields a total
 657 increase of 0.4 ± 0.8 Pg C yr⁻¹, and 0.1 ± 0.8 Pg C yr⁻¹, for C_{nat} based on the combined
 658 estimate (Sabine et al., 2004; Gruber et al., 2019), and the estimate by Gruber et al. (2019),
 659 respectively. Given the lack of statistical significance, we conclude that we cannot detect a
 660 global change in C_{nat} during our study period.

661 Nonetheless, the reduction of ΔC_{nat} in the North Pacific stands out. We link this
 662 change to the phasing of the Pacific Decadal Oscillation (PDO) as it shifted between 2004
 663 and 2019 from positive to negative. While negative PDO regimes are associated with a
 664 shallow thermocline in the Kuroshio Extension, which results in deep mixing and cooler
 665 SSTs in the North Pacific, positive PDO regimes are associated with warmer SSTs (Mantua
 666 & Hare, 2002). Thus, we speculate that during our analysis period, the shoaling of the
 667 thermocline in the North Pacific brought DIC stored at depth to the surface, allowing it to
 668 outgas, leading to an overall loss of DIC in this region. We expect that the opposite would
 669 occur during positive phases of the PDO, so that the net change over multiple decades
 670 would be close to zero, and thus not impact the long-term trend. To test this hypothesis,
 671 we plotted the trend in the surface sDIC from OceanSODA-ETHZ (Gregor & Gruber, 2021)
 672 over their entire study period (1985 through 2018). Over that extended period, we do not
 673 observe negative trends in surface sDIC in the North Pacific (not shown), indicating that
 674 the observed negative trend in the North Pacific sDIC and C_{nat} between 2004 and 2019 is
 675 not a long-term signal. The loss of C_{nat} in the North Pacific during our analysis period
 676 is partially balanced by a gain in C_{nat} in most parts of the Indo-Pacific, especially in the
 677 Western tropical Pacific. We find that this signal is associated with the phasing of the
 678 El Niño Southern Oscillation (ENSO), as further discussed in Supporting Information Fig.
 679 S13. We speculate that over a longer period than our 16 years, this signal of increased C_{nat}
 680 in the Western tropical Pacific would also be dampened.

681 Similarly, we link the changes in C_{nat} in the North Atlantic to the phasing of the
 682 Atlantic Multidecadal Oscillation (AMO; Kerr (2000) as between 2004 and 2019, the AMO
 683 index moved from positive to negative (see [https://climatedataguide.ucar.edu/
 684 climate-data/atlantic-multi-decadal-oscillation-amo](https://climatedataguide.ucar.edu/climate-data/atlantic-multi-decadal-oscillation-amo)). Negative AMO phases are
 685 associated with increased vertical mixing in the North Atlantic Subpolar Gyre, and thus,
 686 an increase of upper ocean DIC and C_{nat} in this region (Breedon & McKinley, 2016).
 687 Concurrently, in the North Atlantic Subtropical Gyre, negative AMO phases are
 688 associated with a decrease in DIC and C_{nat} in this region due to changes in the
 689 temperature affecting the solubility of CO₂. In the tropical Atlantic, the increase in C_{nat}
 690 during our study period might be associated with teleconnections from the AMO phasing.
 691 The loss of C_{nat} in the South Atlantic is in line with the findings by Keppler and
 692 Landschützer (2019) who reported a weakening of the Southern Ocean carbon sink in the
 693 Atlantic sector since ~2012. They linked this weakening to shifts in sea level pressure and
 694 associated changes in surface winds. We note that these links between changes in C_{nat} and
 695 the PDO and the AMO are speculative at this point, as the relatively short temporal
 696 extent of MOBO-DIC (16 years) prevents us from robustly concluding on the effect of
 697 multi-decadal modes of variability.

Fig. 3 reveals how the trend in sDIC varies with depth at the scale of entire ocean basins split into latitude bands (black line). Near the surface, sDIC is reconstructed to have increased, on average by about $0.6 \mu\text{mol kg}^{-1} \text{yr}^{-1}$, with some regions having a higher accumulation (e.g., $0.8 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ in the North Atlantic) and other regions less (e.g., $0.3 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ in the North Pacific). In all regions, the trend in sDIC increases between the mixed layer and the intermediate waters and then decreases with depth below that, reaching values of around $0.2 \mu\text{mol kg}^{-1} \text{yr}^{-1}$ at 1500 m. We observe the largest increase in sDIC in the Atlantic between ~ 200 m and 500 m ($\sim 0.9 \mu\text{mol kg}^{-1} \text{yr}^{-1}$).

Comparing the temporal trends in sDIC with the estimated changes in C_{ant} (blue and red lines in Fig. 3) highlights strong similarities but also distinct differences. Near the surface, sDIC increased less than C_{ant} during our study period. This difference is significant in all regions except for the Southern Ocean and North Atlantic. In the deeper ocean, the difference between the trend in sDIC and ΔC_{ant} is not significant in the Southern Ocean, North Pacific, tropical Indian Ocean, and the South Atlantic, while in the other regions, the trend in sDIC tends to be larger than the two estimates of ΔC_{ant} . The differences between the trends in sDIC compared to those in C_{ant} imply a loss of C_{nat} in the upper ocean, and a gain of C_{nat} in the ocean's interior below a few hundred meters depth. Combined with the lack of an overall change in C_{ant} , this suggests a strong internal redistribution of oceanic C_{nat} over our analysis period.

The similarities and differences in the vertical distribution of the trends in sDIC, C_{ant} , and C_{nat} become even more evident when analyzing zonal mean sections of these components (Fig. 4). Due to methodological constraints, there are some discontinuities at 500 m in the MOBO-DIC derived sDIC (Fig. 4a-c), which are associated with boundaries generated by the depth slabs. Aside from that, the trend in sDIC and ΔC_{ant} (Fig. 4d-f) are very similar, as noted above for the mean profiles. This figure again highlights the loss of C_{nat} (Fig. 4g-i) at the surface, except in the North Atlantic. We also observe a loss of C_{nat} in the North Pacific, extending down to 1500 m but most pronounced in the upper ~ 800 m. The northern high latitudes tend to lose C_{nat} at depth, while the low latitudes tend to gain C_{nat} at depth. Overall, the redistribution of C_{nat} occurs both horizontally, as demonstrated in Fig. 2, and vertically (Fig. 3 and 4), but as pointed out above, the signal in C_{nat} is within the uncertainty bounds.

We cannot identify the potential reasons for this redistribution, but the upper ocean loss of C_{nat} may be at least partially driven by the warming of the ocean, which is strongest in the upper ocean (IPCC, 2021). In addition, such a redistribution pattern is reminiscent of the impact of the ocean's biological pump, where an increased efficiency of this pump would lead to a depletion of C_{nat} in the upper ocean and an accumulation at depth. As we observe this pattern most prominently in the tropics, we speculate that biology may be driving the change in sDIC here. Conversely, as we already hypothesized above, the other regions, including the North Pacific, North Atlantic, and Southern Ocean are likely driven by physical changes.

It should also be noted that the vertical profile in the trend is strongly influenced by interannual variations, such as variations in the thermocline and surface outgassing. Thus, the signal in the mixed layer is prone to large interannual to decadal variations, which are especially dominant in the Southern Ocean (Le Quéré et al., 2007; Landschützer et al., 2015; Keppler & Landschützer, 2019). Therefore, the observed trends in the mixed layer depend greatly on the start and end year and should be interpreted with care. A longer time series would yield a more robust result.

5.3 Interannual variability at global and basin-scale

The interannual variability of sDIC, defined here as the standard deviation in time (seasonal cycle and trend removed), is in our product rather small, especially when compared to the magnitude of the trend (previous section) and the amplitude of the seasonal cycle

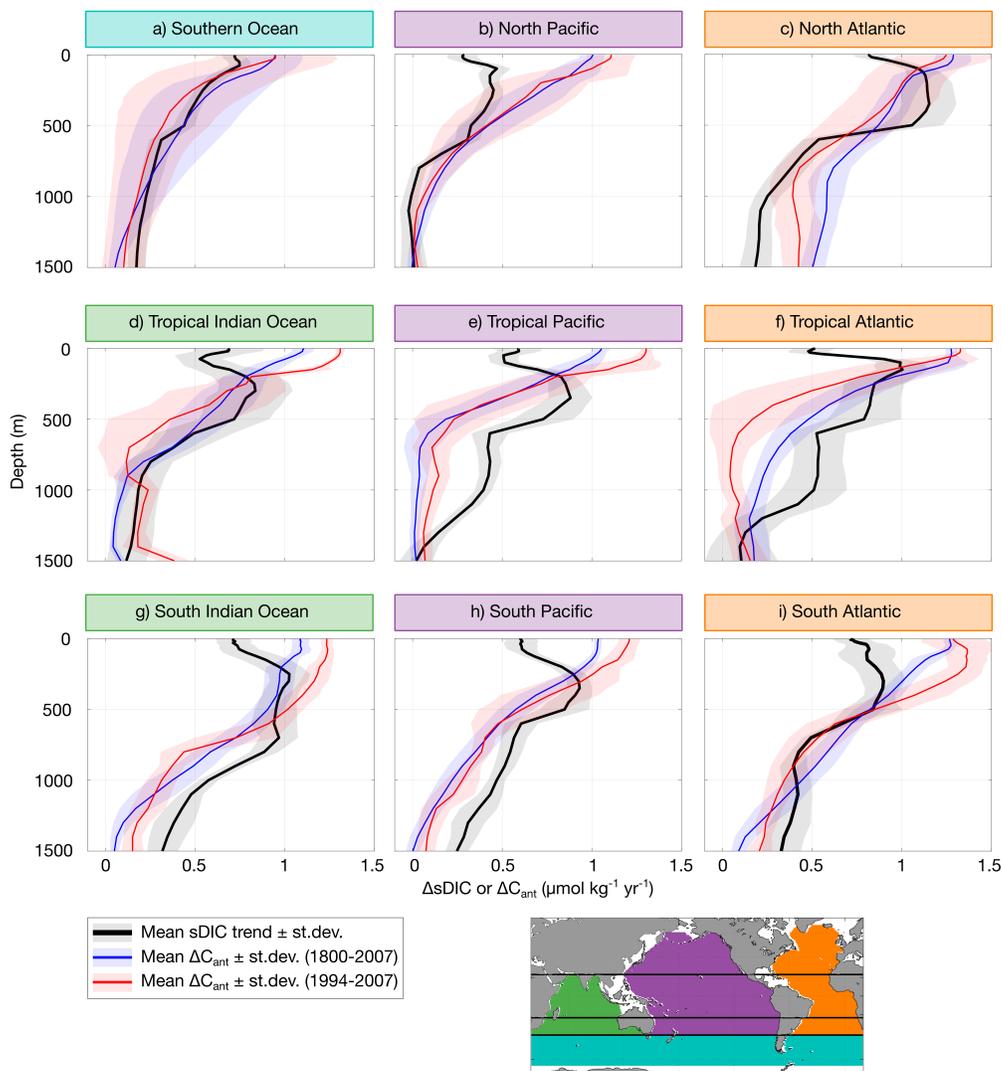


Figure 3. Vertical profiles of the mean trend in subregions for sDIC estimated by MOBO-DIC (black) and ΔC_{ant} from 1800 to 2007 scaled to the period 2004 through 2019 (blue), and from 1994 to 2007, scaled to 2004 through 2019 (red). The uncertainty of the trend in sDIC, based on the standard deviation across the trend in the 15 ensemble members, is illustrated in shading. The uncertainty of the trend in ΔC_{ant} , based on the standard deviation in the latitude-longitude space, is illustrated in shading. Separately for the Southern Ocean (a), Indian Ocean (d,g), Pacific (b,e,h), Atlantic (c,f,i), in the northern temperate regions (until 23°N , b,c), the tropics (23°N to 23°S , d-f), and in the southern temperate regions (from 23°S to 40°S , g-i). The map at the bottom indicates the limits of the ocean basins in color, and the climatic regions are delimited by black lines.

749 (Keppler et al., 2020b). With a global mean temporal standard deviation of $2 \mu\text{mol kg}^{-1}$
 750 at 150 m (the depth level with the largest mean standard deviation), compared to a global
 751 mean uncertainty of $18 \mu\text{mol kg}^{-1}$ at 150 m, the interannual variability is well within the
 752 product uncertainty of MOBO-DIC in most parts of the ocean. However, as highlighted in
 753 Section 4.3, MOBO-DIC likely underestimates the interannual variability.

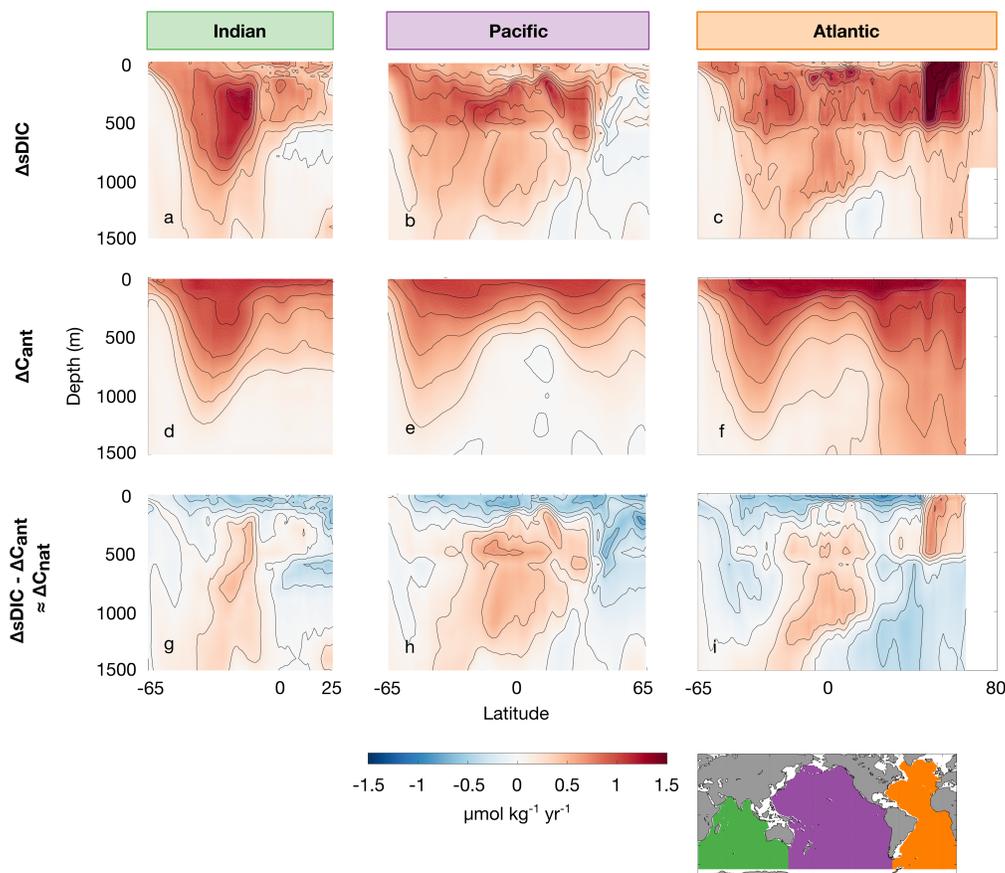


Figure 4. Zonal mean sections of the trend in sDIC from 2004 through 2019 (a-c), of ΔC_{ant} from 1800 to 2007, scaled to our period (d-f), and the difference between the two, i.e., ΔC_{nat} (g-i), for the Indian Ocean (a,d,g), Pacific (b,e,h), and the Atlantic (c,f,i). The map at the bottom right illustrates the boundaries of the basins.

754 Nevertheless, some clear regional signals of the interannual variability emerge (Fig. 5):
 755 the largest interannual signal is generally in the thermocline region (~ 150 m), while in the
 756 mixed layer and below 700 m, the interannual variability is minimal. The equatorial Pacific
 757 stands out as a region with the largest variance, while we observe very little interannual
 758 variability in the Southern Ocean, a region with large decadal variability in the air-sea CO_2
 759 flux estimates (Le Quéré et al., 2007; Landschützer et al., 2015; Keppler & Landschützer,
 760 2019). A recent study has suggested that the decadal variations of the air-sea CO_2 fluxes in
 761 the Southern Ocean may be overestimated in the mapped surface estimates (Gloege et al.,
 762 2021); however, the strongest variations occur around the year 2000 (see, e.g., Friedlingstein
 763 et al. (2022)), i.e., before the start of our time-series here.

764 We further illustrate the nature of the mean vacillations of the vertically integrated sDIC
 765 (upper 1500 m) for large subregions in Fig. 6. The most dominant interannual variations
 766 are found in the Pacific, where we see a steep increase in sDIC between 2010 and 2014 in the
 767 tropics. The northern temperate Pacific also stands out: Here, the trend in sDIC is initially
 768 weak until 2010, increases until 2014, and then we observe a negative trend until the end of
 769 the time series in December 2019. Both in the northern and southern temperate regions of
 770 the Atlantic, the sDIC trend has weakened since around 2012. In contrast, averaged over the

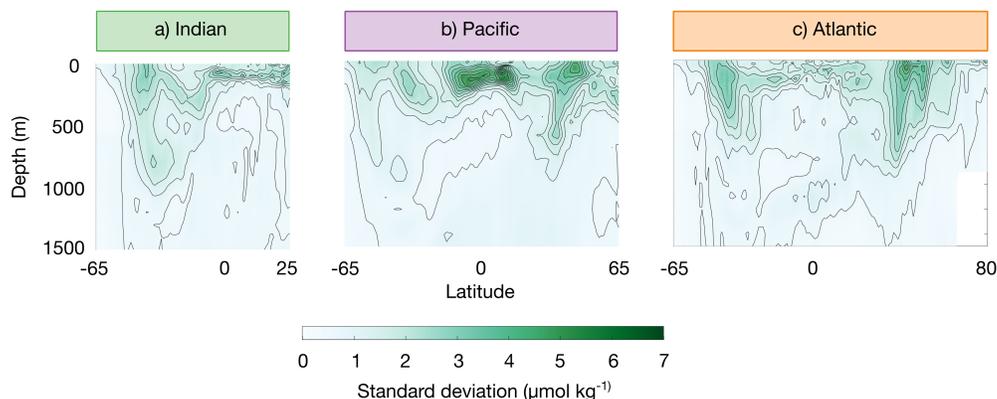


Figure 5. Zonal mean sections of the interannual variability of sDIC, defined as the standard deviation of the time-dimension of sDIC (detrended, seasonal cycle smoothed with a 12-month running mean) for the Indian (a), Pacific (b), and Atlantic Oceans (c). See the map at the bottom right of Fig. 4 for the boundaries of the basins.

771 whole Southern Ocean, we find very few interannual variations here. Similarly, the Indian
 772 Ocean, the South Pacific, and the tropical Atlantic all vary little interannually in the upper
 773 1500 m when averaging over these areas.

774 We link the sDIC increase in the tropical Pacific at least partially to a shift from La
 775 Niñas (especially in 2008 and 2011) to El Niños (especially in 2015 and 2019, see
 776 Supporting Information Fig. S13). The other ENSO-related signals during our study
 777 period are considerably weaker and seem mostly dampened when considering the whole
 778 tropical Pacific. The large variation in the North Pacific is likely to be influenced by the
 779 phasing of the PDO, but may also have an ENSO-related teleconnection. We find that the
 780 weakening of the vertically integrated sDIC around 2012, illustrated in Fig. 1, stems
 781 largely from the high latitude South Atlantic and the tropical Pacific. The weakening of
 782 the sink in the high latitude South Atlantic is in line with the findings by Keppler and
 783 Landschützer (2019), who report a weakening of the CO₂ uptake in the Atlantic sector of
 784 the Southern Ocean around 2012. While this signal is not dominant when averaging over
 785 the whole Southern Ocean, this weakening sink around 2012 is also visible in the global
 786 changes in sDIC (Fig. 1), highlighting the important role of the Southern Ocean carbon
 787 uptake (here: specifically its Atlantic sector) globally. A longer time series is needed to
 788 investigate if this is a long-term decline or part of multi-decadal oscillations, such as the
 789 AMO. We know from previous studies that this weakening may be due to changes in the
 790 circulation, as suggested by DeVries et al. (2017) or linked to atmospheric circulation, as
 791 proposed by Keppler and Landschützer (2019). An alternative hypothesis for these
 792 changes is that volcanoes are the driving force for such sudden changes (McKinley et al.,
 793 2020). However, during our study period, no large volcanic eruptions occurred that may
 794 explain the observed signals.

795 6 Caveats and Uncertainties

796 Given the sparsity of DIC observations, our product fills substantial observational gaps
 797 in time and space. However, our evaluation with independent data provides confidence
 798 in the robustness of the presented numbers within the uncertainty limits. Nonetheless,
 799 there are good reasons to conclude that MOBO-DIC tends to underestimate the trend and
 800 interannual variability. Although this underestimation is within the uncertainty limits, it

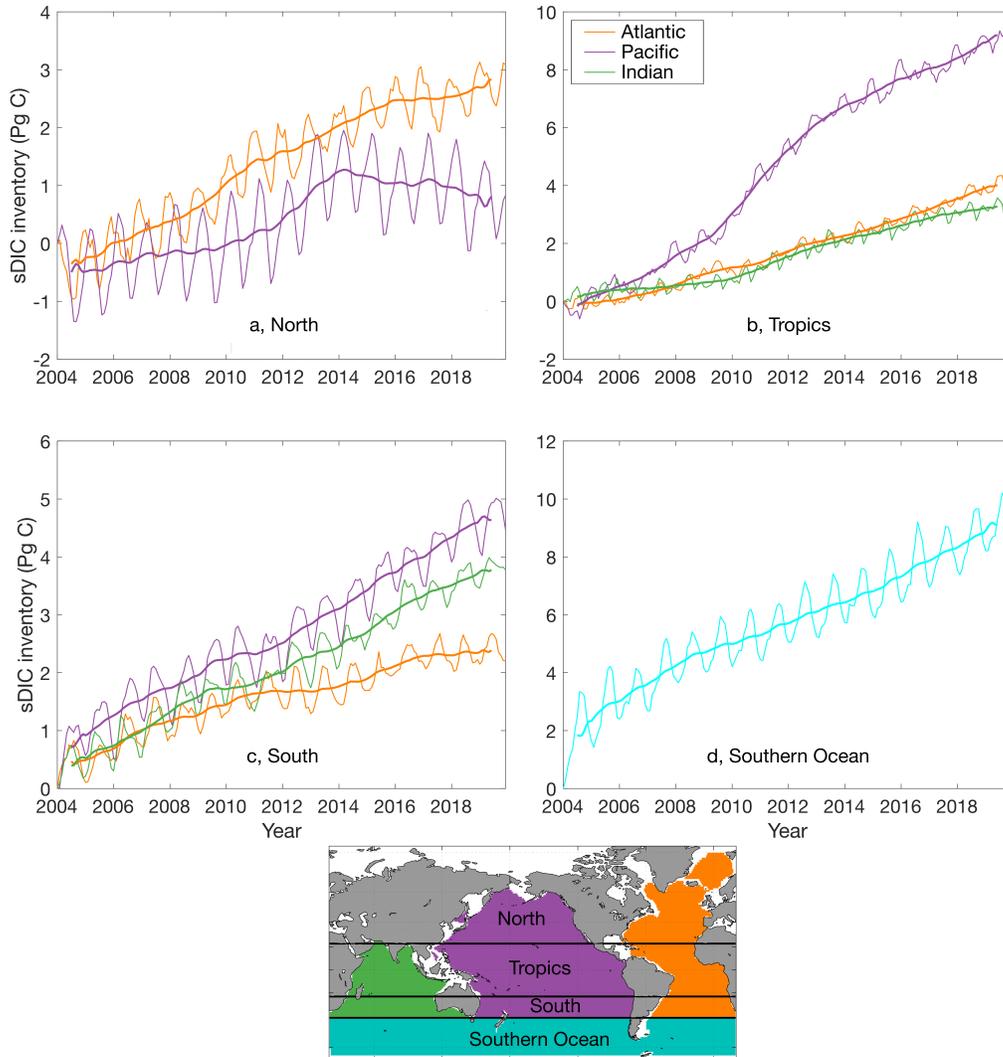


Figure 6. Timeline of the vertical integral (upper 1500 m) of sDIC relative to January 2004 (think solid lines) in the northern temperate regions (a), tropics (b), southern temperate regions (c), and the Southern Ocean. In a-c, separately for the Atlantic (orange), Pacific (purple), and Indian Ocean (green). Note that the y-axes differ in each subplot. Thick solid lines have the seasonal cycle smoothed with a 12-month moving average (first and last six months removed). The inserted map illustrates the boundaries of the subregions.

801 could be significant when integrating in the water column (see Section 4 and Supporting
 802 Information S5 and S6). Further research should be conducted on this, especially as more
 803 data becomes available with new GLODAP releases and BGC Argo data.

804 The uncertainties of the MOBO-DIC estimated sDIC at the level of a single grid cell
 805 are relatively large (global mean of $18 \mu\text{mol kg}^{-1}$) and are often larger than the signal
 806 in the trend or variability. As our mapping method minimizes the overall bias between
 807 the target data (i.e., the GLODAP DIC measurements) and the mapped estimate, we can
 808 assume that any local imprecisions average out when integrating or averaging over large
 809 areas. This means that MOBO-DIC is most robust when considering large areas, while

810 analyses at single grid cells should be interpreted carefully. In this study, we present such
811 integrals and averages over whole ocean basins.

812 We want to note that the linear trend analysis used to quantify and assess the long-
813 term changes in DIC has its limitations, too. First, it is not a given that the increase in
814 oceanic carbon is, in fact, linear. Second, the linear trends are based on a relatively short
815 period of 16 years, and thus, the conditions of the start and end years tend to considerably
816 affect the trend over such a short period (Fay & McKinley, 2013). Furthermore, decadal
817 variations might also affect the linear trends. We found that, locally, some trends are not
818 robust (see Supporting Information Fig. S12) but anticipate that our global trend estimate
819 is robust within the uncertainty, as overestimates of the trend in some regions are likely to
820 be balanced by underestimates elsewhere.

821 Our comparison with C_{ant} also relies on many assumptions. First, as there is not yet
822 a published estimate of C_{ant} for the current period, we scaled previous estimates to our
823 period, assuming a steady state (see Section 2.4). Further, our estimate of C_{nat} is based
824 on the difference in the change in total sDIC and ΔC_{ant} . However, due to the steady-state
825 assumption when estimating ΔC_{ant} , the difference may also contain an element of the non-
826 steady-state component of anthropogenic ΔC_{ant} . Thus, the analyses with C_{ant} and C_{nat}
827 could be improved in the future by using a C_{ant} estimate of the same period.

828 7 Summary and Conclusions

829 This release of the Mapped Observation-Based Oceanic Dissolved Inorganic Carbon
830 (MOBO-DIC) extends the climatological estimate by Keppler et al. (2020b) in time, thus
831 giving insights into the spatiotemporal evolution of the ocean DIC stock at a monthly
832 resolution from January 2004 through December 2019. With a spatial resolution of 1° ,
833 extending from 65°N to 65°S , and until 80°N in the Atlantic, and covering the entire upper
834 and middle ocean (depths from 2.5 m to 1500 m on 28 uneven depth levels) this dataset
835 provides a near-global view. We conducted an in-depth validation of our new data product,
836 which considers sources of uncertainties from the measurements, representation errors, and
837 uncertainties stemming from our mapping method. We trust that our estimate of DIC is
838 robust within the uncertainty ranges provided (global mean uncertainty of $18 \mu\text{mol kg}^{-1}$).

839 Our analysis of the trend in sDIC provides the first direct assessment of the changes in
840 the total sDIC stock (natural + anthropogenic) based on observations. It should be noted
841 that at large scales, the changes in sDIC and DIC are numerically equal because the trend
842 in salinity is negligible once integrated vertically and over large regions (Cheng et al., 2020).
843 Our estimate of the global increase of sDIC during our study period ($3.2 \pm 0.7 \text{ Pg C yr}^{-1}$)
844 is approximately $31 \pm 7\%$ of the anthropogenic CO_2 emissions from fossil fuels and land use
845 change during our study period (Friedlingstein et al., 2022). We find that this increase
846 in sDIC is largely associated with the increase in anthropogenic carbon (C_{ant}) during this
847 period ($2.8 \pm 0.4 \text{ Pg C yr}^{-1}$ or $3.1 \pm 0.4 \text{ Pg C yr}^{-1}$, depending on the method).

848 MOBO-DIC also allows for the first assessment of changes in natural oceanic carbon
849 (ΔC_{nat}) by subtracting ΔC_{ant} from the changes in the total sDIC, yielding a statistically
850 insignificant global mean ΔC_{nat} of $0.4 \pm 0.8 \text{ Pg C yr}^{-1}$ or $0.1 \pm 0.8 \text{ Pg C yr}^{-1}$, depending on
851 the method used to estimate C_{ant} . Previous studies had suggested a potential outgassing
852 of C_{nat} due to elevated sea surface temperatures (McNeil & Matear, 2013), which would
853 affect the global climate. While the large uncertainties in MOBO-DIC and C_{ant} do not
854 rule out such a net outgassing signal of C_{nat} , we observe no statistically detectable change
855 in C_{nat} between 2004 and 2020. Instead, our analysis reveals a redistribution of C_{nat} -
856 a phenomenon that had not been previously investigated at a global scale. During our
857 study period, the upper ocean appears to have mostly lost C_{nat} , while below that, large
858 parts of the ocean increased in C_{nat} . The loss of C_{nat} near the surface could be driven
859 by increased ocean temperatures, as proposed by IPCC (2021). The redistributions in the

860 Pacific correspond to the phasing of the El Niño Southern Oscillation (ENSO) and the Pacific
861 Decadal Oscillation (PDO), while the redistributions in the North Atlantic correspond to
862 the phasing of the Multidecadal Atlantic Oscillation (AMO). However, at this stage, our
863 study period from 2004 through 2019 is too short to robustly conclude on (multi-) decadal
864 signals.

865 The interannual variability in sDIC is substantially weaker than the seasonal cycle and
866 temporal trend in most parts of the ocean. However, it should be noted that MOBO-DIC
867 likely underestimates the interannual variability. We find a mean standard deviation in time
868 of detrended, deseasonalized sDIC at the depth level with the largest variability (150 m)
869 is $2 \mu\text{mol kg}^{-1}$. We find that most of the global-scale variations in sDIC stem from the
870 North and tropical Pacific, in correspondence with ENSO and the PDO, and to a lesser
871 extent from the high latitude South Atlantic. We find a weakening of the positive trend
872 in the high-latitude South Atlantic around the year 2012. This signal is also visible in the
873 global integral of the sDIC, and future studies should examine the continued evolution of
874 this signal as well as its drivers. The interannual variations are comparably weak in the
875 other sectors of the Southern Ocean and the Indian Ocean.

876 We can now constrain the ocean carbon sink from surface measurements in combination
877 with riverine flux estimates (previous studies) and based on the direct DIC measurements
878 that reflect the changes in the DIC pool (this study). While the surface-based estimates
879 benefit from more observations, large uncertainties are associated with such an indirect
880 approach. The interior perspective suffers from considerably less data but, due to the direct
881 approach, does not need a riverine flux adjustment or gas transfer parametrization. The
882 two perspectives each have their strengths and weaknesses, so having both perspectives
883 substantially improves our understanding and the quantification of the global ocean carbon
884 sink. The two estimates are in good agreement ($3.2 \pm 0.7 \text{ Pg C yr}^{-1}$ and $2.7 \pm 0.6 \text{ Pg C yr}^{-1}$
885 for the interior and surface perspective, respectively), despite being based on independent
886 data (SOCAT vs. GLODAP). However, the surface-based estimates would be larger (i.e.,
887 closer to our estimate) when considering a higher-end riverine flux estimate (e.g., 0.8 ± 0.4
888 Pg C yr^{-1} by Resplandy et al. (2018), compared to $0.6 \pm 0.4 \text{ Pg C yr}^{-1}$ by Friedlingstein
889 et al. (2022) used in this study). In addition, the agreement between the surface-based
890 estimates and our interior ocean estimate would be even higher if the proposed temperature
891 corrections were applied to the surface estimates. Specifically, Dong et al. (2022) estimated
892 that accounting for these corrections would increase the ocean uptake of the surface pCO_2
893 based products by 0.6 Pg C yr^{-1} from 1982 through 2020.

894 Further, within the Global Carbon Budget (Friedlingstein et al., 2022), the observation-
895 based methods that estimate the carbon fluxes based on surface measurements are higher
896 than the model-based estimates. Our analysis from the interior ocean perspective suggests
897 that the true value likely lies closer to the observation-based surface estimates in the Global
898 Carbon Budget than to the model-based estimates, as also suggested by Terhaar et al.
899 (2022). Thus, the current approach of averaging all ocean carbon sink estimates from
900 observations and models in the Global Carbon Budget could be revisited and improved to
901 obtain the best estimate, e.g., by weighting the observation-based estimates stronger than
902 the models.

903 Our new data product is available for the scientific community and can be used to
904 further investigate the temporal changes in DIC and its effect on marine organisms.
905 Potential further insights into the processes and drivers could be gained by prolonging the
906 timespan and investigating the multi-decadal variations. Additionally, our product
907 provides the basis to compare the decadal variations of observation-based DIC to the
908 changes in the upper Meridional Overturning Circulation, which weakened in the 1980s,
909 strengthened in the 1990s, and weakened again in the 2000s (DeVries et al., 2017).
910 Similarly, further comparing the decadal variations of the Southern Ocean carbon sink
911 (Le Quéré et al., 2007; Landschützer et al., 2015; Keppler & Landschützer, 2019) to the

912 variations in the DIC pool in this region could lead to important new insights on the
913 global carbon cycle.

914 **Acknowledgments**

915 The research leading to these results has received funding from the European
916 Community's Horizon 2020 Project under grant agreement no. 821003 (4C). This work
917 was also sponsored by NSF's Southern Ocean Carbon and Climate Observations and
918 Modeling (SOCCOM) Project under the NSF Awards PLR-1425989 and OPP-1936222,
919 with additional support from NOAA and NASA. The authors are grateful for the Argo
920 data collected and made freely available by the International Argo Program and the
921 national programs that contribute to it (<https://www.argo.ucsd.edu> and
922 <https://argo.jcommops.org>). The Argo Program is part of the Global Ocean Observing
923 System. This publication uses Hawaii Ocean Time-series observations supported by the
924 U.S. National Science Foundation under Grant OCE-1260164. It also used data from the
925 Southern Ocean Carbon and Climate Observations and Modeling project (SOCCOM).
926 Data were collected and made freely available by SOCCOM, funded by the National
927 Science Foundation, Division of Polar Programs (NSF PLR-1425989), supplemented by
928 NASA and by the International Argo Program and the NOAA programs that contribute
929 to it. This work benefited from discussions with Birgit Klein (BSH), Jochem Marotzke
930 (MPI-M), Johanna Baehr (UHH), and Jens D. Müller (ETHZ).
931

932 **Data availability**

933 The data product created during this study are freely available from NCEI/OCADS: **XXX**
934 and should be cited as **XXX TBC**.

935 The GLODAP DIC ship measurements are available at
936 <https://www.ncei.noaa.gov/data/oceans/ncei/ocads/data/0237935/>. The mapped
937 Argo-based fields of temperature and salinity are available at
938 <http://sio-argo.ucsd.edu/RG.Climatology.html>. The WOA-mapped climatologies of
939 silicic acid, nitrate, and dissolved oxygen are available at
940 <https://www.nodc.noaa.gov/OC5/woa18/woa18data.html>. The atmospheric pCO₂
941 based on the GlobalView xCO₂ is available at
942 https://www.nodc.noaa.gov/ocads/oceans/SPC02_1982_present_ETH_SOM_FFN.html.
943 The mapped annual climatology of DIC is available at
944 <https://www.ncei.noaa.gov/data/oceans/ncei/ocads/data/0162565/mapped/>. The
945 time-series data from HOT, BATS, and Drake Passage are available at
946 <http://hahana.soest.hawaii.edu/hot/hot-dogs/bextraction.html>,
947 <http://bats.bios.edu/bats-data/>, and
948 <https://www.nodc.noaa.gov/archive/arc0118/0171470/2.2/data/0-data/>,
949 respectively. The DIC estimated based on BGC-Argo floats in the Southern Ocean
950 (SOCCOM floats) is available at <http://socompu.princeton.edu/www/index.html>.
951 The OceanSODA surface DIC fields are available at [https://www.ncei.noaa.gov/](https://www.ncei.noaa.gov/access/metadata/landing-page/bin/iso?id=gov.noaa.nodc:0220059)
952 [access/metadata/landing-page/bin/iso?id=gov.noaa.nodc:0220059](https://www.ncei.noaa.gov/access/metadata/landing-page/bin/iso?id=gov.noaa.nodc:0220059). The
953 MOBO-DIC monthly climatology is available at [https://www.ncei.noaa.gov/access/](https://www.ncei.noaa.gov/access/metadata/landing-page/bin/iso?id=gov.noaa.nodc:%3A0221526)
954 [metadata/landing-page/bin/iso?id=gov.noaa.nodc:%3A0221526](https://www.ncei.noaa.gov/access/metadata/landing-page/bin/iso?id=gov.noaa.nodc:%3A0221526). The monthly
955 climatology of DIC by Broullón et al. (2020) is available at
956 <https://doi.org/10.20350/digitalCSIC/10551>. The data for C_{ant} are available at
957 <https://www.ncei.noaa.gov/access/ocean-carbon-data-system/oceans/ndp-100/>
958 [ndp100.html](https://www.ncei.noaa.gov/access/ocean-carbon-data-system/oceans/ndp-100/) and [https://www.ncei.noaa.gov/access/metadata/landing-page/bin/](https://www.ncei.noaa.gov/access/metadata/landing-page/bin/iso?id=gov.noaa.nodc:0001644)
959 [iso?id=gov.noaa.nodc:0001644](https://www.ncei.noaa.gov/access/metadata/landing-page/bin/iso?id=gov.noaa.nodc:0001644) for the periods 1800 to 1994 and 1994 to 2007,
960 respectively. We use the bathymetry from Etopo2 (2001), and the Multivariate El Niño
961 Index (MEI; Wolter and Timlin (2011); <https://psl.noaa.gov/enso/mei/>).

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Figure 1.

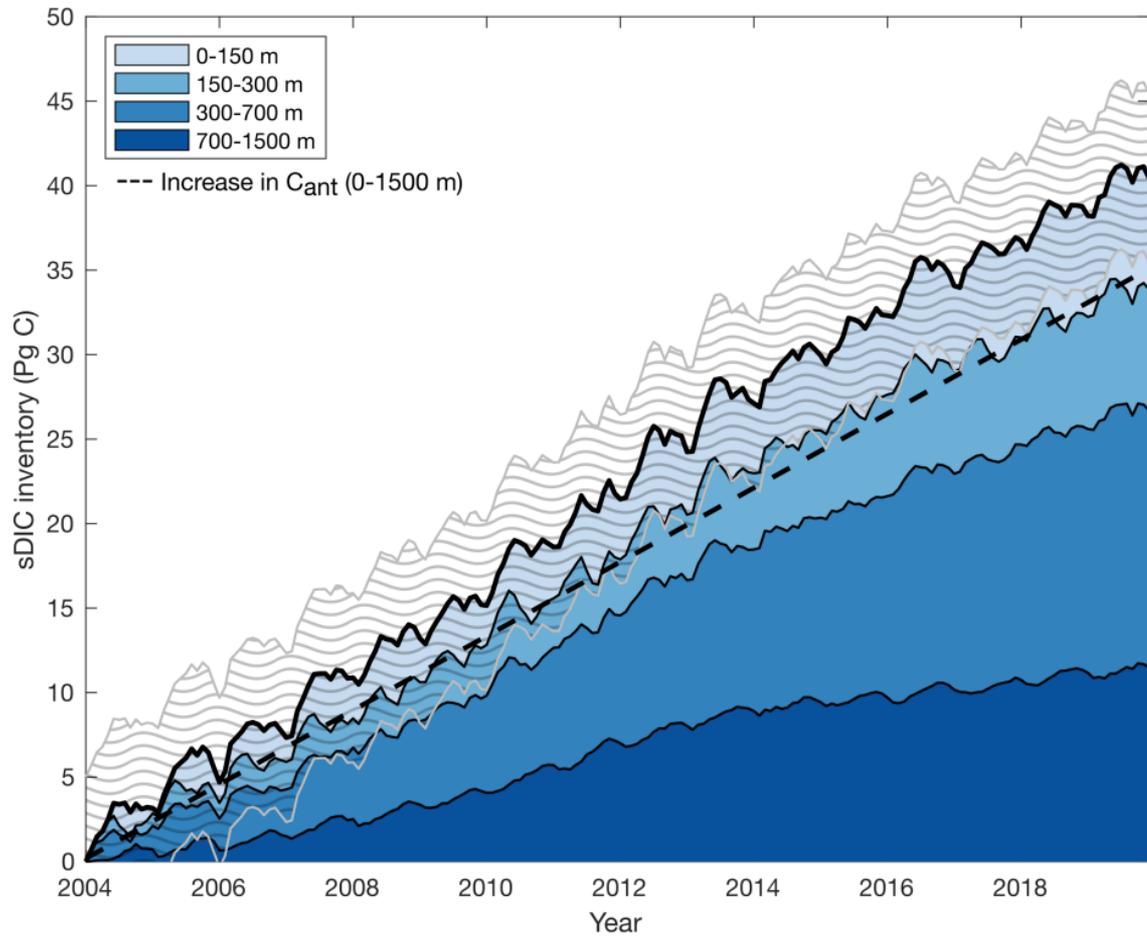
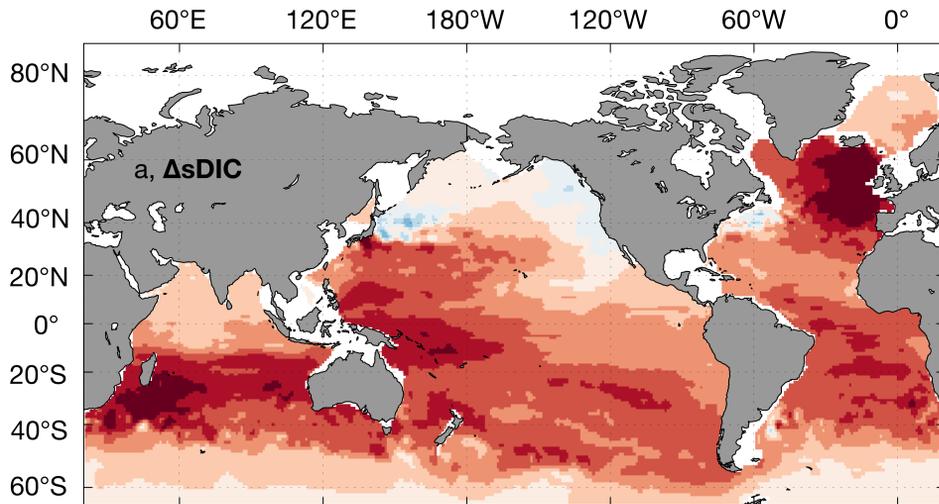


Figure 2.



Sabine + Gruber (1800 to 2007)
scaled to 2004 to 2020

Gruber only (1994 to 2007)
scaled to 2004 to 2020

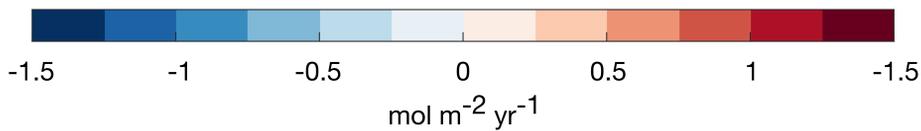
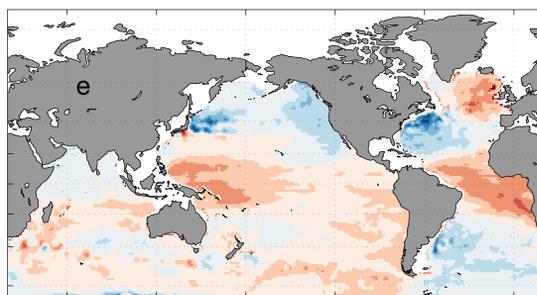
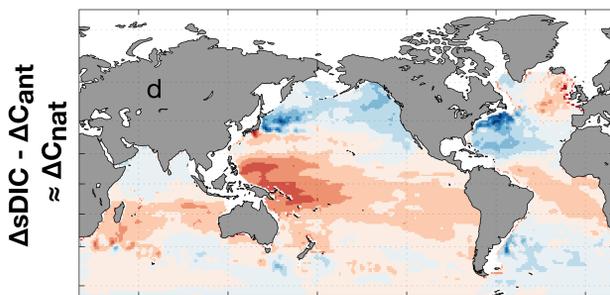
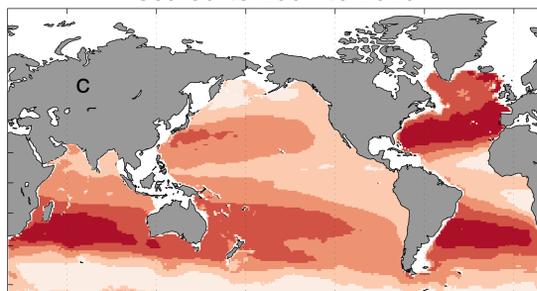
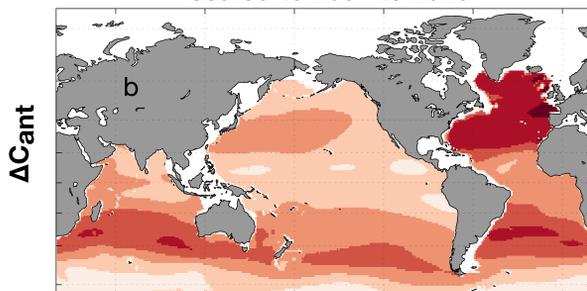
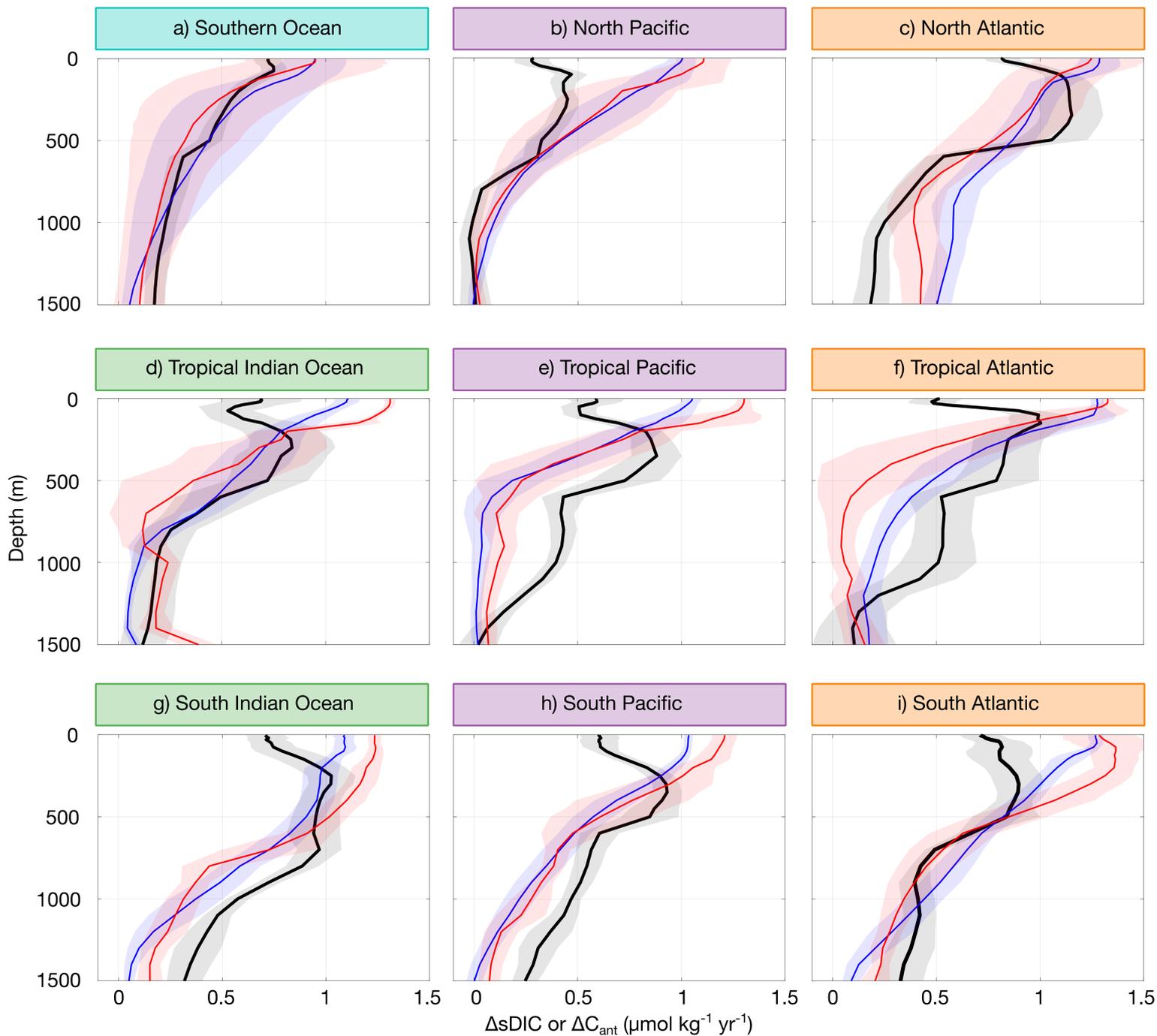


Figure 3.



— Mean $sDIC$ trend \pm st.dev.
 — Mean ΔC_{ant} \pm st.dev. (1800-2007)
 — Mean ΔC_{ant} \pm st.dev. (1994-2007)

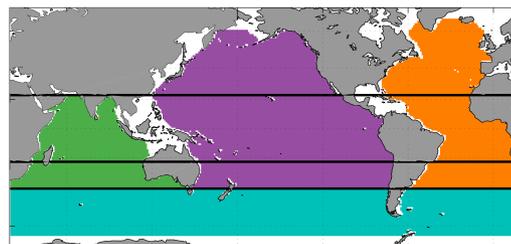


Figure 4.

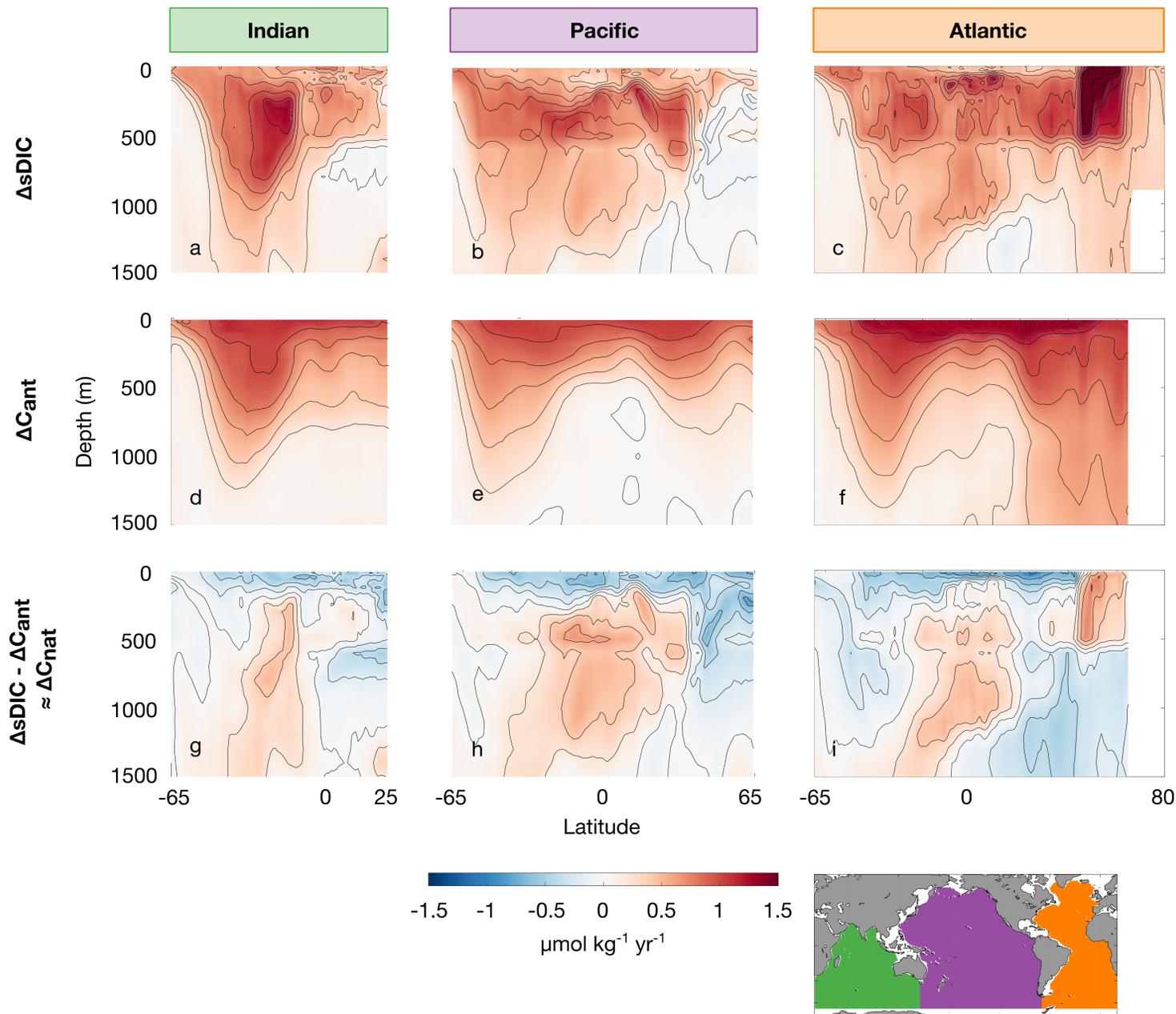


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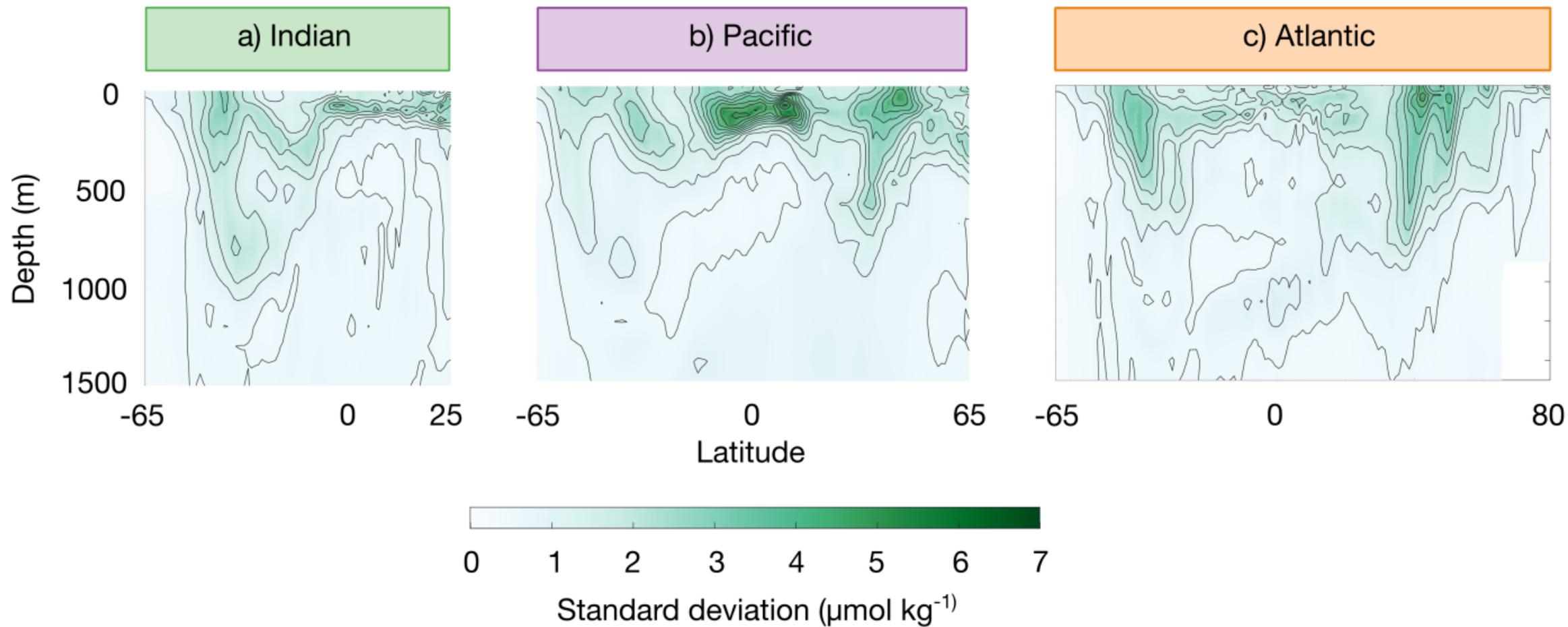


Figure 6.

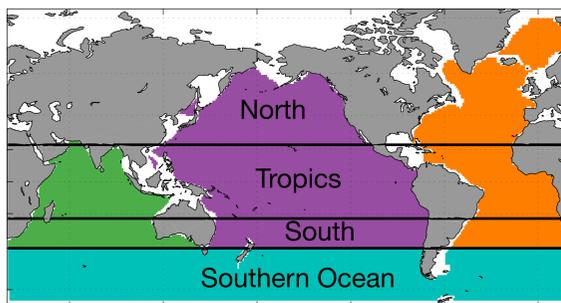
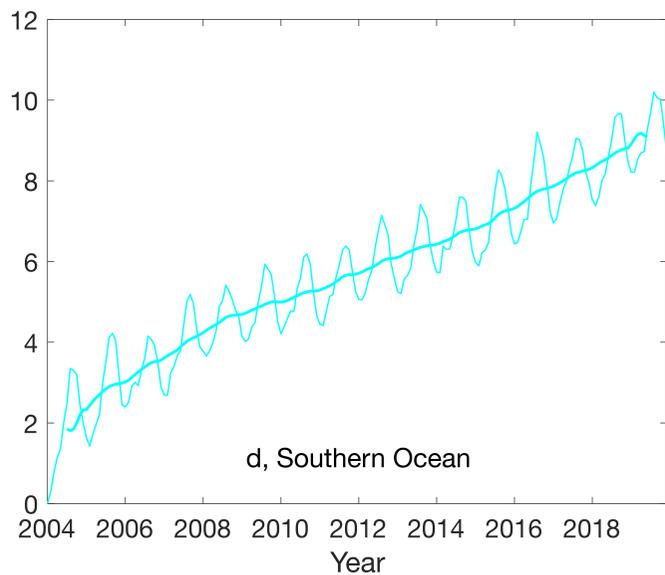
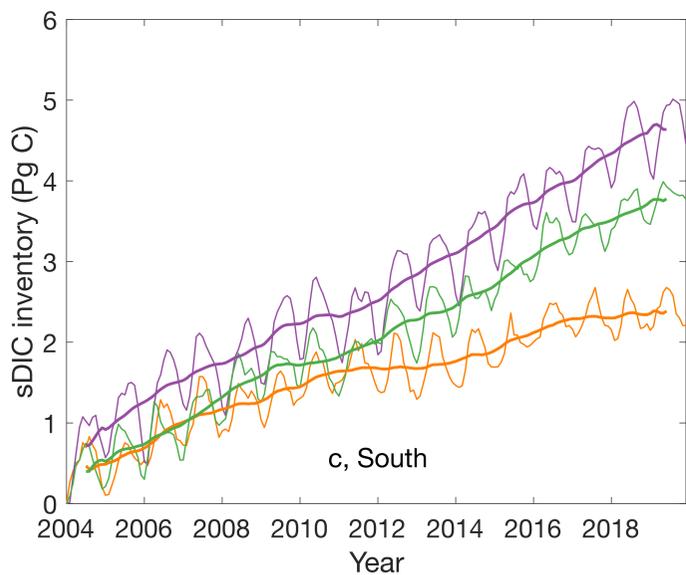
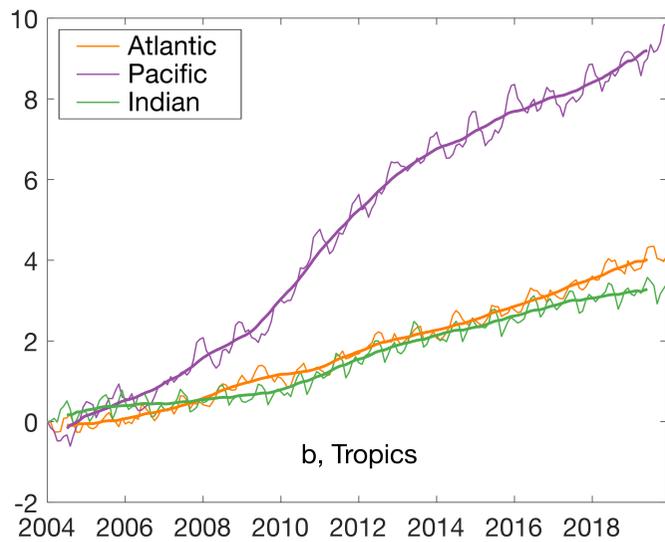
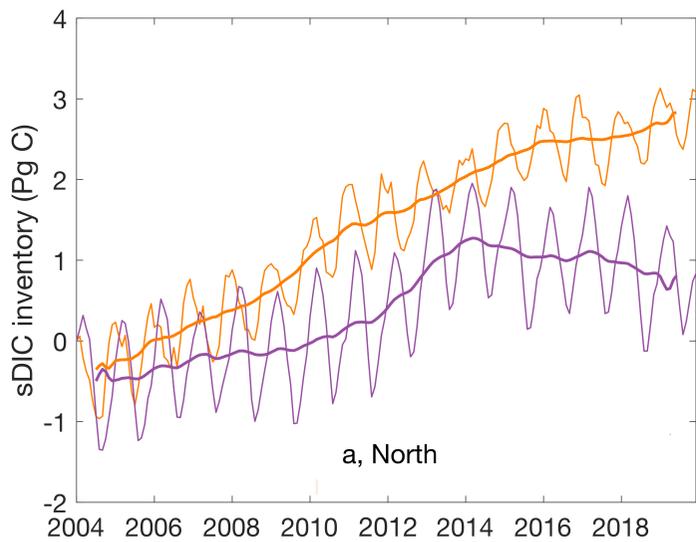


Table 1.

Compared data set	Type of data	Bias ($\mu\text{mol kg}^{-1}$)	RMSD ($\mu\text{mol kg}^{-1}$)	MOBO-DIC uncertainty ($\mu\text{mol kg}^{-1}$)	Comparison uncertainty ($\mu\text{mol kg}^{-1}$)
GLODAPv2.2021	Ship data, without interpolation or mapping (used to train the network)	0	16	18	2
Lauvset climatology	Global climatology (optimal interpolation)	7	17	18	7
Broullón climatology	Global monthly climatology (single-step neural network)	10	17	18	N/A
MOBO-DIC _{clim}	Global monthly climatology (cluster-regression)	11	20	18	9
HAMOCC	Synthetic data	-1	12	18	N/A
BATS	Time-series station	12	17	17	2
HOT	Time-series station	-4	15	17	2
Drake Passage	Time-series station (surface)	16	42	18	1
SOCOM floats	Calculated DIC from BGC floats (pH) with LIAR algorithm	-5	14	17	6
OceanSODA-ETHZ	Global surface estimate (cluster-regression)	4	15	18	21

Table 2.

Trend

Compared data set → Depth ↓	BATS	MOBO-DIC at BATS	HOT	MOBO-DIC at HOT	Drake Passage (surface)	MOBO-DIC at Drake Passage	SOCCOM floats	MOBO-DIC at SOCCOM floats
20 – 40 m	1	7	5	2	8	1	-20	-9
100 – 150 m	3	8	13	6	N/A	N/A	3	1
600 -800 m	16	5	4	5	N/A	N/A	19	26

IAV

20 – 40 m	5	2	11	4	9	5	4	3
100 – 150 m	4	2	6	2	N/A	N/A	2	2
600 -800 m	4	1	3	1	N/A	N/A	3	3