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## 1 Seasonal variability of the surface ocean carbon cycle: a

## 2 synthesis

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40 The seasonal cycle is the dominant mode of variability in the air-sea  $CO_2$  flux in most regions 41 of the global ocean, yet discrepancies between different seasonality estimates are rather large. 42 As part of the Regional Carbon Cycle Assessment and Processes phase 2 project 43 (RECCAP2), we synthesize surface ocean  $pCO_2$  and air-sea  $CO_2$  flux seasonality from 44 models and observation-based estimates, focusing on both a modern climatology and decadal 45 changes between the 1980s and 2010s. Four main findings emerge: First, global ocean 46 biogeochemistry models (GOBMs) and observation-based estimates ( $pCO_2$  products) of 47 surface  $pCO_2$  seasonality disagree, primarily due to discrepancies in the seasonal variability 48 in surface DIC. Second, the seasonal cycle in  $pCO_2$  has increased in amplitude over the last 49 three decades in both  $pCO_2$  products and GOBMs. Third, decadal increases in  $pCO_2$  seasonal 50 cycle amplitudes in subtropical biomes for both  $pCO_2$  products and GOBMs are driven by 51 increasing DIC concentrations stemming from the uptake of anthropogenic  $CO_2$  ( $C_{ant}$ ). In 52 subpolar and Southern Ocean biomes, however, the seasonality change for GOBMs is 53 dominated by C<sub>ant</sub> invasion, whereas for pCO<sub>2</sub> products an indeterminate combination of C<sub>ant</sub> 54 invasion and climate change modulates the changes. Fourth, we have shown that biome-55 aggregated decadal changes in the amplitude of  $pCO_2$  seasonal variability are largely 56 detectable against both mapping uncertainty (reducible) and natural variability uncertainty 57 (irreducible), but not at the gridpoint scale over much of the northern subpolar oceans and 58 over the Southern Ocean, underscoring the importance of sustained high-quality seasonally-59 resolved measurements over these regions.

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## 61 Plain Language Summary

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63 Changes in the amplitude of seasonal variations in the surface ocean carbon dioxide partial 64 pressure  $(pCO_2)$  over the period 1985-2018 are described over the historical period spanning 65 1985-2018, using both observation-based and model-based estimates. We identify increasing 66  $pCO_2$  seasonality over most regions that is largely driven by the impact of the invasion flux of 67 anthropogenic carbon into the surface ocean, with observation-based products also revealing

68 important modulations of  $pCO_2$  seasonality in high latitude regions also being impacted by 69 perturbations to the climate system. We also identified that there are important discrepancies 70 between observation-based and modeled  $pCO_2$  seasonality over global scales, with much of 71 this likely associated with systematic biases in model representations of the seasonal cycle in 72 surface dissolved inorganic carbon (DIC). Both reducible and irreducible forms of uncertainty 73 in monitoring  $pCO_2$  seasonality changes are quantified, with both cases highlighting the need 74 for sustained seasonally-resolving measurements over the high latitudes as part of an 75 optimized marine carbon observing system.

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77 Key points:

- (1) Changes in *p*CO<sub>2</sub> seasonal cycle amplitude over 1985-2018 are detectable against both
   mapping uncertainty and natural variability uncertainty.
- 80 (2) The dominant driver of pCO<sub>2</sub> amplitude increases over decadal timescales is attributed
   81 to the direct effect of C<sub>ant</sub> invasion.
- (3) A discrepancy is identified with surface DIC variability being systematically lower in
   GOBMs than in surface DIC observation-based products.
- 84 **1** Introduction
- 85

86 How is the ocean carbon cycle changing as a consequence of sustained increases in emissions 87 of carbon to the atmosphere? Important steps toward answering this question over the last 88 several decades have been provided via estimates of ocean carbon uptake from both interior 89 hydrographic measurements (Gruber et al., 2019; Sabine et al., 2004), surface fluxes inferred 90 from measurements of the sea surface partial pressure of  $CO_2$  ( $pCO_2$ ) (Chau et al., 2022; 91 Gregor et al., 2019; Landschützer et al., 2014; Rödenbeck et al., 2015), global ocean 92 biogeochemistry model simulations (Friedlingstein, et al., 2022; Hauck et al., 2020; Orr et 93 al., 2001) and ocean inverse models (Gloor et al., 2003; Gruber et al., 2009). A first global 94 synthesis was performed roughly a decade ago through the REgional Carbon Cycle 95 Assessment and Processes (RECCAP) project (https://www.globalcarbonproject.org/reccap), 96 highlighting the inevitable forced carbon cycle changes, while also identifying sources of 97 uncertainty.

99 In a parallel direction of inquiry, it has also become clear that the seasonal cycle in surface 100 ocean  $pCO_2$  (and thereby air-sea  $CO_2$  flux) has been changing, with this first identified in 101 modeling studies (Rodgers et al., 2008; Riebesell et al., 2009; Gorgues et al., 2010; Hauck 102 and Völker, 2015; Gallego et al., 2018) and subsequently inferred from observational 103 constraints (Fassbender et al., 2018; Landschützer et al., 2018) and more recently also 104 identified from changes in the seasonal cycle of atmospheric CO<sub>2</sub> induced by air-sea CO<sub>2</sub> 105 fluxes in the Southern Ocean (Yun et al., 2022). Studies to date show that the dominant 106 drivers of this trend towards increasing  $pCO_2$  seasonal amplitude are the increasing surface 107 concentrations of dissolved inorganic carbon (DIC), due to the invasion of anthropogenic  $CO_2$ 108 into the ocean, and the associated decline in surface ocean buffering capacity. The trend 109 towards increased seasonal amplitude is also expected to be modulated by natural variability 110 and by warming (Schlunegger et al., 2019). The net effect of warming in high emission 111 projections by the end of the 21st century is to enhance increases in the seasonal amplitude of 112 CO<sub>2</sub> fluxes over the expansive subtropical domain by approximately 15% (Rodgers et al., 113 2020).

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115 Our synthesis effort builds on the earlier studies of Fassbender et al. (2018), Landschützer et 116 al. (2018), and Fassbender et al. (2022), but here we rely on a greatly expanded set of  $pCO_2$ 117 products and GOBMs, spanning the time period 1985 to 2018, to assess the drivers of multi-118 decadal changes in the seasonal cycle of surface  $pCO_2$ . Such changes are of interest for three 119 primary reasons. First, they may impact stressors of marine ecosystems, as emphasized in the 120 analysis of future changes in pH, hydrogen ion concentration, and calcium carbonate mineral 121 saturation state seasonality by Kwiatkowski and Orr (2018). Second, they can have potential 122 impacts for climate feedback mechanisms, as demonstrated by Fassbender et al. (2022) for 123  $21^{st}$  century climate change under both strong and moderate CO<sub>2</sub> emissions pathways. Third, 124 they will have implications for optimizing the design of the global marine carbon observing 125 system, as summer-biased measurements can lead to significant errors for a system where the 126 seasonal cycle itself is not stationary.

128 For the majority of the global ocean, the seasonal cycle in sea surface  $pCO_2$  exhibits strong 129 variability that is up to two orders of magnitude larger than the seasonal cycle in atmospheric 130  $pCO_2$  (e.g. Takahashi et al., 2002). Seasonal variations in  $pCO_2$  reflect the interplay of four 131 principal underlying drivers: sea surface temperature (SST), sea surface salinity (SSS), 132 dissolved inorganic carbon concentrations (DIC), and total alkalinity (TA) (Takahashi et al., 133 1993, 2002). Variations in these drivers are the result of changes in ocean circulation and 134 mixing, biological processes, as well as exchanges of heat, freshwater, and carbon with the 135 atmosphere. While the temperature-driven or thermal component of observed  $pCO_2$ 136 seasonality is well constrained by laboratory studies and by high-quality satellite-derived SST 137 products, this is not the case for the other drivers. Therefore, observational studies often refer 138 to changes related to DIC, TA, or salinity collectively as changes in the nonthermal 139 component (Takahashi et al., 1993, 2002). Importantly, thermal and nonthermal drivers of 140  $pCO_2$  seasonality are typically found to be opposed in phase and large in amplitude relative to 141 the resulting net  $pCO_2$  variations. For this reason, skillful modeling of seasonal  $pCO_2$  cycling 142 can be compromised if any of the underlying processes are not well-represented, both for the 143 amplitude and the phase/timing of seasonality changes. Therefore, state-of-the-art models still 144 face challenges in skillfully simulating the seasonal cycle of  $pCO_2$  in regions that are known 145 to be important for  $CO_2$  exchange with the atmosphere (Anav et al., 2013; Goris et al., 2018; 146 Kessler amd Tjiputra, 2016; Mongwe et al., 2018; Pilcher et al., 2015). For the important case 147 of the North Atlantic, it has been shown that future carbon uptake in ESM projections can be 148 constrained by diagnosing the timing and magnitude of modeled seasonal variations in  $pCO_2$ 149 (Goris et al., 2018). Similarly, Nevison et al. (2016) demonstrated, for the Southern Ocean, 150 that the way an ESM represents the seasonal carbon cycle is related to the model's projected 151 carbon uptake. A much weaker seasonal cycle than in higher latitudes is observed in the 152 tropics, where seasonal  $pCO_2$  amplitudes are obscured by interannual variability. In polar 153 latitudes where there is seasonal ice cover, the challenge of data sparsity associated with the 154 difficulty of sampling  $pCO_2$  through the seasonal cycle presents challenges and uncertainty 155 that are beyond the scope of this study.

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157 An important source of uncertainty with detecting anthropogenic trends in the carbon cycle is 158 natural variability. As a matter of definition, we consider the forced anthropogenic signal to 159 represent the sum total of the effects of anthropogenic forcing on atmospheric radiation 160 (greenhouse gases, anthropogenic aerosols, etc.) and thereby warming ensuing physical and 161 biogeochemical state changes in the ocean, as well as the effect of the invasion flux of 162 anthropogenic carbon into the ocean. For the analysis presented here, even if  $pCO_2$  products 163 were able to perfectly resolve multi-decadal changes in the seasonal amplitude of sea surface 164  $pCO_2$  over 1985 to 2018, it would not be possible to confidently isolate the anthropogenic 165 trend from the impacts of natural variability. Given that natural variability is an intrinsic 166 property of the Earth system, natural variability uncertainty is understood to be an irreducible 167 form of uncertainty that can nevertheless be quantified. To address the degree to which 168 natural variability obscures detection of anthropogenic trends, we include analyses of large 169 ensemble (LE) simulations with Earth system models (ESMs) in this study. LE simulations 170 were considered previously in the study of Schlunegger et al. (2019) for the case of 21st 171 century climate change projections under strong emissions, and they showed that over the 172 subtropics forced changes in  $\Delta p CO_2$  seasonal amplitude are more emergent than forced trends 173 in annual mean  $\Delta p CO_2$ .

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175 Previous research has pointed towards significant model biases in representing the seasonal 176 cycle of  $pCO_2$  and its underlying drivers (Goris et al., 2018; Mongwe et al., 2018; Rodgers et al., 2014). The question naturally arises as to whether models have improved since the 177 178 evaluation of seasonal variability in an early generation of prognostic biogeochemistry 179 models (McKinley et al., 2006; Tjiputra et al., 2012). For the case of the North Pacific, the 180 forward models in McKinley et al. (2006) had their seasonal cycles in  $pCO_2$  evaluated 181 directly against time series measurements from observing stations spanning the subpolar and 182 subtropical gyres to identify non-trivial biases in models. Our study differs in that, rather than 183 direct comparisons with time series stations, our observation-based data sources consist of mapped global sea surface  $pCO_2$  products constructed from the same underlying ungridded 184 185 SOCAT (Surface Ocean CO<sub>2</sub> Atlas; Pfeil et al., 2013) data, as well as ocean interior seasonal 186 climatological fields of DIC.

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188 More extensive analyses for the regions with seasonal ice cover, which are not considered 189 here due to their sparsity of carbon observations for estimating the seasonal cycle, will be

190provided by the Arctic and Southern Ocean regional contributions to RECCAP2. Our analysis191and synthesis is developed using a diverse set of resources: nine observationally-based  $pCO_2$ 192products, three of which include associated time-varying monthly surface DIC and TA fields;193two independent three-dimensional seasonally-resolved DIC climatologies; hindcast194simulations from eleven GOBMs; five large ensemble simulations; and individual realizations195of 11 CMIP6 models.

196

197 This paper is structured as follows. We begin in the methods section by introducing the 198 RECCAP2  $pCO_2$  products and GOBMs, along with ancillary products used for this study. In 199 the main results section, we present a descriptive account of changes in  $pCO_2$  seasonality as 200 well as discrepancies between  $pCO_2$  products and GOBMs, followed by attribution of these 201 changes and discrepancies to their drivers. We finally present an analysis focused on 202 identifying detectability of forced/anthropogenic signals, as well as an assessment of the 203 robustness of our chosen biome aggregation for detectability of the forced changes in seasonality to changes due to natural variability. Our objective is to present a synthesis of the 204 205 carbon cycle community's current knowledge of how the seasonal cycle of sea surface  $pCO_2$ 206 and the air-sea CO<sub>2</sub> exchange has been changing over the last three decades.

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#### 208 **2 Methods**

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## 210 **2.1 Considered regions**

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Ocean biomes offer a number of advantages as aggregation regions for studying the global carbon cycle (Fay and McKinley, 2014). For our interests in seasonality, biomes appropriately reflect the structures that are determined by real-world interactions between ocean circulation and biogeochemistry. For all of the oceanic studies within RECCAP2, a discrete number of ocean biomes based on Fay and McKinley (2014) are used to facilitate consistent intercomparison between regions (described in the RECCAP2 Global Ocean contribution by DeVries et al., submitted to the RECCAP2 special issue). This study analyzes  $pCO_2$  and  $CO_2$  219 flux seasonality for six biomes (Table 1 and central panel of Fig. 1) aggregated from the 220 aforementioned. Our decision to aggregate some biomes and exclude others is based on the 221 following: First, this is a synthesis study and the seasonal cycle in surface ocean  $pCO_2$ 222 exhibits important similarities across the subtropics of each hemisphere. Second, we have 223 elected to not include either the Arctic, the Bering Sea, or the regions of the Southern Ocean 224 that are impacted by ice cover, with the analysis of those regions left to the individual 225 regional RECCAP2 studies. Third, we have chosen to not include the equatorial regions, 226 given that the seasonal cycle there is relatively weak and often obscured by large interannual 227 variability.

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229 Thus, our six aggregated biomes (Table 1 and Fig. 1) consist of North Atlantic subpolar 230 seasonally stratified (NA-SPSS), North Pacific subpolar seasonally stratified (NP-SPSS), 231 Northern Hemisphere subtropical seasonally stratified (NH-STSS), Northern Hemisphere 232 subtropical permanently stratified (NH-STPS), Southern Hemisphere subtropical permanently 233 stratified (SH-STPS), and Southern Hemisphere seasonally stratified (subpolar and 234 subtropical combined, SH-SS). We have elected to keep the North Atlantic and North Pacific 235 subpolar biomes separate in our analysis in light of important differences that arise in their 236 seasonal cycles.

237

Besides the aggregation as designated in **Table 1**, adjustments have been taken to avoid confounding changes in observational coverage with seasonality or changes in seasonality. We limit our analysis to regions that have valid observation-based (and GOBM) fields for the whole time period 1985-2018 for all  $pCO_2$  products and for all variables considered in this study (CO<sub>2</sub> fluxes,  $pCO_2$ , SST, SSS, surface DIC, and surface TA). Thereby we make sure that the same region is considered for all variables over the whole seasonal cycle but also over the whole observational period (see Fig.1).

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#### 246 **2.2. Temporal filtering of RECCAP2 products**

248 In general, the monthly mean  $pCO_2$  products and GOBMs considered here can be expected to 249 represent  $pCO_2$  seasonality modulations that are sustained through the combined effects of the 250 invasion flux of Cant, natural variability, and warming. In order to identify the amplitude of 251 the seasonal cycle (with its modulations), we follow Landschützer et al. (2018) in using a 252 quadratic polynomial fit to remove the decadal trend from monthly time series over 1985-253 2018. Our analysis through the majority of this manuscript characterizes multi-decadal 254 changes in  $pCO_2$  seasonal variations by considering differences between the five-year 255 intervals 1985-1989 and 2014-2018. We also investigated the degree to which the main 256 results are sensitive to the choice of five-year versus 10-year versus 15-year intervals for 257 describing climatologies (Fig. S3), and upon finding that the results are relatively insensitive 258 to the approach we opted to use the difference between the two five-year intervals 1985-1989 259 and 2014-2018. As such, our approach is consistent with that used by Landschützer et al. 260 (2018), who also used five-year climatologies in calculating decadal changes.

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## 262 **2.3.** Surface observation-based products of *p*CO<sub>2</sub>, DIC, and TA

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264 Observation-based  $pCO_2$  products that are compared for their representation of  $pCO_2$ 265 seasonality are listed in **Table 2**. All of these products are built on surface ocean  $pCO_2$ 266 measurements within the Surface Ocean  $CO_2$  Atlas database (SOCAT, Bakker et al., 2016) yet they differ significantly in the way they spatiotemporally interpolate or map the sparse 267 268 observations. Methodologically they include multiple linear regression (Iida et al., 2021), 269 gradient boosted decision trees (Gloege et al., 2022), neural networks (Chau et al., 2022; 270 Landschützer et al., 2016; Zeng et al., 2022), ensembles of various machine learning approaches (Gregor et al., 2019; Gregor and Gruber, 2021), and a Bayesian approach that is 271 272 additionally constrained by mixed-layer dynamics (Rödenbeck et al., 2013, 2022). 273 Additionally, one neural network product applies adjustments to the SOCAT data to account 274 for temperature gradients between the ocean surface and the depth of the seawater intake on 275 the observing ships and for the cold skin layer effect (Watson et al., 2020).

277 Of central importance to our study is that three of these products (JMAMLR, OceanSODA-278 ETHZ, and CMEMS-LSCE-FFNN) additionally provide time-varying surface DIC and TA 279 products spanning 1985-2018. While we use all available  $pCO_2$  products listed in **Table 2** for 280 our analysis of seasonal cycles of  $CO_2$ -fluxes and  $pCO_2$ , our analysis greatly benefits from 281 using these three products to attribute changes of  $pCO_2$  seasonality to its drivers. We will 282 refer to them as the pCO<sub>2</sub>/TA products. We provide a summary of how DIC and TA are 283 derived for these three  $pCO_2$  products in the Supplementary Materials, but we refer the 284 reader to the papers cited in Table 2 for a more complete description of methods for the 285 individual observation-based products.

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- 287 2.4. Three-dimensional DIC climatologies
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289 For our evaluation of seasonal variability in surface DIC concentrations, we consider two 290 three-dimensional DIC climatologies constructed from observational products: the Mapped 291 Observation-Based Oceanic DIC (MOBO-DIC) product by Keppler et al. (2020a, 2020b), and 292 the NNGv2LDEO monthly climatology of interior DIC by Broullón et al. (2020). Unlike the 293  $pCO_2/TA$  products, these products are based on direct observations of DIC and as such 294 provide an independent estimate of surface DIC seasonality, with the caveat that they offer a 295 climatological view only. Both of these three-dimensional DIC climatologies are described in 296 greater detail in the Supplementary Materials.

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## 298 2.5. Global Ocean Biogeochemistry Models (GOBMs)

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Global ocean biogeochemistry models (GOBMs) are compared against the  $pCO_2$  products for modulations of CO<sub>2</sub> fluxes and  $pCO_2$  for the six aggregated biomes over the period 1985-2018. Many of the GOBMs considered here have contributed to the Global Carbon Project's estimate of the ocean carbon sink (Friedlingstein et al., 2022), and have been evaluated by Hauck et al. (2020). Both the phase and amplitude of seasonal variability in the GOBMs will be evaluated against the  $pCO_2$  products, as will the behavior of the drivers (SST, DIC, SSS,

306 and TA). The GOBMs are listed in Table 3, and they are further described for their 307 initialization, forcing, and resolution in the submitted global RECCAP2 study of DeVries et 308 al. All but one of the GOBMs listed in Table 3 provide the full suite of variables and 309 timescales needed for this analysis. The exception is the CCSM model for which output is 310 available only through 2017 (rather than 2018). For this model alone we construct 311 climatologies over 2014-2017. Additionally we have included the abiotic data-assimilation 312 model OCIM (DeVries, 2014, 2022) where appropriate in our analysis. Although more 313 commonly applied to the uptake of anthropogenic carbon, the abiotic nature of the OCIM 314 model is of value as an endmember case in evaluating potential biotic biases in the GOBMs.

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Air-sea heat fluxes for the GOBMs used throughout RECCAP2 are calculated with bulk formulae following the protocols of Large and Yeager (2009). This method does not impose a specific nudging or restoring of simulated sea surface temperature (SST) to observed SST, but rather imposes a negative feedback to SST through heat fluxes determined using observed surface boundary layer atmospheric temperatures. In this way, the simulation of seasonal variations in SST shows strong fidelity to observations when aggregated over biome scales.

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## 323 2.6. Attribution Analysis

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## 325 2.6.1 Attribution of drivers of the climatological seasonal cycle in *p*CO<sub>2</sub>

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327 The first stage of the attribution analysis has the goal of identifying the mechanisms 328 responsible for discrepancies between  $pCO_2$  products and GOBMs in representing the 329 climatological seasonal cycle of  $pCO_2$ . We will consider both a decomposition into thermal 330 and nonthermal drivers, using the analysis methods presented by Fassbender et al. (2022). 331 The thermal/nonthermal decomposition provides a valuable means to identify spatially 332 regions where seasonal SST variations are or are not the dominant driver of seasonal 333 variations in  $pCO_2$ . We will also apply the Taylor Series decomposition method previously 334 considered by Sarmiento and Gruber (2006). A Taylor Series decomposition is a more extensive decomposition into individual drivers of the seasonal cycle in  $pCO_2$ , where we are specifically interested in parsing the contributions of SST, SSS, DIC, and TA. In this way we will evaluate any discrepancies that may call into question the fidelity of modeled  $pCO_2$  to the real ocean. This analysis is possible for all GOBMs and the three observation-based products that include DIC and TA.

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Additionally mixed layer depth (MLD) products will be used as part of our attribution analysis. For the sake of consistency with our choice of the relatively data-rich period 2014-2018 for evaluating climatologies of pertinent variables, we present a MLD product for this period 2014-2018 derived from the gridded monthly Argo product of Roemmich and Gilson (2009) for temperature and salinity. We define MLD by a density threshold through application of the MLD definition of Holte and Talley (2009).

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## 348 **2.6.2** Attribution of decadal changes in the seasonal cycle of *p*CO<sub>2</sub>

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350 The second stage of attribution analysis consists of applying the method of Fassbender et al. 351 (2022) to identify the drivers of decadal changes in  $pCO_2$  seasonality over 1985-2018. The 352 method provides a means to separate the impacts of the invasion flux of anthropogenic carbon 353 from the atmosphere from other climate signals that can modulate the seasonal cycle in  $pCO_2$ . 354 More specifically, the goal for this work will be to identify the direct impact of an increase in 355 the anthropogenic carbon (Cant) content (i.e., a decrease of the surface ocean buffering 356 capacity) relative to climate change and natural variability impacts on  $pCO_2$  seasonality 357 changes over 1985-2018. The original application by Fassbender et al. (2022) was for 21<sup>st</sup> 358 century LE projections with an ESM, where the ensemble mean was applied to identify the 359 forced component of change. Here our intention is to apply the method to the period 1985-360 2018 for all GOBMs (**Table 3**) and gridded  $pCO_2$  products that include associated monthly 361 DIC and TA fields along with SST and SSS. A description of the method is provided in the Supplementary Materials. 362

### 364 **2.7.** Uncertainty analysis for detecting the anthropogenic signal

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366 As in the study of Fassbender et al. (2022), we apply large ensemble (LE) simulations with 367 ESMs to aid in our interpretation of forced changes in  $pCO_2$  seasonality in the presence of 368 non-negligible background variability. Whereas Fassbender et al. (2022) focused on 369 centennial timescale projections and could rely on the ensemble mean to isolate forced 370 signals, we are here faced with a different challenge of interpreting historical records for which we have only three out of nine  $pCO_2$  products that are viable for attribution analysis, all 371 372 aiming to represent the same individual realization experienced by the chaotic Earth system. 373 Thus, LE simulations will be applied to interpret changes measured over the historical period 374 1985-2018 to estimate uncertainty in isolating a forced change in the presence of natural 375 background variability.

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377 The LE models considered here are listed in Table 4. The first set of models were run under 378 CMIP5 protocols using historical/RCP8.5 forcing (CanESM2, ESM2M, and CESM1) and the 379 second set were run under CMIP6 protocols with historical/SSP3-7.0 forcing (CanESM5 and 380 CESM2). Such a mix of CMIP5 and CMIP6 is appropriate to our interest in the historical 381 period spanning 1985-2018, as the total radiative forcing component for the CMIP5 and 382 CMIP6 forcing pathways is very similar through 2018 (Riahi et al., 2017). These LE outputs 383 are not part of the resources made available through RECCAP2, but were rather collected by 384 the authors through a modest expansion of the collection of LE models considered by 385 Schlunegger et al. (2020) and Gloege et al. (2021). We are particularly interested in 386 evaluating confidence levels for emergent changes between the time intervals 1985-1989 and 387 2014-2018, where the ensemble mean changes (signal) and natural variability in the changes 388 (noise) provide a means to identify the signal-to-noise ratio (SNR) to characterize the degree 389 of emergence or detectability.

390

For the attribution of decadal changes in  $pCO_2$  seasonal amplitude, we have applied the CESM2-LE, as this was deemed to have better correspondence with climatological  $pCO_2$ variability over biome scales than the other models (this will be addressed in **Fig. 12a**). The application of CESM2-LE will allow us to address the degree of confidence that we have in distinguishing the impact of the invasion flux of anthropogenic carbon from climate-driven perturbations. CESM2-LE is also applied to identify the degree to which thermal and nonthermal drivers of  $pCO_2$  seasonality changes are emergent.

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#### 399 **2.8. CMIP6 models**

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401 This study also includes analyses of 11 CMIP6 ESMs that are evaluated for their agreement 402 with the  $pCO_2$  products over biome scales, with the models listed in **Table 6**. As ESMs are 403 used for future projections of the ocean carbon sink, it is of interest to assess their fidelity in 404 reproducing the seasonal cycle and its underlying processes. The CMIP6 models are not 405 intended to correspond to coupled versions of the GOBMs considered here, but our goal is 406 rather to identify similarities or discrepancies between these models, the GOBMs, and 407 observational  $pCO_2$  products. For the CMIP6 models we have opted to focus on a slightly 408 different period, namely the five years (2010-2014) at the end of the historical component of 409 CMIP6 simulations, rather than the 2014-2018 interval considered for the GOBMs.

410

#### 411 3 Results

412

## 413 **3.1.** Overview of Multi-Decadal Changes in CO<sub>2</sub> Flux and *p*CO<sub>2</sub> seasonality

414

The evolution of air-sea  $CO_2$  flux seasonality, considered as averages over the six biomes, is shown in **Fig. 1** as time series of the annual minimum and annual maximum of the monthly air-sea  $CO_2$  fluxes for the  $pCO_2$  products and GOBMs. Here, we have chosen to show the maximum and minimum of the fluxes to represent the full range of the seasonal cycle. One should keep in mind that the across-product ensemble spread also includes the impacts of wind speed on gas exchange that are not treated the same way across products (more details are provided in the submitted DeVries et al. global ocean contribution to RECCAP2).

423 A prominent feature in **Fig. 1** is that the amplitude of the seasonal cycle in  $CO_2$  fluxes is 424 consistently larger for the GOBMs than it is for the  $pCO_2$  products, across all biomes (see also 425 **Fig. S1)**. From the RECCAP2 GOBMs alone it is not possible to determine the degree to 426 which these internal disagreements reflect structural differences between the GOBMs and the 427 degree to which it represents differences in forcing of the GOBMs. It is also worth noting that 428 both the GOBMs and  $pCO_2$  products exhibit a non-negligible degree of natural variability 429 (interannual to decadal), for both the maxima and minima.

430

431 A decadal trend towards increased seasonal amplitude of biome-integrated  $CO_2$  fluxes is 432 evident over the subtropical biomes, as well as for NP-SPSS, whereas for NA-SPSS and SH-433 SS biomes this is less evident (especially in the  $pCO_2$  products, see also Fig. S1). The 434 increase in the amplitude, where it exists, can be accounted for mainly by a decrease of the 435 annual minima (i.e., an increase of the uptake flux), whereas the annual flux maxima 436 remained comparably constant over time. To better constrain whether there are discernable 437 decreases in the spread amongst the  $pCO_2$  products over 1985-2018 as  $pCO_2$  observations 438 increase, the biome-averaged  $pCO_2$  for seasonal maximum and seasonal minimum are shown 439 in Fig. S2, where although there is a decrease for both NA-SSS, SH-STPS, and SH-SS 440 between 1985 and 2018, for the other biomes the changes are rather small. The broad-scale 441 view of the evolution of CO<sub>2</sub> fluxes in Fig. 1 serves to motivate much of the analysis that 442 follows, with emphasis devoted to  $pCO_2$  rather than  $CO_2$  fluxes as a means to facilitate 443 attribution and mechanistic understanding. Nevertheless we will return to CO2 fluxes in the 444 discussion of decadal trends in seasonal cycle amplitude.

445

#### 446 **3.1.1** Climatological seasonal cycle of *p*CO<sub>2</sub> and multi decadal changes

447

The climatological monthly  $pCO_2$  seasonal cycle over 2014-2018 as a zonal mean is shown in Fig. 2. For the  $pCO_2$  products (Fig. 2a),  $pCO_2$  is highest in the subtropics in summer and highest in the Northern Hemisphere subpolar region and in the Southern Ocean in winter. The GOBMs (Fig. 2e) are in agreement with the  $pCO_2$  products in their seasonal phasing in the subtropics but are in disagreement in the Northern Hemisphere subpolar regions and the 453 Southern Ocean. As a measure of the internal consistency across the nine  $pCO_2$  products and 454 the 11 GOBMs for this 2014-2018 climatology, the standard deviation is calculated month-455 by-month across the  $pCO_2$  products (Fig. 2b) and across the GOBMs (Fig. 2f). From this we 456 learn that the agreement across the  $pCO_2$  products is robust (<10% disagreement relative to 457 the annual cycle amplitude), whereas internal disagreements are much larger across the 458 GOBMs (larger than 20% of the annual cycle amplitude at some locations), with the GOBMs 459 diverging the most north of 30°N and south of 30°S. For the Northern Hemisphere subpolar 460 regions, the disagreement in the GOBMs is largest in summer, and over the Southern Ocean it 461 is largest in summer and in late winter.

462

We also consider the multi-decadal changes in the climatological monthly  $pCO_2$  seasonal cycle between 1985-1989 and 2014-2018 for the  $pCO_2$  products (**Fig. 2c**) and for the GOBMs (**Fig. 2g**). For both the  $pCO_2$  products and the GOBMs (**Fig. 2c** and **Fig. 2g**, respectively), the meridional patterns of increases in seasonal amplitude to first order reflect and amplification of the climatological seasonal cycle in the Northern Hemisphere subtropical and subpolar biomes. However, this is less clear in the Southern Hemisphere subtropics and the Southern Ocean.

470

#### 471 **3.1.2 Spatial structure in the seasonal amplitude change**

472

473 To understand the spatial structure of the amplitude change and the coherence between  $pCO_2$ 474 products and between GOBMs, maps of changes in the amplitude of the  $pCO_2$  seasonal cycle 475 between 1985-1989 and 2014-2018 are shown for the  $pCO_2$  products in Fig. 3a and for the 476 GOBMS in Fig. 3b. We use two definitions for the amplitude assessment: winter minus 477 summer (JFM minus JAS for the Northern Hemisphere and JAS minus JFM for the Southern 478 Hemisphere) in Figs. 3a and 3b and climatological monthly maximum minus minimum in 479 **Figs.** 3c and 3d. For both the GOBMs and the  $pCO_2$  products, regions where the multi-480 decadal trends are not significant (defined using a one standard deviation threshold) are 481 stippled. From this we can see that the Northern Hemisphere has more significant changes 482 than the Southern Hemisphere. Nevertheless for both hemispheres for GOBMs and  $pCO_2$  products the broadest regions of significant changes are in the subtropics, although there isalso a significant trend in the eastern subpolar North Pacific.

485

486 The two different definitions of seasonal amplitude (winter-minus-summer and maximum-487 minus-minimum) result in very similar large-scale patterns, although, due to their definition, 488 with largely opposite sign. While for the maximum-minus-minimum definition positive 489 changes always indicate an increase in seasonal amplitude, the winter-minus-summer 490 amplitude is negative over the subtropical domain and positive in high latitudes, such that 491 negative (positive) values for the multi-decadal amplitude change in Fig. 3a and 3b over the 492 subtropics (high latitudes) also indicate an amplitude increase. Phasing differences in 493 seasonality are treated differently with the two definitions, since winter-minus-summer refers 494 to fixed time intervals while the maximum and minimum can occur at any time throughout 495 the year. While the winter-minus-summer definition highlights differences in phasing 496 between  $pCO_2$  products and GOBMs, it conceals some of the changes in the seasonal 497 amplitude, specifically if the maximum or minimum of the seasonal cycle occurs outside of 498 winter or summer months (as seen in Fig. 3d in the high latitude North Atlantic and North 499 Pacific, where the discrepancy between GOBMs and  $pCO_2$  products is much larger for the 500 winter-minus-summer definition).

501

502 For the remainder of our analysis we will use the winter minus summer definition of 503 seasonality due to its simplicity when it comes to analysis of multiple models and products, 504 and also for consistency with previous work (e.g., Landschützer et al., 2018), keeping in mind 505 that the specifics of this definition (the choice of JFM and JAS for defining seasonal 506 amplitude) are sensitive to phasing differences between GOBMs and  $pCO_2$  products. The 507 sensitivity of the choice of five-year versus 10-year versus 15-year climatologies for the case 508 of the  $pCO_2$  products (Fig. S3) indicates that the results are robust to the length of the 509 climatologies, the main difference being that using 10-year or 15-year climatologies shortens 510 the effective decadal-change interval and weakens the signal of interest.

512 The fields shown in Fig. 3a and 3c reveal important information about uncertainty in 513 detecting multi-decadal changes in  $pCO_2$  seasonality using  $pCO_2$  products. As all of the 514 products are based upon the same un-gridded SOCAT data product, differences in multi-515 decadal changes shown in Fig. 3a and 3c are due to mapping differences rather than 516 differences in the basis data itself. As such, the stippled regions indicate where mapping 517 uncertainty is large relative to resolved decadal changes. We interpret mapping uncertainty to 518 be a reducible variety of uncertainty, in that with improved data coverage and expansion of 519 the ocean observing system the products should be expected to converge.

520

For the GOBMs (**Figs. 3b** and **3d**) the regions of significance are very similar to what is shown for the  $pCO_2$  products, but the underlying reasons for uncertainty are of course fundamentally different. For the GOBMs, regions where trends are not significant reflect the combined effect of structural differences between models and differences in GOBM forcing (described in the submitted global RECCAP2 contribution of DeVries et al. for differences in forcing). The uncertainty in **Fig. 3b** and **3d** for the GOBMs should also be considered as a reducible variety of uncertainty.

528

Nevertheless it should also be noted that for both  $pCO_2$  products and GOBMs that it is the subtropics where the thermal component of  $pCO_2$  seasonality has the weakest opposition from the nonthermal component that there is emergence. Thus both mapping uncertainty and presumably structural uncertainty with GOBMs are mostly problematic in regions where the nonthermal drivers become important. This will be discussed further in subsequent analyses of this study.

535

## 536 **3.1.3 Biome aggregated climatologies**

537

538 Climatological monthly anomalies in  $pCO_2$  and its primary drivers, SST and surface DIC, 539 relative to their annual means are displayed in **Fig. 4** as (area-weighted) averages across the 540 RECCAP2 biome regions. A first comparison between the full collection of  $pCO_2$  products 541 (blue) and all GOBMs (green) is shown in the first column of **Fig. 4** by biome region, where 542 continuous lines and associated shading represents the mean and one standard deviation, 543 respectively. Our first objective is to identify discrepancies between the GOBMs and the 544  $pCO_2$  products, with such discrepancies to be addressed subsequently in the **Attribution** 545 section of the paper.

546

547 In broad terms, the seasonal phasing agreement between GOBMs and  $pCO_2$  products is best 548 in subtropical biomes (NH-STSS, NH-STPS, and SH-STPS) and worst in the Northern 549 Hemisphere subpolar and the Southern Ocean biomes (NA-SPSS, NP-SPSS, and SH-SS). In 550 the NA-SPSS and NP-SPSS biomes, GOBMs consistently represent seasonal pCO<sub>2</sub> minima 551 that occur earlier than in the  $pCO_2$  products, and that are weaker for NA-SPSS. For NA-SPSS 552 and NP-SPSS, after the spring minimum in  $pCO_2$  in the GOBMS there is an increase towards 553 a maximum in August, whereas the  $pCO_2$  products remain low throughout the summer before 554 increasing in autumn and winter towards a maximum in January/February. Analogous to what 555 is found in the Northern Hemisphere subpolar biomes, there are also discrepancies in phasing 556 for the GOBMs relative to the  $pCO_2$  products for SH-SS. For the higher latitude regions 557 where phasing in models diverges greatly from that seen in the  $pCO_2$  products, it is widely 558 understood that the early summer minimum in  $pCO_2$  (as seen in  $pCO_2$  products) is directly 559 related to biological drawdown of DIC (Gregor and Gruber, 2021). It is a known bias of 560 coupled models that primary production is often not realistically sustained through summer 561 after a pronounced spring bloom. This has been shown in the North Atlantic (Goris et al., 562 2018), and more broadly for the high latitudes of both hemispheres (Cabré et al., 2016; 563 Mongwe et al., 2018; Nevison et al., 2016), and the same limitation appears to be pertinent in 564 our study.

565

In the subtropical biomes, on the other hand, the GOBMs consistently simulate a  $pCO_2$ seasonal cycle that is larger than what is seen in the  $pCO_2$  products (**Table 5**). For NH-STSS the amplitude averaged across the GOBMs is 80% larger than for that averaged across the  $pCO_2$  products, for NH-STPS it is 26% larger, and for SH-STPS it is 34% larger. This occurs with phasing that is largely consistent between the GOBMs and  $pCO_2$  products. As seen in **Fig. 4** (second column), this discrepancy in the amplitude of  $pCO_2$  variations does not reflect 572 biases in the seasonal amplitude of SST variations, as the thermodynamic boundary 573 conditions for forcing the GOBMs result in SSTs that have relatively strong fidelity to 574 observations when averaged over biome scales. Rather, the discrepancy in the amplitude of 575 the  $pCO_2$  seasonal amplitude indicates that surface DIC (Fig. 4, second column) as the main 576 driver of the nonthermal component of  $pCO_2$  seasonality is not providing sufficient 577 counterbalance to the realistic thermal component of seasonal forcing (i.e., the seasonal 578 amplitude seems to be too small in GOBMs). For the subtropical biomes it is worth noting 579 that the OCIM model (Fig. 4, thin line) has an even larger  $pCO_2$  seasonal amplitude than the 580 GOBMs. The OCIM model has a realistic SST seasonal evolution (not shown), but simulates 581 only circulation- and surface flux-driven DIC variations without including a contribution of 582 the biological drawdown of DIC. The OCIM  $pCO_2$  and DIC seasonal cycle amplitude 583 discrepancies relative to the GOBMs provide an estimate of the contribution of the biological 584 pump to DIC and  $pCO_2$  seasonality.

585

586 The fact that the DIC seasonal amplitude is weaker for the GOBMs relative to the  $pCO_2$ 587 products means that the nonthermal component of  $pCO_2$  seasonality is smaller. Over the 588 subtropical regions, this will serve to enhance the thermal dominance of seasonal  $pCO_2$ 589 variations and in the subpolar and circumpolar regions this will reduce the nonthermal 590 dominance of seasonal  $pCO_2$  variations. This provides a means to alter the phasing of  $pCO_2$ 591 variations, although the degree to which this occurs will be modulated by the background 592 mean state of DIC and TA (Fassbender et al., 2018). In all biomes, the DIC seasonal 593 amplitude for GOBMs lies between that of the abiotic OCIM model and that of the  $pCO_2/TA$ 594 products, suggesting that for the GOBMs there may be as-yet undetermined shortcomings in 595 the representation of the seasonal processes that increase DIC (entrainment) and decrease DIC 596 (net biological consumption of DIC).

597

598 Comparing the decadal changes (**Fig. 4**, right column) with the climatologies over 2014-2018 599 (left column), we see again that the decadal changes in  $pCO_2$  to first order amplify the 600 climatological seasonal cycle in the Northern Hemisphere as well as for SH-STSS, consistent 601 with what was identified in the Hovmöller diagrams in **Fig. 2**. Importantly, the  $pCO_2$  products 602 show significantly less internal disagreement for the climatological seasonal cycle (left

603 column of Fig. 4 and Table 5) than the GOBMs. Yet, for the decadal changes (right column 604 of Fig. 4), the spread across the GOBMs and across the  $pCO_2$  products is similar (one 605 standard deviation for the biome averaged seasonal amplitude changes are roughly between 1 606 and 10 ppm, **Table 5**). While this is of the same order of magnitude as for their seasonal 607 amplitude for the  $pCO_2$  products, GOBMs show significantly less internal disagreement for 608 the decadal change than for the seasonal amplitude. This indicates that the processes 609 responsible for the increase in seasonal  $pCO_2$  amplitude are relatively well represented in 610 GOBMs We hypothesize here that the pertinent process in the GOBMs is the invasion flux of 611  $C_{ant}$  dominating  $pCO_2$  seasonality modulations associated with climate-driven changes in the 612 state of the ocean, a hypothesis to which we will return in the Attribution section. The best 613 agreement for decadal changes in the  $pCO_2$  seasonal amplitude occurs in the subtropical 614 biomes, consistent with Fig. 2. For NH-STSS, the GOBMs show a mean net increase in the 615 seasonal amplitude of 10.7  $\mu$ atm and the pCO<sub>2</sub> products of 9.8  $\mu$ atm, with the difference in 616 their increase being less than 10%. For NH-STPS, the mean net increase in the seasonal 617 amplitude for the GOBMs and  $pCO_2$  products are the same at 6.3 µatm. For SH-STPS the 618 increase for GOBMs is 2.7  $\mu$ atm, which is approximately half of the mean pCO<sub>2</sub> product 619 increase of 5.3 µatm. It can also be seen in **Table 5** that the relatively consistent ranges of 620 change over the subtropical biomes between GOBMs and  $pCO_2$  products are approximately 621 within each other's uncertainty range.

622

## 623 **3.2. Attribution**

Having evaluated the seasonal cycle of  $pCO_2$  in terms of its phasing and amplitude, as well as related discrepancies between the  $pCO_2$  products and the GOBMs, we now turn our attention to attribution of the drivers. We have already inferred from the biome-aggregated analyses in **Fig. 4** that DIC should be explored as having a potential first-order role in modulating the differences between  $pCO_2$  products and GOBMs in the  $pCO_2$  seasonal cycle by serving as the main nonthermal driver.

630

#### 631 **3.2.1 Spatial patterns of the seasonal cycle in surface DIC concentrations**

633 The spatial patterns for the surface DIC seasonal cycle amplitude (winter minus summer, 634 climatology over 2014-2018) for the  $pCO_2/TA$  products is shown in Fig. 5a (average across 635 JMAMLR, CMEMS-LSCE-FFNN, and OceanSODA-ETHZ) as well as for the GOBMs in 636 Fig. 5b. We see a good agreement between the amplitude of the surface DIC seasonal cycle 637 from  $pCO_2/TA$  products and the GOBMs for the overall patterns with maximum values seen 638 in the Northern Hemisphere subpolar domains. For the  $pCO_2/TA$  products we also see a 639 zonally-oriented local surface DIC maximum across 40°S-45°S that can be distinguished from 640 a local minimum over the Antarctic Circumpolar Current (ACC) region between 50°S-60°S, 641 with this feature being more weakly present in the GOBMs. The differences in the seasonal 642 amplitude of DIC (GOBMs minus  $pCO_2$  products) in Fig. 5c indicates a negative bias of the 643 GOBMs that is relatively consistent over large scales, and that is particularly pronounced at 644 northern high latitudes .

645

The zonal average of the amplitude of the seasonal cycle (winter minus summer) in DIC is shown in **Fig. 5d**. Additionally the amplitude of the surface ocean DIC seasonal variability for the two three-dimensional DIC climatologies, namely MOBO-DIC and NNGv2LDEO are also shown. The MOBO-DIC product tends to have a seasonal amplitude that is somewhat larger than the surface DIC products over most latitudes, except over the Southern Ocean and north of 50°N. NNGv2LDEO is also larger than the GOBMs over most latitudes, also with an exception to the north of 50°N but not over the Southern Ocean.

653

654 The same collection of products considered as zonal means in Fig. 5d are shown as biome 655 averages in Fig. S4, along with the OCMIP product. This further illustrates that there is a 656 discrepancy between the weaker DIC seasonal amplitude of GOBMs relative to not only the 657  $pCO_2/TA$  products but also to MOBO-DIC and NNGv2LDEO, although the seasonal DIC 658 amplitude of MOBO-DIC is smaller than that of all other observation-based products (and 659 thus closer to the GOBMs) in the high latitude biomes (NA-SPSS, NP-SPSS, and SH-SS). 660 Interestingly, for both large permanently stratified biomes (NS-STPS and SH-STPS) the late 661 summer seasonal minimum in DIC concentrations for the MOBO-DIC and NNGv2LDEO 662 products precedes that of the  $pCO_2/TA$  products.

665

663

666 To systematically attribute discrepancies in the  $pCO_2$  seasonal cycle between GOBMs and 667  $pCO_2$  products to their drivers, we begin with an analysis of the relative importance of the 668 thermal and nonthermal drivers of  $pCO_2$  seasonal variability following the framework of 669 Fassbender et al. (2022) (see the Supplementary Materials for a description). For this 670 approach, in contrast to the widely applied deconvolution method of Takahashi et al. (1993, 671 2022), monthly  $pCO_2$  deviations in a given year are considered relative to the  $pCO_2$  value 672 calculated from annual mean values of SST, SSS, DIC, TA, PO<sub>4</sub> and SiO<sub>4</sub> rather than relative 673 to a direct mean of the monthly  $pCO_2$  values, which could obscure seasonally asymmetric 674  $pCO_2$  responses to its drivers.

675

Within this analysis we consider for each grid point the absolute value of the differences between the winter and summer seasonal averages over 2014-2018. The difference between the thermal and nonthermal  $pCO_2$  amplitudes is shown for the observational products (**Fig. 6a**) and for the GOBMs (**Fig. 6b**). For both panels, positive values (red) indicate that the thermal  $pCO_2$  component dominates the seasonal cycle amplitude and negative values (blue) indicate that the nonthermal component dominates for this climatological diagnostic.

682

683 We see consistency between the  $pCO_2$  products (Fig. 6a) and the GOBMs (Fig. 6b) in that 684 the thermal  $pCO_2$  component is dominant over the subtropics, with this dominance being even 685 stronger for the GOBMs. Over the northern subpolar gyres and the Southern Hemisphere 686 circumpolar regions there is a pronounced disagreement between the  $pCO_2$  products and the 687 GOBMs. Specifically, for the  $pCO_2$  products the nonthermal  $pCO_2$  component is dominant in 688 these biomes, while the GOBMs exhibit a relatively strong thermal component in these 689 regions. These discrepancies between the  $pCO_2$  products and GOBMs are also shown for each 690 GOBM and  $pCO_2$  product separately in Fig. 6c, revealing the relatively strong agreement 691 between the  $pCO_2$  products and the disparity between the GOBMs that was also in evidence 692 in Fig. 4 for  $pCO_2$  itself averaged over biome scales.

693

As we have identified in **Figs. 4** and **5**, there is a systematic discrepancy between GOBMs and  $pCO_2$  products, with GOBMs indicating a weaker climatological seasonal cycle in DIC variations, which is consistent with a more pronounced dominance of the thermal component of  $pCO_2$  seasonality highlighted in **Fig. 6**. Next, we evaluate whether the nonthermal  $pCO_2$ component discrepancy between GOBMs and  $pCO_2$  products can be explained by the smaller DIC seasonal variability in the GOBMs.

700

701 We consider the drivers of the climatological  $pCO_2$  seasonal cycle during 2014-2018 over the 702 six aggregated biomes in Fig. 7. The left column shows  $pCO_2$  climatologies for the  $pCO_2$ 703 products (blue) and the GOBMs (green). This serves as a reminder that over the NA-SPSS, 704 NP-SPSS and SH-SS domains, there are fundamental discrepancies between the GOBMs and 705 the  $pCO_2$  products in their representation of both the phase and amplitude of  $pCO_2$ 706 seasonality, while in NH-STSS, NH-STPS, and SH-STPS regions the amplitude of the 707 seasonal cycle of  $pCO_2$  is consistently larger in GOBMs. In order to understand the cause of 708 these discrepancies, a decomposition of the month-to-month changes in  $pCO_2$  into its main drivers (SST, SSS, DIC, and TA) is presented in the second and third column of Fig. 7. For 709 710 the  $pCO_2$  products (Fig. 7, second column) the SST term dominates over the DIC term in the 711 subtropics, while the DIC contribution is dominant in the subpolar biomes of the Northern 712 Hemisphere and high latitude regions of the Southern Hemisphere. For each of these six 713 biomes, we see that the GOBMs have a smaller DIC contribution (third column of Fig.7) 714 when compared to the  $pCO_2$  products (second column of Fig. 7).

715

It is worth noting for the observational  $pCO_2$  products (second column in **Fig. 7**) that although the effect of the spring bloom in NA-SPSS is marked by a short DIC-driven minimum in the  $pCO_2$  tendency in April, for NP-SPSS the DIC-driven  $pCO_2$  tendency is sustained from April through July (emphasis on the blue DIC tendency line in each case). For the GOBMs (third column in **Fig. 7**) there is no sustained dominance of the DIC contribution relative to SST over most of the seasonal cycle for NA-SPSS and NP-SPSS. For both of these biomes the GOBMs simulate weak phytoplankton blooms in late spring as indicated by the DIC tendency term for both cases, with the discrepancies in amplitude being larger than discrepancies in phase. This results in an SST dominance of the  $pCO_2$  tendency through the summer months, thereby driving  $pCO_2$  to peak 6-7 months earlier than that of the  $pCO_2$  products for each of the two biomes. This represents the impact of the weaker representation of the seasonal amplitude of surface DIC variations for the GOBMs.

728

729 In the fourth column of Fig. 7, the difference between the GOBM contributions (third 730 column) and the  $pCO_2$  product contributions (second column) allows us to identify DIC 731 discrepancies as the underlying term that leads to the principal different tendencies in  $pCO_2$ 732 seasonal amplitude over biome scales. In other words, over these aggregated scales the most 733 important driver that causes the disagreement between the  $pCO_2$  seasonal cycle of the 734 GOBMs and the  $pCO_2$  products lies in the processes that control DIC seasonality. It is 735 important to emphasize that the GOBMs provide a distinct advantage over coupled ESMs in 736 studying disagreements between modeled and observation-based pCO<sub>2</sub> seasonality, because 737 their climatological seasonal evolution of SST is effectively imposed from observations. This 738 allowed us to identify and understand the potential magnitude of  $pCO_2$  seasonality biases that 739 are imposed by a seeming systematic bias with GOBMs to underestimate the importance of 740 DIC seasonality. It is worth recalling here that the DIC from the  $pCO_2/TA$  products is more 741 consistent with the independent products (MOBO-DIC and NNGv2LDEO) than with the 742 GOBMs (Fig. 5d), lending credence to the discrepancies highlighted in Fig. 7.

743

744 A candidate mechanism that could potentially account for why the GOBMs have smaller 745 seasonal surface DIC variability than the DIC products could be in the modeled seasonal 746 variability of biological drawdown of surface DIC. However, in addressing this question we 747 are faced with the caveat that seasonal variability in biological drawdown of surface DIC over 748 global scales is poorly constrained by observations. To shed light on this question, we 749 consider in Fig. 8 the relationship between climatological (2014-2018) surface salinity-750 normalized DIC (nDIC) variations and mixed layer depth (MLD) averaged over the six 751 biomes. The MLD product is derived from Argo observations, described above. Fig. 8 shows 752 that MLD variations averaged across the GOBMs are smaller throughout the seasonal cycle 753 than they are for the observational product. Additionally, during summer months when the MLD becomes minimum, strong nDIC reductions are more pronounced in the observations than in the GOBMs. This suggests that the surface summer DIC drawdown, likely associated with biological drawdown of DIC, is underestimated in GOBMs. This could reflect a spurious exhaustion of nutrients by early summer in GOBMs. Investigating these questions further is not possible with the tools available through RECCAP2. However, given their importance for  $pCO_2$ , it is strongly advised that process understanding of the potential shortcomings of models in this regard be further investigated.

761

## 762 **3.2.3.** Attribution for decadal increases in *p*CO<sub>2</sub> seasonality

763

764 Here we adopt the framework of Fassbender et al. (2022) to address both mechanistically and 765 quantitatively the drivers of enhanced  $pCO_2$  seasonal amplitude. Specifically, this method 766 allows one to deconvolve the relative importance of these drivers, namely the transient 767 invasion flux of anthropogenic carbon (denoted as the Cant contribution) and other drivers 768 (denoted collectively as climate contribution). As the attribution methodology of Fassbender 769 et al. (2022) requires not only surface  $pCO_2$  but also surface DIC and TA concentrations (in 770 addition to SST, SSS, PO<sub>4</sub>, and SiO<sub>4</sub>), we focus here on the three RECCAP2  $pCO_2$  products 771 that include the necessary quantities, specifically JMAMLR, OceanSODA-ETHZ, and 772 CMEMS-LSCE-FFNN, as well as the GOBMs listed in Table 3. For the observation-based 773 products, monthly climatological PO<sub>4</sub> and SiO<sub>4</sub> fields were taken from World Ocean Atlas 774 2018 (Garcia et al., 2019).

775

776 We begin the analysis for this section on decadal changes in  $pCO_2$  and  $CO_2$  flux seasonality 777 with a descriptive assessment of seasonal asymmetries in the changes in seasonal amplitude 778 between 1985-1989 and 2014-2018 in Fig. 9.  $pCO_2$  anomalies are shown relative to the 779 annual mean value computed from annual means of SST, SSS, DIC, TA, and nutrients ( $pCO_2$ ) 780 AM) for both observationally-based products and GOBMS, following the method described in 781 Fassbender et al. (2022). Calculations are performed separately for each year and then 782 averaged over the time intervals 1985-1889 and 2014-2018. This provides an estimate of the 783 annual mean if  $pCO_2$  were to respond linearly to its individual drivers over the seasonal cycle,

making it possible to identify seasonally asymmetric changes in  $pCO_2$  that are obscured when

computing a mean of monthly  $pCO_2$  values over an annual cycle.

786

787 We first consider the case of the subtropics as approximately 15°N-40°N and 15°S-40°S, where summer anomalies of  $pCO_2$  for 1985-1989 (Fig. 9a) and 2014-2018 (Fig. 9b) are 788 789 larger for the GOBMs than for the  $pCO_2$  products, reflecting discrepancies in the amplitude of 790 DIC seasonality between the GOBMs and observation-based products discussed previously 791 (Fig. 5d). Importantly, between  $25^{\circ}$ N-40°N and  $25^{\circ}$ S-40°S the summer anomalies are larger 792 than the winter anomalies over both time intervals. For the difference between the 2014-2018 793 and 1985-1989 intervals (Fig. 9c), we see that over the Northern Hemisphere subtropical 794 latitudes the summer changes are larger than the winter changes. In other words, notable 795 asymmetry can be identified over the Northern Hemisphere subtropics with larger increases in 796 summer highs than decreases in winter lows. North of 45°N there are relatively symmetric 797 changes in summer and winter for the observation-based products and the GOBMs, but the 798 changes for the two product classes are of reverse sign (see next paragraph) and larger for the 799  $pCO_2$  products. The changes for the Southern Hemisphere are less pronounced, with some 800 degree of asymmetry over 10°S-25°S where for the observation-based products the winter 801 lows decrease more than the summer highs increase. There is no discernable asymmetry over 802 the Southern Ocean.

803

804 The disagreement between the GOBMs and  $pCO_2$  products, both for seasonal  $pCO_2$ 805 anomalies (Fig. 9a,b) and their changes (Fig. 9c), is largest in the northern subpolar latitudes 806 (45°N-60°N). For both the 1985-1989 and 2014-2018 climatologies, the  $pCO_2$  products show 807 relatively strong positive  $pCO_2$  anomalies in winter, and relatively strong negative  $pCO_2$ 808 anomalies in summer, reflecting the interplay between winter convective mixing and warm-809 season export production in this region where nonthermal  $pCO_2$  effects are dominant. In 810 contrast, over the same region the anomalies for  $pCO_2$  in GOBMs are relatively small in both 811 winter and summer, reflecting the near balance between thermal and nonthermal  $pCO_2$  cycles. 812 Although it is not explicitly shown here, it is important to keep in mind that Fig. 2 and Fig. 4 813 revealed that there are discrepancies in the phenology or phasing of seasonal variations 814 between the GOBMs and  $pCO_2$  products, such that the results with the GOBMs are sensitive 815 to the specific definition of the seasonal cycle amplitude with  $pCO_2$  products showing indeed 816 the largest seasonal change in winter and summer for 45°N-60°N, while GOBMs show the 817 largest change in early spring and late summer/early autumn (Fig. 2 and Fig. 4). For the 818 Southern Hemisphere region between 45°S and 60°S, the GOBMs and  $pCO_2$  products are 819 much more similar than they are over the Northern Hemisphere subpolar regions over both 820 1985-1989 and 2014-2018, with relatively modest decadal changes for the northern subpolar 821 regions where nonthermal  $pCO_2$  drivers are larger than thermal  $pCO_2$  drivers for the  $pCO_2$ 822 products.

823

824 The corresponding asymmetries in zonally-integrated seasonal CO<sub>2</sub> fluxes, shown in Fig. 9d-825 **9f**, are quite different from what was seen for surface  $pCO_2$  anomalies. For both time periods 826 1985-1989 and 2014-2018, seasonal CO<sub>2</sub> fluxes are larger over the subtropics in winter 827 (ingassing) than in summer (outgassing), which indicates that seasonal wind speed 828 asymmetries more than compensate the summer-skewed  $pCO_2$  seasonal asymmetry over the 829 same latitude range (Fassbender et al., 2022). This is consistent with the winter CO<sub>2</sub> fluxes 830 showing local extrema for ingassing near 40°N and 40°S where westerly winds exhibit 831 meridional maxima. This feature is also found in the 1985-1989 to 2014-2018 decadal 832 change. For the Northern Hemisphere subpolar regions spanning 45°N-60°N, the seasonal 833 amplification in CO<sub>2</sub> fluxes between 1985-1989 and 2014-2018 shows enhanced ingassing 834 during summer for the  $pCO_2$  products, whereas the GOBMs show enhanced winter uptake 835 over these latitudes. For the circumpolar regions of the Southern Hemisphere spanning 45°S-836  $60^{\circ}$ S, both the GOBMs and the pCO<sub>2</sub> products broadly exhibit enhanced ingassing in summer 837 (JFM) and winter (JAS) seasons relative to the 1985-1989 period.

838

We next turn our attention in **Fig. 10** to deconvolve the role of  $C_{ant}$  invasion and climate perturbations (e.g., changes to the ocean physical or biogeochemical state caused by anthropogenic climate change) in driving multi-decadal changes in  $pCO_2$  seasonality between 1985-1989 and 2014-2018. The changes in the subtropics are qualitatively similar in the  $pCO_2$  products and GOBMs (top and model rows of **Fig. 10**), with the changes in the total seasonal cycle amplitude (**Fig. 10a**) dominated by the  $C_{ant}$  invasion impact on the thermal component with a small climate-driven increase in the thermal  $pCO_2$  component (**Fig. 10b**).

However, for the subpolar regions of the Northern Hemisphere and the Southern Ocean, the  $pCO_2$  products show that the nonthermal  $pCO_2$  component changes driven by climate perturbations (**Fig. 10c**) are sufficiently large to dominate the total response in these regions (**Fig. 10a**), a feature that is not seen (or considerably weaker) for the GOBMs. C<sub>ant</sub>-induced declines in carbonate buffering in the high latitudes also work to slightly amplify the total nonthermal  $pCO_2$  seasonal cycle changes in both the  $pCO_2$  products and GOBMs (red lines in right column).

853

854 For the total change in the GOBM  $pCO_2$  values (Fig. 10d), the impact of climate 855 perturbations is relatively minor so that the change in  $pCO_2$  seasonality is largely dominated 856 by the invasion flux of  $C_{ant}$ . For the thermal component of  $pCO_2$  changes in the GOBMs, both 857 the subtropical and subpolar latitudes of the Northern Hemisphere indicate dominance of the 858 effect of transient Cant invasion, with the Southern Ocean posing one region where this is 859 neutralized by climate perturbations. For the nonthermal component of  $pCO_2$  changes in the 860 GOBMs, the impact of C<sub>ant</sub> invasion is relatively minor, so that the changes are dominated by 861 climate perturbations, which are also small. In summary, for the GOBMs, increases in the 862 total  $pCO_2$  seasonal cycle amplitude are dominated by the direct influence of  $C_{ant}$  on the thermal pCO<sub>2</sub> component seasonal cycle amplitude through the enhanced temperature 863 864 sensitivity associated with elevated ocean  $pCO_2$  values.

865

## 866 3.3. Estimating Irreducible Uncertainty in Seasonal Cycle Amplitude Changes

867

868 We now turn our attention to the emergence of decadal changes in  $pCO_2$  seasonality, with 869 applications at the gridoint, zonal mean, and biome scales.

870

3.3.1 Natural variability uncertainty in detecting forced changes in pCO<sub>2</sub> seasonal
amplitude

873

874 As was emphasized in the study of Landschützer et al. (2018), natural variability in the 875 seasonal amplitude of  $pCO_2$  variations can obscure detection of anthropogenic trends, an 876 effect that can be termed natural variability uncertainty. For sea surface  $pCO_2$ , the signal (Fig. 877 11a) and the signal-to-noise ratio (SNR) (Fig. 11b) are calculated for a single LE, to illustrate 878 patterns of emergence. We choose here the CESM2 model (Danabasoglu et al., 2020) for 879 which the phasing of the  $pCO_2$  seasonal cycle over the Northern Hemisphere subpolar biomes 880 has the best correspondence with the observation-based products out of the LE simulations 881 described in Table 4 or the CMIP6 models shown in Table. 6.

882

883 According to the CESM2-LE, the  $pCO_2$  seasonality increases are most emergent (|SNR|>1) 884 (Fig. 11b) at the grid point level over much of the subtropical Northern Hemisphere, as well 885 as over a band spanning the subtropical convergence regions of the Southern Hemisphere. 886 Large parts of the North Atlantic subpolar gyre exhibit a decrease in the amplitude of seasonal 887  $pCO_2$  variations. This analysis is indicative of  $pCO_2$  seasonality changes over the Southern 888 Ocean being non-emergent above the noise level of natural variability at the grid point level 889 over our three-decade interval. In particular,  $pCO_2$  seasonality changes in the Antarctic 890 Circumpolar Current region spanning 50°S-60°S are non-emergent, along with some parts of 891 the Southern Hemisphere subtropics.

892

893 We next consider the emergence characteristics for the underlying thermal and nonthermal 894 drivers of  $pCO_2$  seasonality as represented by the large ensemble model. We see that the 895 forced decadal change (ensemble mean) for the thermal component (Fig. 11c) and nonthermal 896 component (Fig. 11e) are both considerably larger than the decadal change for the full  $pCO_2$ 897 field itself (Fig. 11a). For the resultant signal-to-noise ratio, we see that the patterns for the 898 thermal component (Fig. 11d) and the nonthermal component (Fig. 11f) are spatially offset 899 from each other, and when considered together help to account for the relatively restricted 900 regions over which there is local grid point emergence, predominantly over the thermally 901 dominated subtropical domain, for the  $pCO_2$  seasonal cycle. From these analyses, we can see 902 that within the subtropics, the emergent regions for total  $pCO_2$  SNR (|SNR|>1 in Fig. 11a) 903 correspond to the regions where the ensemble mean nonthermal component (Fig. 11e) is 904 small.

905

#### 906 **3.3.2 Interpretation of results of attribution analysis**

907

908 Having visualized the patterns of emergence for  $pCO_2$  seasonality changes in Fig, 11 we 909 redirect out attention to the last row of Fig. 10 (panels 10g-10i), where the same set of 50 910 large ensemble members with the CESM2-LE are applied to interpret the zonal mean changes 911 in the  $pCO_2$  products (first row of Fig. 10). The shading for the LE results represents natural 912 variability uncertainty (one standard deviation about the 50-ensemble-member mean), while 913 the shading for the  $pCO_2$  products (Figs. 10a-10c) can be interpreted to represent mapping 914 uncertainty. These forms of uncertainty are distinct, but can be used together to assess the 915 degree to which the  $C_{ant}$  and climate-driven signals for the  $pCO_2$  products are detectable. We 916 consider the means of the  $pCO_2$  products in Figs. 10a-10c to be "best estimates" of the 917 decadal changes. Superimposing the envelope of natural variability from the LE analysis 918 (Figs. 10g-10i) onto the  $pCO_2$  product means would suggest that the  $C_{ant}$  and climate driven 919 signals are emergent when they and their associated natural variability are separated from 920 zero for their decadal change.

921

922 Under this framework, we interpret the  $C_{ant}$  component of the  $pCO_2$  product changes to be 923 detectable for all regions and  $pCO_2$  components ( $pCO_2$ ,  $pCO_2$ <sub>T</sub>, and  $pCO_2$ <sub>NT</sub>). What is more 924 surprising and compelling with the  $pCO_2$  products is the fact that the climate driven changes 925 over the Northern subpolar domains, with much of this signal found in the nonthermal 926 component, cannot be explained by natural variability according to our CESM2-LE results. 927 Further attribution of this climate-driven signal (mixing versus biology, for example) is 928 beyond the scope of this study, but at the very least this underscores the importance of 929 sustained seasonally-resolving observations in high latitude regions to better constrain and 930 understand this behavior.

931

Taken together, the attribution analysis in Fig. 10 and the analyses of  $pCO_2$  and  $CO_2$  flux seasonal asymmetries in Fig. 9 indicate that, over the broad areal extent of the subtropics, the  $pCO_2$  seasonal amplitude increases with an asymmetric larger increase in summer are in large 935 part due to the C<sub>ant</sub> impact on the thermal driver of seasonal amplitude. Through the effect of 936 stronger wind speeds in winter than summer, the seasonal  $CO_2$  flux asymmetry becomes even 937 larger and of opposite sign over the subtropics, with large increases in winter ingassing. Over 938 the broad expanse of the subtropics, the agreement between the  $pCO_2$  products and GOBMs, 939 together with the clear emergence indicated by the LE, gives us reasonable confidence for 940 both the driver ( $C_{ant}$ ) and the asymmetry of  $pCO_2$  seasonality changes and elevated winter 941  $CO_2$  uptake. However, for the high northern latitudes,  $pCO_2$  products and GOBMs disagree 942 on the drivers of the changing  $pCO_2$  seasonality. Here, the products indicate that a rather large 943 climate forcing is responsible for  $pCO_2$  seasonal cycle changes.

944

## 945 **3.3.3 Emergence of forced trends with biome aggregation**

946

947 We next address the degree to which biome aggregation improves the emergence of  $pCO_2$ 948 seasonality trends between 1985-1989 and 2014-2018. To this aim, we use multiple LE 949 simulations with five different ESMs (detailed in **Table 4**), and apply them to interpret the 950 decadal changes in  $pCO_2$  seasonal variations represented by the  $pCO_2$  products. However, 951 before considering multi-decadal trends we consider first in Fig. 12a an analysis for the six 952 aggregated biomes of the simulated seasonal amplitude of  $pCO_2$  variations for a climatology 953 over 2014-2018, both for pCO<sub>2</sub> products and LEs, with uncertainty ranges indicated for each 954 case. For the observation-based  $pCO_2$  products, the uncertainty shown represents the 955 uncertainty across the nine  $pCO_2$  products themselves, which is reflecting the spread across 956 methods (mapping uncertainty) in representing gridded  $pCO_2$ . For an individual LE, the 957 uncertainty represents the range of natural variability represented by the collection of 958 individual ensemble members for that specific LE. We see that for all six aggregated biomes, 959 at most, two of the five LEs have a climatological seasonal amplitude (defined as the absolute 960 value of JFM-JAS) where the first quartile range (25%-75%, solid bar) overlaps with the first-961 to-third quartile range of the  $pCO_2$  products. Over four of the six biomes (NP-SPSS, NH-962 STPS, SH-STPS, and SH-SS) CESM2-LE overlaps with the pCO<sub>2</sub> products, thereby 963 indicating correspondence. We also see that inter-model differences in seasonal amplitude 964 (ensemble median) are much larger than the ensemble spread (natural variability) for the 965 individual ESMs. Overall, the analysis in Fig. 12a indicates that the representation of the 966 climatological seasonal amplitude of  $pCO_2$  in LEs is partly inconsistent with the  $pCO_2$ 967 products, mirroring what was found for GOBMs in the first part of the **Results** section of this 968 study.

969

970 On the other hand, the decadal changes in the amplitude of seasonal  $pCO_2$  variations between 971 1985-1989 and 2014-2018 (Fig. 12b) reveal a much better agreement (relative to Fig. 12a) 972 between the median of the observational products and the ensemble spread of the individual 973 LEs. Most importantly, with one exception over the three subtropical biomes (NH-STSS, NH-974 STPS, SH-STPS) and for each LE within these regions, the range of outcomes for modeled 975  $pCO_2$  seasonality changes (the range indicated by the whiskers) does not intersect with a zero 976 trend or go negative. Thus in the subtropics the biome-aggregated changes in the seasonal 977 amplitude of  $pCO_2$  are predominantly emergent, with SH-SS also being largely emergent. For 978 the NP-SPSS biome, the median decadal change calculated for each LE indicates an increase 979 in  $pCO_2$  seasonal amplitude, although one of the LEs is not emergent while two other LEs are 980 near the one standard deviation threshold for emergence. This is indicative of the NP-SPSS 981 being moderately emergent under aggregation.

982

983 For the NA-SPSS biome the decadal changes in  $pCO_2$  seasonal amplitude are quite distinct 984 from the other regions, in that all LEs are at least moderately emergent, but that they have 985 divergent signs in their simulated decadal changes. Three of the LEs (CESM1, CESM2, and 986 ESM2M) exhibit a decrease in  $pCO_2$  seasonal amplitude, while two of the LEs (CanESM2 987 and CanESM5) exhibit an increase. Under the caveat that we have imposed a somewhat 988 restrictive definition of the seasonal cycle that is optimally suited for the  $pCO_2$  products (see 989 Fig. 2 and Fig. 4), the at least moderately emergent decrease in  $pCO_2$  seasonal amplitude for 990 three LEs is distinctive for the NA-SPSS biomes. The study of Goris et al. (2022) suggests 991 that such behavior may be expected from the disparate responses of the Atlantic Meridional 992 Overturning Circulation (AMOC) for the LEs. However, further exploration of this point 993 should be done while taking into consideration the sensitivity of the models to the definition 994 of the seasonal cycle. It is interesting that one of the  $pCO_2$  products does show zero net 995 change between 1985-1989 and 2014-2018.

997 The fact that the distribution of multi-decadal trends for individual LEs overlaps with the 998 25%-75% (bar) range for the observation-based  $pCO_2$  products gives us a degree of 999 confidence that the biome-aggregated changes can be considered emergent or detectable over 1000 this 30-year period. It should be noted, however, that the Northern Hemisphere biomes tend to 1001 be faced with a triple-whammy of elevated uncertainty, given the relatively high degree of 1002 disaccord between  $pCO_2$  products (mapping uncertainty), the relatively elevated level of 1003 natural variability uncertainty, and the offset of the medians of the different LEs (model 1004 structural uncertainty). The LEs nevertheless bring great value to the parsing of uncertainty in 1005 that they allow us to distinguish between internal variability uncertainty and model structural 1006 uncertainty. Qualitatively they are of similar amplitude for the decadal changes (Fig. 12b), 1007 but the model structural uncertainty is much larger for the 2014-2018 climatology (Fig. 12a).

1008 The Southern Ocean (SH-SS) region is characterized by relatively weak trends (Fig. 12b), yet 1009 due its relatively weak natural variability is also predominantly emergent at the biome scale 1010 despite being non-emergent at the gridpoint scale (Fig. 11b). The reason for there being both 1011 weak natural variability and a weak decadal trend over the Southern Ocean is due to the 1012 combined effects of the tendency of models to have a relatively weak dominance of thermal 1013 over nonthermal seasonality drivers over the Southern Ocean, and that the relatively small 1014 amplitude of seasonal SST variations over the Southern Ocean. Taken together, this limits 1015 the ability of the invasion flux of  $C_{ant}$  to boost  $pCO_2$  seasonality amplitude between the 1980s 1016 and 2010s.

1017

## 1018 4 **Discussion**

1019

1020 The unprecedented collection of  $pCO_2$  products and model simulations available through the 1021 RECCAP2 project has provided an opportunity for not only synthesizing but also 1022 understanding the changing seasonal cycle in sea surface  $pCO_2$ . The use of new observation-1023 based  $pCO_2/TA$  products that include DIC and TA in addition to  $pCO_2$  has made it possible to 1024 address the following scientific questions: (i) what are the underlying causes of discrepancies 1025 in  $pCO_2$  seasonal variability between GOBMs and observation-based products? (ii) what are 1026 the drivers of decadal-timescales trends towards increasing  $pCO_2$  seasonality?, and (iii) are 1027 there asymmetries in the growth of  $pCO_2$  and  $CO_2$  flux seasonal variations over multi-decadal 1028 timescales that could give rise to a feedback mechanism consistent with the mechanism 1029 identified in the modeling study of Fassbender et al. (2022)?

1030

1031 One of the main findings of this study is that there are systematic discrepancies in the 1032 climatological seasonal amplitude of surface DIC variations between GOBMs and 1033 observation-based  $pCO_2/TA$  products over very large spatial scales (Fig. 5c) that are 1034 primarily responsible for compromising the climatological seasonal cycle of  $pCO_2$ . More 1035 specifically, simulated DIC seasonal variability is systematically smaller for GOBMs than for 1036 DIC seasonal variability in the  $pCO_2/TA$  products. The climatological seasonal amplitude of 1037 DIC variations can be impacted by both the seasonal amplitude of net biological removal of 1038 DIC from the surface layer and the seasonal process of entrainment associated with 1039 destratification. Although it is beyond the scope of this study to parse these processes, 1040 important information can be inferred from previous studies. Models have previously been 1041 shown to prematurely exhaust limiting nutrients in summer (Goris et al., 2018), and such 1042 behavior could impact the seasonal amplitude of surface DIC variations

1043

1044 From the biome-averaged time series shown in Fig. 4, we have seen that there are some important discrepancies in phasing of the climatological seasonal cycle for  $pCO_2$  between 1045 1046 GOBMs and  $pCO_2$  products over biome scales. We have opted here to focus largely on a 1047 definition of seasonal cycle amplitude as the difference between winter and summer, defined 1048 as JFM-JAS (JAS-JFM) for the Northern (Southern) Hemisphere, as this was deemed to 1049 capture the main characteristics of  $pCO_2$  seasonality over large scales, based on the  $pCO_2$ 1050 products, as shown in the Hovmöller diagrams in Fig. 2. Although our main story of 1051 seasonality changes in  $pCO_2$  is not strongly sensitive to this definition for the  $pCO_2$  products 1052 or the GOBMs (Fig. 3), the presence of phasing differences between  $pCO_2$  products and 1053 GOBMs in high latitudes influences the seasonal amplitude calculated according to this 1054 definition. We recommend further investigation of such phasing differences to better 1055 understand the underlying reasons for this discrepancy.
We emphasized earlier that for our purposes of a synthesis, the GOBMs potentially offer an important advantage over coupled ESMs for our seasonality study and associated attribution analysis. This is due to the fact that the SSTs of the GOBMs are constrained to correspond

analysis. This is due to the fact that the SSTs of the GOBMs are constrained to correspond 1060 closely to observations through the imposition of a negative feedback in surface heat fluxes 1061 by virtue of using observed surface air temperatures (Large and Yeager, 2009). However, 1062 there is no analogous data-constraint applied for the DIC seasonal evolution, other than an 1063 indirect and much weaker effective restoring of sea surface  $pCO_2$  to atmospheric  $pCO_2$  over a 1064 one-year timescale (an order of magnitude less than for SST). As the SSTs of the GOBMs 1065 effectively track the observed seasonal cycle with fidelity, our attention in this study was 1066 drawn to the dominant role of DIC seasonality discrepancies in distorting  $pCO_2$  seasonality 1067 for the GOBMs relative to the observation-based products.

1068

1069 Nevertheless, as a complementary exercise to the evaluation of GOBMs in Fig. 4, we 1070 consider here an analogous analysis of a collection of the 11 CMIP6 ESMs listed in Table 6. 1071 The ESMs are evaluated for their climatological seasonal cycle in  $pCO_2$ , SST, and DIC, as 1072 well as for decadal changes in  $pCO_2$ . For this case, we have considered changes between 1073 1985-1989 and 2010-2014 (an interval four years shorter than in Fig. 4) to be consistent with 1074 the fact that the historical period in CMIP6 ends in 2014. In general terms there is no 1075 fundamental difference between both classes of models. Despite the absence of observational 1076 constraints, the simulated SST seasonal cycle in ESMs is in good agreement with 1077 observations (dotted lines in central column of Fig. 13), although the model spread is 1078 significantly larger as expected. The DIC seasonality of the ESMs is very similar to that of the 1079 GOBMs (weaker in models than in observation-based  $pCO_2$  products) and the overall  $pCO_2$ 1080 changes are very consistent between fully-coupled ESMs and GOBMs (right column of Fig. 4 1081 and Fig. 13). This serves to further underscore our main finding that there is a pervasive 1082 discrepancy between ocean carbon cycle models and  $pCO_2$  products, not limited to the 1083 RECCAP2 GOBMs, with a seasonal variability in models that is too weak relative to  $pCO_2$ 1084 products. Interpretation of projected future seasonal changes in ESMs should therefore take 1085 into account the systematic model uncertainties highlighted in this study. As we have seen in 1086 Fig. 12, a future extension of this analysis for coupled models could benefit from the inclusion of LEs, which offer a means to deconvolve for models the effects of structuraluncertainty from natural variability uncertainty.

1089

Our attribution analysis indicates that in the subtropics the GOBMs and  $pCO_2$  products are 1090 1091 qualitatively consistent in both the degree of seasonal asymmetry in decadal  $pCO_2$  changes 1092 and  $CO_2$  flux changes (Fig. 9), as well as for the mechanistic drivers (Fig. 10). This reflects 1093 the fact that both the GOBMs and  $pCO_2$  products are dominated by the thermal  $pCO_2$ 1094 component in the subtropics, albeit with the degree of dominance of the thermal  $pCO_2$ 1095 component being exaggerated by the GOBMs. The fact that the Northern Hemisphere 1096 subtropics have a larger response than the Southern Hemisphere subtropics is expected given 1097 that the seasonal cycle in SST is larger in the north than in the south, and thereby the thermal 1098 component of seasonality to be larger in the north than in the south. Thus, C<sub>ant</sub> invasion will 1099 preferentially boost the amplitude of  $pCO_2$  seasonality in the subtropics of the Northern 1100 Hemisphere relative to the Southern Hemisphere, due to the higher amplitude SST variability 1101 to the north.

1102

1103 An important implication of our attribution analysis is that the decadal changes in seasonal 1104  $pCO_2$  and  $CO_2$  flux amplitudes are qualitatively consistent with the existence of a negative 1105 climate feedback over the historical period spanning 1985-2018. Consistent with what was 1106 found by Fassbender et al. (2022) using future projections with an LE simulation, the seasonal 1107 asymmetry in CO<sub>2</sub> fluxes identified here for the historical period is by inference in large part 1108 due to seasonal asymmetries in wind speeds and thereby piston velocities, rather than in  $pCO_2$ 1109 itself. It is our hope that the analysis provided here will motivate future work to quantify the 1110 strength of this feedback over the historical period, as doing so was beyond the scope of this 1111 study.

1112

1113 Two studies that evaluated the early generation of prognostic ocean biogeochemistry models 1114 for their representation of  $pCO_2$  seasonality against observations were McKinley et al. (2006) 1115 for the case of the North Pacific and Tjiputra et al. (2012) for the case of the North Atlantic. 1116 Although an explicit comparison of the models applied by McKinley et al. (2006) and 1117 Tjiputra et al. (2012) with the RECCAP2 GOBMs is beyond the scope of this study, it is 1118 important to ask, "how far have we come?". The message from this study is that the new suite 1119 of  $pCO_2/TA$  products that provide time-varying surface DIC products that have become 1120 available provide invaluable tools for identifying and attributing potential model biases over 1121 global scales. Moving forward, we recommend that efforts be devoted to understanding how 1122 complexity in physical and biogeochemical process representation in models is reflected in 1123  $pCO_2$  seasonality. For the case of the biogeochemical components of models, one opportunity 1124 would be to evaluate with the same circulation state a climatology of  $pCO_2$  seasonality using 1125 different complexity of biogeochemical models, analogous to what was explored by Galbraith 1126 et al. (2015) with the BLING and TOPAZ biogeochemical models, or by Karakus et al. 1127 (2022). Moreover, it could be beneficial to link the simulated strength of key circulation 1128 metrics (e.g. Atlantic Meridional Overturning Circulation for the North Atlantic) to the  $pCO_2$ 1129 seasonality of the GOBMs. In a study with several coupled climate models, Goris et al. 1130 (2022) were able to show that the representation of a model's  $pCO_2$  seasonality in the North 1131 Atlantic is strongly linked to the simulated strength of the Gulf Stream volume transport. In 1132 order to address the question of how missing physical processes may impact biogeochemistry 1133 and  $pCO_2$  seasonality, it would be beneficial to explore independently the impact of 1134 mesoscale turbulence with ocean eddies (Lévy et al., 1998) and shear-induced turbulence 1135 associated with near-inertial oscillations, ocean swells, and wind stirring (Jochum et al., 2013; 1136 Keerthi et al., 2021; Rodgers et al., 2014) through perturbation studies with individual 1137 models. Such efforts would be beneficial in identifying the processes that contribute to the 1138 discrepancies identified in this RECCAP2 synthesis effort.

1139

### 1140 **5** Conclusions

1141

1142 In this study, we have presented as a contribution to the RECCAP2 effort a synthesis of how 1143 the seasonal cycle in sea surface  $pCO_2$  and air-sea  $CO_2$  fluxes has changed over the last three 1144 decades (1985-2018). Our main findings include:

1146 For a 2014-2018 climatology, we identified a systematic discrepancy in the seasonal 1147 amplitude of surface DIC variations between the GOBMs, which are too weak, and the 1148 surface DIC products, such that the  $pCO_2$  seasonal cycle in subtropical biomes is spuriously 1149 large, and both the amplitude and phase of seasonal  $pCO_2$  variations are spuriously modulated 1150 in subpolar and Southern Ocean biomes. Similar biases are found in ESMs as submitted to 1151 CMIP6. When averaged over aggregated biomes, both  $pCO_2$  products and GOBMs represent 1152 consistent increases in the seasonal amplitude of pCO<sub>2</sub> and CO<sub>2</sub> fluxes between 1985-1989 1153 and 2014-2018.

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1155 Decadal increases in  $pCO_2$  seasonal amplitude in subtropical biomes are shown to be 1156 dominated process-wise by the invasion flux of anthropogenic carbon ( $C_{ant}$ ) for both the  $pCO_2$ 1157 products and GOBMs. For pCO<sub>2</sub> seasonality changes over subpolar and circumpolar biomes, GOBMs see this change as being dominated by C<sub>ant</sub> invasion, but for pCO<sub>2</sub> products it is 1158 1159 dominated rather by modulations of the climate state, which can be associated with changes in 1160 SST seasonality or in perturbations occurring in mixing or biological processes. Our analysis 1161 of LE simulations indicates that the climate-driven signal found in the subpolar biomes of the 1162 Northern Hemisphere (bold blue line in **Fig. 10a**) is likely too large to be explained by natural 1163 variability, as estimated from the amplitude of natural variability in the LE (width of blue 1164 shading in Fig. 10g). Considered together, the subtropical biomes exhibit decadal increases in 1165 CO<sub>2</sub> flux seasonality that are larger during winter than summer, consistent with the 1166 mechanism identified by Fassbender et al. (2022) whereby increased seasonal cycles may be 1167 promoting a negative feedback in the climate system.

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1169 We have also identified and quantified two distinct sources of uncertainty that can impact the 1170 detection of anthropogenic trends in  $pCO_2$  seasonal amplitude. The first type is mapping 1171 uncertainty (Fig. 3a and Fig. 3c), which is in essence a reducible form of uncertainty. The 1172 second type is natural variability uncertainty (Fig. 11 and Fig. 12), which we identified with 1173 the LE simulations, with this being an irreducible form of uncertainty. For both cases, we 1174 identified that there are large regions of the global domain where the forced anthropogenic 1175 signal does not rise above the one standard deviation (noise) level at the gridpoint scale. Even 1176 for the idealized case of a perfectly-resolving observational system where mapping error 1177 disappears, one would still need to quantify the degree of confidence one has that a local 1178 measured decadal change represents forced changes in the presence of natural variability. We 1179 tend in the subtropics to find unanimous agreement at the gridpoint scale across  $pCO_2$ 1180 products and GOBMs on the dominant mechanism of enhanced  $pCO_2$  seasonality, namely the 1181 C<sub>ant</sub>-driven boost to the thermally component of  $pCO_2$  seasonality.

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1183 Thus our synthesis provides evidence that the seasonal  $pCO_2$  amplitude has significantly 1184 increased globally, and for many regions, this increase has already emerged from the noise 1185 with biome aggregation. This highlights the importance of including seasonal changes when 1186 considering long-term acidification trends, as we will likely cross critical thresholds in some 1187 seasons, before such thresholds are crossed in the annual mean (Burger et al., 2020; Henson et 1188 al., 2017; Kwiatkowski and Orr, 2018; McNeil and Sasse, 2016). For similar reasons, we 1189 recommend that modeling resources be devoted to identifying missing processes and 1190 implementing representations or parameterizations of the pertinent missing processes in ocean 1191 biogeochemical models.

1192

1193 Our conclusions rest on the growing availability of  $pCO_2$  products that include associated 1194 time-varying DIC and TA products, with this representing a significant advance for scientific 1195 inquiry into the mechanisms controlling the ocean uptake of anthropogenic carbon. The fact 1196 that the three  $pCO_2/TA$  products (considered here as surface DIC products) that exist to date 1197 (JMAMLR, OceanSODA-ETHZ, and CMEMS-LSCE-FFNN) indicate consistent patterns of 1198 seasonal variability over large scales, which are also consistent with depth-resolving three-1199 dimensional monthly DIC climatologies (MOBO-DIC and NNGv2LDEO) over large scales, 1200 should boost community confidence in the scientific utility of these products. We therefore 1201 strongly recommend to the  $pCO_2$  product providers to provide DIC and TA along with sea 1202 surface  $pCO_2$  and air-sea  $CO_2$  fluxes. In parallel, we recommend a rigorous skill-assessment 1203 of the mapping methods that transform from SOCAT measurements to globally-gridded  $pCO_2$ 1204 products, for example by using LEs with ESMs as a synthetic testbed similar to the work 1205 presented by Gloege et al. (2021).

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## **OPEN RESEARCH**

1245	All of the ]	RECCAP2	data will l	be made	available in a	a public re	positor	v before	publication.
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#### 1247 CONTRIBUTOR ROLES TAXONOMY

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1249 Conceptualization (Ideas; formulation or evaluation of overarching goals/aims):

- 1250 KBR, JS, AJF, RY
- 1251 Data Curation (Management activities to annotate (produce metadata)):
- 1252 LGr, YI, TTTC, JDM, LK, PV, JS, PL, AV, JH, NGo, JT

1253 Formal analysis (application of statistical, mathematical, computational, or other formal

- 1254 techniques to analyze or synthesize study data):
- 1255 JS, AJF, RY, HF, KS, SSc, KBR
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- 1258 KR
- 1259 Investigation (Conducting a research and investigation process, specifically performing the 1260 experiments, or data/evidence collection):
- 1261 JS, AJF, RY, JDM, NGo, JEK, HF, KS, PL, SSc, SSh, MI, KT, KBR
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  components):
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1276 N/A

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- 1282 KBR
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- 1284 by those form the original research group, specifically critical review, commentary, or
- 1285 revision including pre- or post-publication stages):
- 1286 Everyone
- 1287
- 1288
- 1289

Names	Aggregated seasonality biomes	RECCAP2 biomes
North Atlantic subpolar seasonally stratified	NA-SPSS	NA-SPSS
North Pacific subpolar seasonally stratified	NP-SPSS	NP-SPSS
Northern Hemisphere subtropical seasonally stratified	NH-STSS	NA-STSS+NP-STSS
Northern Hemisphere subtropical permanently stratified	NH-STPS	NA-STPS + NP-STPS
Southern Hemisphere subtropical permanently stratified	SH-STPS	SA-STPS + SP-STPS + Southern Indian Ocean
Southern Hemisphere seasonally stratified (subpolar and subtropical combined)	SH-SS	SO-STSS + SO-SPSS

**Table 1**: The six aggregated biomes used in this study. The full names are given in the left column, the abbreviated names used throughout this study in the central column, and the original RECCAP2 biomes from which they are constructed are listed in the right column. Our six aggregated biomes are shown in the central panel of Fig. 1. As has been noted in the Methods section, spatial and temporal data coverage of data products and models might further constrain the area of the aggregated biomes used for our analysis.

Product name	References	Duration and Remarks
CSIRML6	Gregor et al., 2019	1985-2018
CMEMS-LSCE-FFNN	Chau et al., 2022	1985-2018; DIC and TA available
JMAMLR	lida et al., 2021	1985-2019; DIC and TA available
Jena-MLS	Rödenbeck et al., 2013, 2022	1985-2018
NIES-ML3	Zeng et al., 2022	1980-2020
OceanSODA-ETHZ	Gregor and Gruber, 2021	1985-2018; DIC and TA available
SOMFFN	Landschützer et al., 2016	1982-2019
UOEX_WAT20	Watson et al., 2020	1985-2019
spco2_LDEO_HPD	Gloege et al., 2022	1985-2018

**Table 2**:  $pCO_2$  data products used for this study. The products are described more completely1301in the global RECCAP2 analysis of DeVries submitted to Global Biogeochemical Cycles.1302Note that the CMEMS-LSCE-FFNN, JMAMLR, and OceanSODA-ETHZ products include1303associated surface DIC and TA fields that are used in this study.

Model Name	References	Remarks
CCSM	Doney et al., 2009	Covers only 1985-2017, For 2014-2018 climatologies an exception is made for this model
CESM-ETHZ	Doney et al., 2009; Lindsay et al., 2014; Yang and Gruber, 2016	
CNRM-ESM2-1	Berthet et al., 2019; Séférian et al., 2016, 2019	
EC-Earth3	Döscher et al., 2022	
FESOM-REcoM-LR	Hauck et al., 2020	No phosphate available, WOA2018 used for attribution analysis
MOM6-COBALT- PRINCETON	Liao et al., 2020; Stock et al., 2020	
MRI-ESM2-1	Nakano et al., 2011; Urakawa et al., 2020	
NorESM-OC1.2	Schwinger et al., 2016	
ORCA025-GEOMAR	Chien et al., 2022; Kriest and Oschlies, 2015; Madec et al., 2017	No silicate available, silicate from WOA2018 used for attribution analysis
IPSL-NEMO-PISCES	Aumont et al., 2015; Friedlingstein et al., 2022	

PlankTOM12	(Le Quéré et al., 2010; Wright et al., 2021)	No silicate available, WOA2018 used for attribution analysis
Assimilation Model	References	Remarks
OCIM v2021	(DeVries, 2014, 2022)	Abiotic model with data assimilated circulation; Used for some analyses but never included in ensemble means for GOBMs

1309 **Table 3**: Global Ocean Biogeochemical Models (GOBMs) and an assimilation model used

1310 for this study. Model names are given in the first column, references are given in the second

1311 column, and comments pertinent to the application of the products are given in the third

1312 column. The CCSM model differs from the others in that output only extends to 2017 rather

1313 than to the RECCAP2 protocol end-year of 2018.

Earth System Model	References	Comments
CESM2 (CMIP6)	Rodgers et al., 2021	50 members
CanESM2 (CMIP5)	Kirchmeier-Young et al., 2017	50 members
CanESM5 (CMIP6)	Swart et al., 2019b	24 members
CESM1 (CMIP5)	Kay et al., 2015	34 members
ESM2M (CMIP5)	Rodgers et al., 2015	30 members
MPI-GE (CMIP5)	Maher et al., 2019	100 members

**Table 4**: Large Ensemble Simulations used in the analysis in **Fig. 12**.

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	GOBMs		<i>p</i> CO <sub>2</sub> products	
Region	$pCO_2$ amplitude	<i>p</i> CO <sub>2</sub> amplitude change	<i>p</i> CO <sub>2</sub> amplitude	<i>p</i> CO <sub>2</sub> amplitude change
NA-SPSS	-18.5 +/- 18.5	-2.1 +/- 9.9	46.0 +/- 4.9	5.9 +/- 6.1
NP-SPSS	-34.1 +/- 29.3	-8.3 +/- 6.0	17.1 +/- 4.3	2.8 +/- 7.7
NH-STSS	-67.8 +/- 16.2	-10.7 +/- 3.2	-37.2 +/- 1.7	-9.8 +/- 3.9
NH-STPS	-42.0 +/- 6.7	-6.3 +/- 1.1	-33.2 +/- 2.4	-6.3 +/- 2.8
SH-STPS	-39.5 +/- 10.7	-2.7 +/- 2.0	-29.4 +/- 3.2	-5.3 +/- 2.7
SH-SS	-5.8 +/- 14.7	0.6 +/- 1.4	11.4 +/- 2.4	1.0 +/- 2.5

**Table 5:** Seasonal amplitude (winter-minus-summer) of biome averaged  $pCO_2$  for GOBMs1322and  $pCO_2$  products for the period 2014-2018. Seasons are defined as the three month periods

1323 JFM and JAS. Change represents the difference between 1985-1989 and 2014-2018,

Model Name	ESM reference	Data reference
ACCESS-ESM1-5	Law et al., 2017	Ziehn et al., 2019
CanESM5	Swart et al., 2019b	Swart et al., 2019a
CESM2	Lauritzen et al., 2018	Danabasoglu, 2019
CMCC-ESM2	Lovato et al., 2022	Lovato et al., 2021
CNRM-ESM2-1	Séférian et al., 2019	Séférian, 2018
GFDL-ESM4	Dunne et al., 2020	Krasting et al., 2018
IPSL-CM6A-LR	Boucher et al., 2020	Boucher et al., 2018
MIROC-ES2L	Hajima et al., 2020	Hajima et al., 2019
MRI-ESM2-0	Yukimoto et al., 2019b	Yukimoto et al., 2019a
NorESM2-MM	Seland et al., 2020	Bentsen et al., 2019
UKESM-0-LL	Sellar et al., 2019	Tang et al., 2019

**Table 6**: CMIP6 models evaluated against  $pCO_2$  products in **Fig. 13**.

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1732 Fig. 1: Central panel showing biomes used for this study (see Table 1 for details): North 1733 Atlantic subpolar seasonally stratified (NA-SPSS), North Pacific subpolar seasonally 1734 stratified (NP-SPSS), Northern Hemisphere subtropical seasonally stratified (NH-STSS), 1735 Northern Hemisphere subtropical permanently stratified (NH-STPS), Southern Hemisphere 1736 subtropical permanently stratified (SH-STPS), and Southern Hemisphere seasonally stratified, 1737 incorporating the subpolar and subtropical components (SH-SS). Surrounding panels show 1738 time series plots of biome-integrated sea-air CO<sub>2</sub> fluxes (annual maximum and minimum 1739 values irrespective of the month of occurrence).  $pCO_2$  products (blue) and GOBMs (green) 1740 are shown for both the ensemble mean (bold) and for one standard deviation (shaded). 1741 Positive (negative) values indicate outgassing (ingassing) of CO<sub>2</sub>. The regions in white in the 1742 central panel are not included in this analysis., in each panel, winter is designated by W, and 1743 summer by S, in order to distinguish the seasonal phasing between the subtropical and 1744 subpolar/Southern Ocean biomes.


1746 Fig. 2: Hovmöller diagrams of zonally-averaged climatological (2014-2018)  $pCO_2$  anomalies 1747 for  $pCO_2$  products (a) and GOBMs (e), and corresponding standard deviations (b, f); multidecadal change in  $pCO_2$  climatology between 1985-1989 and 2014-2018 for  $pCO_2$  products 1748 1749 (c) and GOBMs (g) and corresponding standard deviation (d, h). The seasonal maxima (black 1750 triangles) and minima (white triangles) are highlighted in panels a and e. Anomalies were 1751 calculated, grid-point by grid-point, by first fitting a quadratic polynomial through the full 1752 monthly time series over the period 1985-2018 and removing this trend from the data. Then, 1753 mean  $pCO_2$  values over 1985-1989 and 2014-2018 were calculated for each month as a 1754 deviation from the detrended mean  $pCO_2$  over the whole five year period.

#### Sea surface pCO<sub>2</sub> seasonal cycle anomalies [ $\mu$ atm]



1756

Fig. 3: Multi-decadal changes in the  $pCO_2$  seasonal amplitude between the two five-year 1757 1758 periods 1985-1989 and 2014-2018, for (a,c) pCO<sub>2</sub> products and (b,d) GOBMs. For panels a 1759 and b, seasonal amplitude is calculated as a winter-minus-summer difference, where winter 1760 and summer are defined as JFM and JAS (JAS and JFM) for the Northern (Southern) 1761 Hemisphere, respectively. For panels c and d seasonal amplitude is calculated as maximum-1762 minus-minimum. Dotted regions indicate where the spread of the ensemble members (STD=1 1763 level) is larger than the ensemble mean multi-decadal change. For this analysis, the nine  $pCO_2$ 1764 products listed in Table 2 and the 11 GOBMs in Table 3 (excluding OCIM) are used. The 1765 sensitivity to the choice of five-year versus 10-year versus 15-year averaging for 1766 characterizing decadal changes is displayed in Fig. S3.





1768 Fig. 4: Climatological seasonal cycle anomalies for 2014-2018 for pCO<sub>2</sub> (left column), SST 1769 (center column, dashed) and DIC (center column, solid), and change in pCO<sub>2</sub> seasonale cycle 1770 anomalies between 1985-1989 and 2014-2018 (right column) over the six aggregated biomes 1771 for GOBMs (green),  $pCO_2$  products (blue), and for OCIM (brown, dashed). The biome names 1772 as defined in Table 1 and Fig. 1 are given in each panel title. Ensemble means for GOBMs 1773 and  $pCO_2$  products are shown as thick lines while the shading indicates the standard deviation 1774 around the mean. Note the different scales of the left and right columns, and that the Southern 1775 Hemisphere sub-plots begin with the month of July.





1779 Fig. 5: Climatological seasonal amplitudes in surface DIC concentrations over 2014-2018 1780 considered as winter-minus-summer (JFM-JAS for the Northern Hemisphere and JAS-JFM 1781 for the Southern Hemisphere) for (a) the average of three  $pCO_2/TA$  (DIC) products 1782 (JMAMLR, CMEMS-LSCE-FFNN, and OceanSODA-ETHZ), (b) the average across 11 1783 GOBMs, and (c) the difference between (b) and (a), where negative (positive) values indicate 1784 that GOBMs under-represent (over-represent) the climatological seasonal amplitude of DIC 1785 variability. (d) shows latitudinal averages for GOBMs (green) and data products (blue), where 1786 bold lines represent product averages, and light lines indicate individual products. Also shown 1787 in (d) are MOBO-DIC (red dashed) and NNGv2LDEO (red solid). The MOBO-DIC 1788 climatology corresponds to the period 2004 through 2017 and NNGv2LDEO to 1995.



1791 Fig. 6: Ensemble mean difference between the thermal and nonthermal  $pCO_2$  component of 1792 its seasonal cycle amplitude ( $\Delta A$ ) for (a) the three pCO<sub>2</sub> products that include associated DIC 1793 and TA products (JMAMLR, OceanSODA-ETHZ, and CMEMS-LSCE-FFNN and (b) 11 1794 GOBMs. (c) Zonal mean of  $\Delta A$  values for GOBMs and pCO<sub>2</sub> products with bold lines 1795 showing the ensemble mean values. The olive-colored line shows the abiotic OCIM model, 1796 which is not included in the model ensemble mean.  $pCO_2$  component seasonal cycle 1797 amplitudes were computed as the absolute value of the difference between the winter minus 1798 summer seasonal averages over 2014-2018. For each case the separation into thermal and 1799 nonthermal components follows the approach of Fassbender et al. (2022), where a carbon 1800 system calculator (CO2SYS) is used to quantify the thermal component, and thereby the 1801 nonthermal component via difference from the full  $pCO_2$  annual cycle. Seasons are defined as 1802 JFM and JAS.

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Fig. 7: Decomposition of the seasonality drivers for  $pCO_2$  products and GOBMs (one standard deviation shading for GOBMs) for all six biomes. The 2014-2018 climatological seasonal cycle of  $pCO_2$  (left column) is decomposed into month-to-month changes due to two physical (SST (orange) and SSS (purple)) and two biogeochemical (salinity-normalized DIC (nDIC, blue) and salinity-normalized TA (nAlk, yellow)) drivers (2nd and 3rd column). The

1813 observational products are the three products that include DIC and TA, namely JMAMLR, 1814 OceanSODA-ETHZ, and CMEMS-LSCE-FFNN. Calculations are performed first 1815 individually on each product or GOBM, and subsequently averaged to arrive at a mean 1816 respectively for the observation-based products and GOBMs. The differences in the drivers 1817 between the  $pCO_2$  products and the GOBMs are shown in the right column - note the 1818 differences in scale. The drivers are identified via Taylor Series decomposition using 1819 PYCO2SYS to calculate the coefficients for DIC and TA, and using constant values for SST and SSS (dlnpCO<sub>2</sub>/dT=0.0423°C<sup>-1</sup> and dlnpCO<sub>2</sub>/dlnS=1) as in Sarmiento and Gruber (2006). 1820 For  $pCO_2$  itself (left column) the monthly dots are positioned mid-month, mid-way between 1821 1822 tick marks. For tendencies (columns two and three) dots are located at the end of the month, 1823 so on tick marks. SUM in columns two and three represents the sum of the four underlying 1824 tendencies.

1825



Fig. 8: Seasonal evolution of the relationship between MLD and anomalies of nDIC over the six biomes for a climatology over 2014-2018 for observation-based products (blue) and GOBMs (green). The stars indicate January. The  $pCO_2$  products that include DIC and TA, namely JMAMLR, OceanSODA-ETHZ, and CMEMS-LSCE-FFNN, are considered along with the full suite of 11 GOBMs. Observationally-based MLDs have been derived for this study from the gridded Argo-derived temperature and salinity product of Roemmich and Gilson (2009) using a density threshold criterion (Holte and Talley, 2009) using the years 2014-2018, for consistency with the nDIC fields.



Fig. 9: Mean summer (S) and winter (W) (a-c) surface pCO<sub>2</sub> anomalies and (d-f) CO<sub>2</sub> sea-to-air flux. Results for the 1985-1989 period, 2014-2018 period, and the difference between periods ( $\Delta$ ) are shown in separate columns. Sea pCO<sub>2</sub>, seasonal anomalies are calculated relative to the annual mean computed from annual means of temperature, salinity, dissolved inorganic carbon, total alkalinity, and nutrients for each year (pCO<sub>2 AM</sub>). Different magnitudes in the multi-decadal changes for each season (panel c) reflect asymmetry in the  $pCO_2$  seasonal cycle change. Ensemble mean GOBM changes are shown in green hues (11 models listed in Table 3, excluding OCIM). Ensemble mean  $pCO_2$  product (Prod.) changes are shown in blue hues (same  $pCO_2$  products as in Fig. 6). Seasons are defined as JFM and JAS. 





Fig. 10: Mean change in the seasonal cycle amplitude (winter minus summer) of  $pCO_2$  and 1859 1860 its thermal (T) and nonthermal (NT) components from the period 1985-1989 to the period 1861 2014-2018. Changes are broken down into the total change and the portion of the change attributed to the accumulation of anthropogenic carbon (Cant) versus climate (Clim: changes to 1862 1863 the ocean physical or biogeochemical state caused by anthropogenic climate change). The top, 1864 middle, and bottom rows show the ensemble mean results for the three  $pCO_2$  products, the 1865 eleven GOBMs listed in Table 3, and the CESM2 large ensemble, respectively. Shading 1866 reflects the standard deviation across the  $pCO_2$  products, GOBMs, and ensemble members. 1867 Seasons are defined as JFM and JAS. 1868





1870 Fig. 11: Multi-decadal changes in  $pCO_2$  seasonality between 1985-1989 and 2014-2018 (left 1871 column) and signal-to-noise ratio (SNR) maps for pCO<sub>2</sub> seasonality changes between 1985-1872 1989 and 2014-2018 (right column). For each case, the seasonal amplitude is defined as the 1873 absolute value of the difference between JFM and JAS averages of  $pCO_2$  in the model, such 1874 that negative values indicate decreasing amplitude. (a) the full decadal change in  $pCO_2$ 1875 seasonal amplitude, (b) the SNR for the full  $pCO_2$  signal, (c) the absolute value of the change 1876 in the thermal component of  $pCO_2$  seasonal amplitude, (d) the SNR for the thermal 1877 component change, (e) the absolute value of the change in the nonthermal component, and (f) 1878 the SNR for the nonthermal component. All fields are from the CESM2 large ensemble (50 1879 members). For each ensemble member, the change in the amplitude of the annual cycle was 1880 measured by (i) detrending the 1985-2018 time series by subtracting a quadratic polynomial 1881 fit, (ii) calculating the annual cycle strength as the absolute value of the JFM-JAS difference 1882 in  $pCO_2$  for each year, (iii) averaging the five yearly values for each period, and (iv) 1883 subtracting the average over the earlier period from the average over the later period. For the 1884 full  $pCO_2$  seasonal cycle, (i) the signal is defined as the ensemble mean change, and (ii) the

1885 noise as the ensemble standard deviation, and (iii) the SNR indicates the confidence levels for 1886 emergence. This is done analogously for the thermal component of seasonality changes for (iv) the signal, (v) the noise, and (vi) the confidence intervals for emergence. Likewise for the 1887 1888 nonthermal component, (vii) the signal, (viii) the noise, and (ix) the confidence intervals for 1889 emergence. Absolute magnitudes are chosen to emphasize the strength of the signal, so that 1890 the thermal and nonthermal fields in the left column do not necessarily sum to the total  $pCO_2$ 1891 signal. The decomposition of the thermal and nonthermal components of the  $pCO_2$  seasonal 1892 cycle is described in the Appendix.



1894

1895 Fig. 12: Box plots of the absolute amplitude of the sea surface  $pCO_2$  seasonal cycle for the 1896 period 2014-2018 (left) and the change in the amplitude of the  $pCO_2$  seasonal cycle between 1897 1985-1989 and 2014-2018 (right) based on pCO<sub>2</sub> products (blue) and five large ensemble 1898 experiments. For the large ensemble experiments, the number of ensemble members used for 1899 each model is shown in parentheses in the legend. The seasonal cycle amplitude is calculated 1900 as the absolute value of biome-averaged winter-minus-summer differences (JFM-JAS and 1901 JAS-JFM for the Northern and Southern Hemisphere, respectively). The boxes show the 1902 values between the first quartile (25%) and the third quartile (75%) with a line at the median 1903 (50%), with the whiskers indicating the minimum and maximum values. Note that for the 1904  $pCO_2$  products, the uncertainty range reflects uncertainty between the available nine

1905 observational  $pCO_2$  products, whereas the uncertainty range for the large ensemble 1906 simulations indicates natural climate variability as simulated by each model. For each 1907 ensemble member and data product, the change of the amplitude of the seasonal cycle for each region was measured by (i) detrending the 1985-2018 time series by subtracting a 1908 1909 quadratic polynomial fit, (ii) calculating the seasonal cycle amplitude as the winter-minus-1910 summer  $pCO_2$  values for each year, (iii) averaging the five yearly values for each period, (iv) 1911 subtracting the earlier period average from the latter, and (v) calculating the spatial mean with 1912 latitude weighting. The amplitude of the seasonal cycle in 1985-1989 was obtained in step 1913 (iii). For each biome the sequencing of the colors is identical to the sequencing in the legend.



1915

1916 Fig. 13: Same as Fig. 4 but with CMIP6 models as listed in Table 6. Note that the 1917 climatologies for  $pCO_2$  (left column) and both DIC and SST (middle column) are taken over 1918 the years 2010-2014 (last five years of the CMIP6 historical simulations) and that the change 1919 in  $pCO_2$  seasonality (right column) is calculated as the difference between 2010-2014 and 1920 1985-1989.

- 1921
- 1922
- 1923

# 2 Seasonal variability of the surface ocean carbon cycle: a

synthesis 3 4 **Supplementary Materials** 5 Draft: April 3<sup>rd</sup> 2023 6 7 Keith B. Rodgers<sup>1,2,\*,†,</sup> Jörg Schwinger<sup>3,\*,†</sup>, Andrea J. Fassbender<sup>4,†</sup>, Peter Landschützer<sup>5</sup>, 8 Ryohei Yamaguchi<sup>6,†</sup>, et al. 9 10 † principal scientific contribution 11 \* corresponding authors: keithbrodgers@gmail.com, jrsc@norceresearch.no 1. Center for Climate Physics, Institute for Basic Science, Busan, Republic of Korea 12 13 2. Pusan National University, Busan, Republic of Korea 3. NORCE Climate & Environment, Bjerknes Centre for Climate Research, Bergen, 14 15 Norway 4. NOAA/OAR Pacific Marine Environmental Laboratory, Seattle, USA 16 17 5. Flanders Marine Institute (VLIZ), Oostende, Belgium 6. Japan Agency for Marine-Earth Science and Technology, Yokosuka, Japan 18 19 20 21

### 22 Surface DIC products associated with *p*CO<sub>2</sub> products

23

24 Here we provide more details on the  $pCO_2/TA$  products that provide time-varying surface

- 25 DIC. These have been provided for CMEMS-LSCE, OceanSODA-ETHZ, and JMAMLR.
- 26

27 For CMEMS-LSCE, surface ocean DIC is calculated by using the CO2SYS speciation 28 software (Lewis & Wallace, 1998; Van Heuven et al., 2011) given the reconstructions of 29  $pCO_2$  and TA, nutrient concentrations (silicate, nitrate, phosphate), physical variables (SST 30 and SSS), and dissociation constants. Total dissociation constants follow the best practices of 31 (Dickson, 2010), wherein K<sub>1</sub> and K<sub>2</sub> are from Lueker et al. (2000). K<sub>HSO4</sub> is from Dickson et 32 al. (1990), and the formulation of boron to salinity ratio is from Uppstrom (1974). Details of 33 the algorithms and derivations are documented in Chau et al. (2022). TA is derived from 34 LIAR (Carter et al., 2018) - an approach of multiple linear regressions.

For OceanSODA-ETHZ, TA is (equivalent to the sea surface  $pCO_2$ ) estimated using ensembles of two-step cluster-regression approaches, using K-means clustering for the first step, and a combination of neural networks, gradient-boosted trees and support vector regression for the second step (Gregor & Gruber, 2021). K<sub>1</sub> and K<sub>2</sub> are from the Dickson and Millero (1987) refit of Mehrbach et al. (1973), as these constants had the lowest error for the  $pCO_2$ -TA pairing (Raimondi et al., 2019). The K<sub>HSO4</sub> and boron-salinity ratios are consistent with the CMEMS-LSCE approach.

42 For the JMAMLR product, salinity-normalized DIC (nDIC) is calculated by coupling each set 43 of  $pCO_2$ , SST and SSS in SOCATv2019 (Bakker et al., 2016) with monthly fields of surface 44 ocean TA expressed as a function of sea surface dynamic height (SSDH) and SSS that was 45 built on GLODAPv2.2019 (Olsen et al., 2019; Takatani et al., 2014). The dissociation 46 constants used are those recommended in the best practices of Dickson (2010). They are then 47 fitted by multiple linear regressions for respective ocean domains with explanatory variables 48 such as SST, SSS, SSDH, chlorophyll, mixed layer depth and year of measurements to 49 account for the trend of DIC increase due to anthropogenic CO<sub>2</sub> uptake. Monthly fields of 50 surface ocean nDIC are derived by combining these empirical expressions of nDIC with data 51 sets of these explanatory variables derived from satellite measurements and an ocean data 52 assimilation (Iida et al., 2021).

53

#### 54 Three Dimensional DIC climatologies

55

Here we provide a brief overview of the three-dimensional DIC climatologies used to assess GOBM representation of vertical gradients in DIC. The two climatologies described below are MOBO-DIC and NNGv2LDEO, and they are both independent of the surface DIC products that are directly associated with the surface  $pCO_2$  products described above.

60

61 An interior DIC climatology DP is provided from the Mapped Observation-Based Oceanic 62 DIC (MOBO-DIC) product by Keppler et al. (2020a; 2020b). MOBO-DIC is a machine-63 learning-based approach, and is built on the same 2-step neural network method that has been 64 used previously to reconstruct surface ocean  $pCO_2$  for the SOM-FFN product by 65 Landschützer et al. (2016) and adjusted to resolve DIC fields in the interior ocean. This two-66 step cluster-regression approach first divides the ocean into clusters using self-organizing 67 maps (SOM), and then runs a feed-forward network (FFN) in each of the SOM-clusters. During the FFN step, this version of MOBO-DIC uses mapped observation-based temperature 68 69 and salinity fields from the Argo program (Roemmich & Gilson, 2009) together with 70 climatological nutrient and oxygen fields from the World Ocean Atlas (WOA18; (Garcia et 71 al., 2019)) to obtain the statistical relationships between these predictor data and interior 72 ocean DIC measurements from the GLODAPv2019 database (Olsen et al., 2019). These 73 relationships are then applied to reconstruct gap-filled maps of the interior oceanic DIC. The differences between MOBO-DIC and the original SOM-FFN approach include that the target 74 75 variable to be mapped is DIC (compared to  $pCO_2$  in SOM-FFN), the fields have an additional 76 dimension (depth), and different predictors are used due to data availability and different 77 processes in the water column relative to the surface. In addition, this version of MOBO-DIC 78 only resolves a monthly climatology (compared to monthly resolution in SOM-FFN). As a 79 result of these differences, the structure of the networks differs in MOBO-DIC, with fewer 80 clusters in the SOM-step, and fewer neurons in the FFN step. More information on the method is provided in Keppler et all (2020a). This MOBO-DIC monthly climatology is at 1°
horizontal resolution on 28 depth levels (2.5-2000 m), and is based on data from 2004 through
2017.

84

85 We also use the NNGv2LDEO monthly climatology of interior DIC by Broullón et al. (2020). 86 This DIC product is based on a two-layer FFN neural network, that utilizes physical and 87 biogeochemical predictor data from the World Ocean Atlas (WOA13; Garcia et al., 2013; 88 Locarnini et al., 2013; Zweng et al., 2019), as well as information on the depth, horizontal 89 location, and year of the measurement, to reconstruct gap-filled maps of the interior oceanic 90 DIC. The relationship from predictors and DIC measurements is established through a 91 combination of observation-based DIC measurements from GLDAPv2.2019 (Olsen et al., 92 2019) and DIC obtained from pCO2 from the Lamont Doherty Earth Observatory (LDEO) 93 database (Takahashi et al., 2017), combined with TA from the NNGv2 neural network of 94 Broullón et al. (2019). These NNGv2LDEO DIC fields are at 1° horizontal resolution, on 102 95 depth levels (0–5500 m), where the upper 1500 m resolve a monthly climatology and below 96 1500 m it is at annual resolution; the data are centered around the year 1995.

97

## 98 Attribution of discrepancies in the *p*CO<sub>2</sub> seasonal cycle phase and amplitude

99

In the main text section describing climatological  $pCO_2$  seasonal cycle drivers, we applied the method of Fassbender et al. (2022), which is a modified version of the framework originally developed by Takahashi et al. (1993; 2002), where for each grid point local temperature and  $pCO_2$  are used as follows to define the thermal ( $pCO_2$  <sub>T</sub>) and nonthermal ( $pCO_2$  <sub>NT</sub>) components of  $pCO_2$  seasonal variability:

$$pCO_{2T} = pCO_{2am} * exp[0.0423 (T_{mm} - T_{am})]$$
(1)  
$$pCO_{2NT} = pCO_{2mm} * exp[0.0423 (T_{am} - T_{mm})]$$
(2)

Within this approach the subscripts am and mm represent annual means and monthly means, respectively. However, one of the limitations of the method of Takahashi et al. (1993; 2002) is that the two components in (1) and (2) do not sum to reproduce the full  $pCO_2$  seasonal 108 cycle (e.g.,  $pCO_{2 \text{ mm}} \neq pCO_{2 \text{ T}} + pCO_{2 \text{ NT}} - pCO_{2 \text{ am}}$ ). This is because the thermal sensitivity of 109  $pCO_2$  (expressed with a factor of 0.0423/°K in the equations above) in fact varies slightly 110 with background chemistry (Wanninkhof et al., 1999).

111

In this study, we use the alternative approach of Fassbender et al. (2022), where an important focus of the analysis is to identify asymmetries in seasonal  $pCO_2$  variations. For this case, rather than working with an annual mean  $pCO_2$  to characterize asymmetries, one defines a neutral  $pCO_2$  with respect to the annual cycle as being the  $pCO_2$  one would obtain using annual means of its drivers (T, S, DIC, TA, PO<sub>4</sub>, and SiO<sub>4</sub>), which reflects the mean  $pCO_2$  if  $pCO_2$  were to respond linearly to the seasonal variability of its drivers. With this in mind,  $pCO_2 AM$  is given by:

$$pCO_{2AM} = f(\overline{T}, \overline{S}, \overline{DIC}, \overline{TA}, \overline{PO_4}, \overline{SIO_4}) \quad (3)$$

where the function f() represents a carbon system calculator. The time-varying (monthly mean) thermally-driven  $pCO_2$  component can be identified using the same approach but by using monthly varying output temperature for the carbonate system calculator:

$$pCO_{2TFASS} = f(T_{mm}, \overline{S}, \overline{DIC}, \overline{TA}, \overline{PO_4}, \overline{SIO_4}) \quad (4)$$

122 The thermal  $pCO_2$  component seasonal cycle anomaly is then determined as:

$$pCO_{2T_{anom}} = pCO_{2TFASS} - pCO_{2AM}$$
(5)

123 making it possible to calculate the nonthermal component by difference:

$$pCO_{2 NT FASS} = pCO_2 - pCO_{2 T_{anom}}$$
(6)

124 These definitions for thermal- and nonthermal decomposition in Equations (5)-(6), which are

125 applied in Fig. 6, will prove to be important to our attribution of decadal changes in  $pCO_2$ 

126 seasonality, to identify whether decadal changes project differently onto summer and winter.

127

#### 128 Attribution of decadal changes in the *p*CO<sub>2</sub> seasonal cycle

130 The goal of the attribution analysis is to isolate the direct ( $C_{ant}$ ) and indirect (climate change; 131 Clim) influences of  $C_{ant}$  on  $pCO_2$  seasonal cycle changes between the time periods of interest 132 via offline calculations. Due to the short duration over which this method is applied relative to 133 what it was originally designed for (i.e., >100-year time scales), both the C<sub>ant</sub> and Clim 134 components of the total  $pCO_2$  seasonal cycle change may contain non-trivial interference 135 from natural variability, which we attempt to constrain with the CESM2 LE analysis in 136 Sections 3.3.1 and 3.3.2. Here we provide further description of the method of Fassbender et 137 al. (2022) to facilitate understanding of the attribution analysis for decadal changes; however, 138 readers are encouraged to review the original publication for complete methodological details.

139

140 To perform the attribution analysis, we focus on isolating the direct and indirect impacts of 141 C<sub>ant</sub> on thermal and nonthermal  $pCO_2$  component seasonal cycle changes. Since these 142 calculations are performed offline, the method requires an initial step of reconstructing the 143 model  $pCO_2$  values. A Taylor series decomposition was used previously in **Section 3.2.2** to 144 compare discrepancies between the GOBM and  $pCO_2$  product seasonal  $pCO_2$  variability 145 resulting from each of its drivers. A similar approach is used here for attribution following the 146 method of Fassbender et al. (2022).

147

148 Changes in surface ocean  $pCO_2$  with time can be expressed as the sum of temporal changes in 149 the various  $pCO_2$  drivers multiplied by the sensitivity of  $pCO_2$  to that driver:

$$\frac{dpCO_2}{dt} = \left(\frac{\partial pCO_2}{\partial DIC}\right) \left(\frac{\partial DIC}{\partial t}\right) + \left(\frac{\partial pCO_2}{\partial TA}\right) \left(\frac{\partial TA}{\partial t}\right) + \left(\frac{\partial pCO_2}{\partial SSS}\right) \left(\frac{\partial SSS}{\partial t}\right) + \left(\frac{\partial pCO_2}{\partial SST}\right) \left(\frac{\partial SST}{\partial t}\right) (7)$$

The first three terms on the right-hand side reflect the nonthermal  $pCO_2$  drivers and the last term on the right-hand side reflects the thermal  $pCO_2$  driver. For the thermal sensitivity, we use a carbonate system calculator, as described in the preceding section, to isolate the monthly thermal  $pCO_2$  component anomalies ( $pCO_{2 \text{ Tanom}}$ ) relative to  $pCO_{2 \text{ AM}}$  at each grid point during each year of 1985-1989 and 2014-2018. For the remaining sensitivity terms, we rely on familiar relative sensitivity terms including the Revelle Factor (RF; (Bolin & Eriksson, 1959; Broecker et al., 1979)), Alkalinity Factor (AF; (Takahashi et al., 1993)), and Salinity Factor
(Takahashi et al., 1993), which all take the same form, with an example for the AF given
here:

$$AF = \left(\frac{\Delta pCO_2}{\overline{pCO_2}}\right) \times \left(\frac{\Delta TA}{\overline{TA}}\right)^{-1} \quad (8)$$

Where  $\Delta p CO_2$  reflects the change imposed by a small TA perturbation ( $\Delta TA$ ) to initial TA and  $pCO_2$  values with all other parameters held constant. The RF is provided as direct output from the carbonate system calculator while the AF and SF are calculated using the carbonate system calculator by independently imposing small TA and salinity perturbations ( $\pm 0.01$  $\mu$ mol/kg TA and  $\pm 0.0001$ , respectively) for each grid point and month.

164

165 Monthly anomalies ( $\Delta$ ) in DIC, TA and SSS, relative to their annual mean values, can then be 166 multiplied by their corresponding relative sensitivity factors to estimate the relative change in 167 *p*CO<sub>2</sub>. These relative changes in *p*CO<sub>2</sub> are then summed and multiplied by *p*CO<sub>2 AM</sub> to 168 estimate the corresponding monthly nonthermal *p*CO<sub>2</sub> component anomalies (*p*CO<sub>2 NT</sub>) 169 relative to *p*CO<sub>2 AM</sub> at each grid point during each year of 1985-1989 and 2014-2018, as 170 follows:

$$pCO_{2NT_{anom}} = pCO_{2AM} \times \left( \left[ RF \times \left( \frac{\Delta DIC}{\overline{DIC}} \right) \right] + \left[ AF \times \left( \frac{\Delta TA}{\overline{TA}} \right) \right] + \left[ SF \times \left( \frac{\Delta SSS}{\overline{SSS}} \right) \right] \right)$$
(9)

Summing the thermal and nonthermal component anomalies with  $pCO_{2 AM}$  yields the total pCO<sub>2</sub> value consistent with the model  $pCO_2$  output for the time periods of interest. The offline reconstruction of the model  $pCO_2$  field is important in later methodological steps when we difference a suite of calculated values that are subject to the same offline estimation uncertainties.

176

To isolate the influence of  $C_{ant}$  and Clim on  $pCO_{2 NT}$ , we perform the same calculations outlined above but use the climatological monthly mean RF and AF values from the 1985-1989 period in each year of the 2014-2018 calculations. This holds the sensitivity of  $pCO_2$  to the nonthermal carbonate system drivers constant at the 1980s level, providing an estimate of the indirect impact of  $C_{ant}$  (i.e., Clim) on the nonthermal  $pCO_2$  component ( $pCO_{2 NTanom-Clim}$ ). 182 The difference between  $pCO_{2 \text{ NTanom}}$  and  $pCO_{2 \text{ NTanom-Clim}}$  yields an estimate of the direct 183 impact of C<sub>ant</sub> on the nonthermal  $pCO_2$  component.

$$pCO_{2 NT_{anom}-C_{ant}} = pCO_{2 NT_{anom}} - pCO_{2 NT_{anom}-Clim}$$
(10)

Similarly, to isolate the influence of  $C_{ant}$  and Clim on  $pCO_2$  T, we use the same carbonate system calculator approach described in the previous section but apply the climatological  $pCO_2$  AM value from the 1985-1989 period ( $pCO_2$  AM80s) in each year of the 2014-2018 calculations as input values rather than annual mean DIC.

$$pCO_{2T FASS80s} = f\left(SST_{mm}, \overline{SSS}, pCO_{2AM80s}, \overline{TA}, \overline{PO_4}, \overline{SIO_4}\right) \quad (11)$$

This holds the thermal sensitivity of  $pCO_2$  constant at the 1980s level providing an estimate of the indirect impact of C<sub>ant</sub> (i.e., Clim) on the thermal  $pCO_2$  component ( $pCO_2$  <sub>Tanom-Clim</sub>). We then subtract  $pCO_2$  <sub>AM80s</sub> to yield the thermal term anomalies for each year of the 2014-2018 period.

$$pCO_{2T_{anom}-Clim} = pCO_{2TFASS80s} - pCO_{2AM80s}$$
(12)

192 The difference between  $pCO_{2 \text{ Tanom}}$  and  $pCO_{2 \text{ Tanom-Clim}}$  yields an estimate of the direct impact 193 of C<sub>ant</sub> on the thermal  $pCO_2$  component.

$$pCO_{2T_{anom}-C_{ant}} = pCO_{2T_{anom}} - pCO_{2T_{anom}-Clim}$$
(13)

Summing the thermal and nonthermal anomalies derived using time-varying and constant sensitivity terms with  $pCO_{2 AM}$  yields the total  $pCO_{2 Clim}$  values, respectively, which can be differenced to isolate the C<sub>ant</sub> contribution:

$$pCO_{2 C_{ant}} = pCO_2 - pCO_2 Clim \quad (14)$$

197 With the Clim and  $C_{ant}$  components of total  $pCO_2$  differentiated relative to the 1980s 198 reference period, we can assess attribution for the changes in seasonal cycle characteristics in 199 the thermal, nonthermal, and full  $pCO_2$  values for the 2014-2018 period.

200

201



Fig. S1: Time series of the net amplitude (annual maximum minus minimum) of monthly  $CO_2$ fluxes spatially integrated over the six aggregated biomes shown in Fig. 1. The GOBMs (green) and  $pCO_2$  products (blue) are shown for their product-mean (bold line) and 1-std of their spread amongst individual products (shaded).



Fig. S2: Time series of  $pCO_2$  averaged over biomes showing annual maximum and annual minimum values, both for GOBMs and for  $pCO_2$  products. Lines indicate the mean and shading indicates the one standard deviation range for each ensemble of estimates.



215 Fig. S3: Evaluation of the sensitivity of the decadal changes in  $pCO_2$  seasonal amplitude for

216 the pCO<sub>2</sub> products using (a) five-year climatologies, (b) 10-year climatologies, and (c) 15-

217 year climatologies. Panel (a) is the same field shown in **Fig. 3a**.

218



220 221 Fig. S4: Climatological seasonal cycle anomalies for 2014-2018 DIC over the six aggregated 222 biomes for GOBMs (green), pCO<sub>2</sub> products (blue), for OCIM (brown, dashed), and for the 223 two DIC-climatologies MOBO-DIC (lightblue, dashed) and NNGv2LDEO (lightblue, dash-224 dotted). The biome names as defined in Table 1 and Fig. 1 are given in each panel title. 225 Ensemble means for GOBMs and  $pCO_2$  products are shown as thick lines while the shading 226 indicates the standard deviation around the mean. Note that the DIC climatologies are 227 referenced to time periods 2004 through 2017 for MOBO-DIC and 1995 for NNGv2LDEO 228 different from the averaging period used for GOBMs and  $pCO_2$  products (2014-2018). Also 229 note that, due to differences in spatial data coverage, the inclusion of MOBO-DIC and 230 NNGv2LDEO entails that the areas.

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