Strengthening seasonal marine CO_2 variations due to increasing atmospheric CO_2

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The increase of atmospheric carbon dioxide (CO_2)[1] has been predicted to impact the seasonal cycle of inorganic carbon in the global ocean[2,3], yet the observational evidence to verify this prediction has been missing so far. Here, using an observation-based product of the oceanic partial pressure of carbon dioxide (pCO_2) covering the past 34 years, we find that the winter-to-summer difference of the pCO_2 has increased on average by 2.2$\pm$0.4 μatm decade^{-1} from 1982 through 2015 poleward of 10° latitude. This is largely in agreement with the trend expected from thermodynamic considerations. Most of the increase stems from the seasonality of the drivers acting on an increasing ocean pCO_2 caused by the uptake of anthropogenic CO_2 from the atmosphere. In the high latitudes, the concurrent ocean acidification induced changes in the buffer capacity of the ocean enhance this effect. This strengthening of the seasonal winter-minus-summer difference pushes the global ocean earlier towards critical

thresholds inducing stress to ocean ecosystems and fisheries[4]. Our study provides the observational evidence for this strengthening seasonal difference in the oceanic carbon cycle on a global scale, illustrating the inevitable consequences of anthropogenic CO_2 emissions.

The oceanic uptake of anthropogenic CO_2[1,5] is causing major shifts in the surface ocean inorganic carbon system[6]. These shifts include increasing concentrations of dissolved CO_2, and hence higher pCO_2, but also a reduction of the carbonate ion concentration and pH, and are often collectively referred to as ocean acidification[4,7]. One of the predicted consequence of these chemical changes is an increase in the seasonal variation of the surface ocean pCO_2[2,3,8]. However, this prediction has not been confirmed experimentally by in-situ data on a global scale so far. Here, by analysing surface ocean pCO_2 observations collected over the past 34 years, we demonstrate that the seasonal pCO_2 difference indeed has increased substantially in recent decades, implying stronger stress to organisms that are sensitive to ocean acidification[3,9].

Our results are based on a collection of ship and mooring surface ocean pCO_2 measurements assembled by the Surface Ocean CO_2 Atlas effort (SOCATv4)[10,11]. These measurements were combined with our 2-step neural network data interpolation technique SOM-FFN (self organizing map feed forward network)[12,13] to reconstruct changes in surface ocean pCO_2 from 1982 through 2015 at monthly intervals and with a spatial resolution of $1^\circ \times 1^\circ$. The seasonal differences of pCO_2 were computed by first fitting the interpolated data with a combination of harmonic and polynomial functions and then determining the differences for the January through March means and for the July through September means (see Methods). Even though there exists substantial interannual

variability in the strength of the seasonal pCO_2 difference, expressed here as the seasonal maximum minus the seasonal minimum, its trend is clearly positive in all extratropical regions ranging between 1.1±0.3 μatm decade^{-1} and 2.9±0.3 μatm decade^{-1} (Fig. 1) with an average rate of 2.2±0.4 μatm decade^{-1}.

The seasonal differences observed at the Hawaiian Ocean Timeseries station (HOT)[14] and at the Station S/Bermuda Atlantic Timeseries Station site (StaS/BATS)[15–17], where measurements are available since 1988 and 1983, respectively, support our estimates. The diagnosed trends of 1.5 ± 1.1 μatm decade^{-1} (StaS/BATS) and 3.8 ± 2.4 μatm decade^{-1} (HOT) are statistically indistinguishable from those inferred from our neural network based estimates at these two locations (i.e., 1.5±1.8 and 0.2±1.8 μatm decade^{-1} respectively - see Supplementary Information). The uncertainties of these local trends are quite large, however, largely reflecting the strong year-to-year fluctuations of the seasonal cycle of surface ocean pCO_2 at this local scale. While this prevents a very thorough quantitative validation of our neural network based analyses, they nevertheless support our estimate of a rate of increase of 2.3 ± 0.4 μatm decade^{-1} in the subtropical band (10°N-40°N) of the northern hemisphere. This is particularly encouraging considering the local nature of these station data in comparison to our larger scale SOM-FFN estimates. This conclusion is further supported by the residuals of our interpolated product having no seasonal trend, i.e., the diagnosed trend in the seasonal difference is not an artifact of the interpolation, but stems from the pCO_2 observations (see Supplementary Information). The long-term changes in the winter-minus-summer differences for the large-scale regions analyzed here are also significantly larger than the estimated uncertainty in the reconstructed seasonal cycles for pCO_2.

The seasonal differences of the surface $p\mathrm{CO}_2$ increase everywhere, with the winter-minus-summer differences becoming more negative in the low latitudes (equatorward of $\sim$40°), and more positive in the high latitudes (poleward of $\sim$40°) (Fig. 2). This change in the sign between the low and the high latitudes corresponds to the seasonal maxima of $p\mathrm{CO}_2$ being 6 months out of phase between these two bands (Fig. 2e). In the low latitudes, the seasonal cycle has a maximum in summer, and thus a negative winter-minus-summer difference (Fig. 2a, c). In contrast, the seasonal cycle in the high latitudes has a maximum in winter, leading to a positive winter-minus-summer difference in $p\mathrm{CO}_2$ (Fig. 2b, d). For our further analyses, we will use the winter-minus-summer difference rather than the absolute difference as our metric for changes in the seasonal cycle, mainly because including the sign permits us to investigate the dominant drivers for the winter-minus-summer trends.

We can quantify these drivers by separating the seasonal cycle of the surface ocean $p\mathrm{CO}_2$ into the thermal part driven by the seasonal variations in sea-surface temperature (SST), and into the non-thermal part driven by the seasonal variations by all other factors, namely dissolved inorganic carbon (DIC), alkalinity, and salinity[6, 18, 19] (Fig. 3). This separation reveals that the winter-minus-summer differences of the thermal component became more negative between the 1985-1989 and the 2010-2014 periods, while the winter-minus-summer difference in the non-thermal component became more positive between these two periods. While these trends oppose each other, in the low latitudes (Fig. 3a), the negative winter-minus-summer trend in the thermal component dominates the increase in the seasonal $p\mathrm{CO}_2$ difference, whereas in the high latitude regions (Fig. 3b), the positive winter-minus-summer trend in the non-thermal component is the dominating compo-

nent. Consequently, adding both components, we find an increase in the seasonal $p\mathrm{CO}_2$ difference everywhere in the global ocean.

The regionally varying contribution of the thermal and non-thermal components to the changing seasonal cycle is also clearly visible in a regionally more refined analysis of the winter-minus-summer trends (Fig. 4), even though the regional winter-minus-summer $p\mathrm{CO}_2$ trends vary more substantially than the broad latitude band averages (Fig. 4a). Trends in the winter-minus-summer $p\mathrm{CO}_2$ difference of the thermal and non-thermal components are locally even stronger, reaching 10 μatm decade^{-1}, but the compensatory nature of the thermal and non-thermal trend components occurs nearly everywhere (Fig. 4b, c).

Thermodynamic consideration of the CO_2 system in seawater permits us to determine and quantify the drivers behind the increases in the seasonal differences as well as the compensatory nature between the thermal and non-thermal components. Starting from a Taylor expansion of the $p\mathrm{CO}_2$ drivers [6,18] and using some simplifications, we can show that three mechanisms are driving the increase in the seasonal differences (see Methods). The first mechanism is associated with the long-term increase in the mean concentration of CO_2 in the surface ocean, i.e., $[\mathrm{CO}_2]_{aq}$, caused by the uptake of anthropogenic CO_2 from the atmosphere. This mechanism effects both the thermal and non-thermal driven seasonal cycle because of their respective CO_2 sensitivities. The second mechanism is associated with the reaction of the added CO_2 with the carbonate ion in seawater, resulting in a reduction of the capacity of the surface ocean CO_2 system to buffer against changes. This consequence, reflected in an increase in the Revelle (buffer) factor implies that the surface

ocean CO_2 system becomes more sensitive to the seasonal cycle in DIC and alkalinity[3]. The third mechanism changes the seasonal pCO_2 difference by altering the seasonality of its drivers.

We do not have sufficient long-term observations of surface DIC, alkalinity and salinity to determine the changes in their seasonal cycles required to quantitatively estimate the contribution of the third mechanism. In response, we investigate how much of the observed trends can be attributed to the increase in the CO_2 concentration and buffer factor and then discuss the role of the third mechanism based on the mismatch between the observations and the estimates from the first two mechanisms.

The comparison between our observation-based winter-minus-summer trends and those from the thermodynamic consideration (dashed green lines in Fig. 4) illustrates that the first two mechanisms can explain the majority of the observation-based latitudinal trend pattern. This suggests that the contribution of the third mechanisms, i.e., changes in the seasonality of the pCO_2 drivers, is comparably small. The dominance of the first two mechanisms implies that it is indeed the long-term changes in the surface ocean CO_2 chemistry linked to the increasing uptake of anthropogenic CO_2 that causes the increase in the winter-minus-summer difference in surface ocean pCO_2, confirming model-based analyses[2,3,20].

Further details emerge when the thermal and the non-thermal pCO_2 components are considered separately (Fig. 4b,c). In particular, this analysis allows us to explain why the thermal and non-thermal components drive the change in the low and high latitudes, respectively. As it turns out this is the result of the increasing oceanic CO_2 concentration being responsible for roughly

two thirds of the predicted trend in the non-thermal component and for all of the predicted trend in the thermal component (see Supplementary Figure 8). This dominance results in this driver enhancing both components in roughly equal manner, but because these two components have opposing seasonality, the net effect depends on which component dominates the seasonal cycle. As a result, in regions where the seasonal cycle of $p\mathrm{CO}_2$ is dominated by the thermal component, the trend in the winter-minus-summer difference is dominated by this component as well. In an analogous manner, the non-thermal component drives both the seasonal cycle and the trend in the winter-minus-summer difference in the high latitudes.

While the consideration of the first two mechanisms explain most of the observed trends, there are also substantial differences, implying changes in the seasonality of the $p\mathrm{CO}_2$ drivers. Figure 4 reveals that the underestimation of the winter-minus-summer trend in the low and temperate latitudes of the northern hemisphere stems from an underestimation of the thermal component (Fig. 4b). This implies that a trend toward a stronger seasonal warming of the sea surface contributed to the larger seasonal difference in $p\mathrm{CO}_2$ as well. In contrast, the underestimation of the observation-based trend in the low and temperate latitudes of the southern hemisphere (Fig. 4a) stems primarily from the non-thermal $p\mathrm{CO}_2$ component (Fig. 4c). This implies that a reduction or shift of the seasonal cycle of DIC, or an increase in the seasonal cycle of alkalinity contributed to the weaker increase of the winter-minus-summer difference. A plausible cause is natural variability of the surface ocean $p\mathrm{CO}_2$ in the subtropical Pacific in response to El Niño Southern Oscillation (ENSO). The decadal scale variability of the Southern Ocean carbon sink[21,22] may matter as well, particularly since a weaker seasonal cycle of DIC would be consistent with recent estimates sug-

gesting a weaker accumulation of CO_2 in the surface waters of the Southern Ocean[22] linked to changes in the shallow overturning circulation[23]. This mismatch in the non-thermal seasonal difference trends, however, might also reflect our limitation to estimate seasonal trends in the austral winter due to the limited amount of seasonal CO_2 measurements[24].

Our ability to experimentally verify the increase in the seasonal variations of the surface ocean pCO_2 is very encouraging. It demonstrates the great advances of the surface ocean observing networks and their interpretation through various interpolation schemes[11,25]. Our finding that the majority of the increase in the seasonal winter-minus-summer difference is driven by the rise in atmospheric CO_2 implies that this increase should be a very robust feature across different models and observations, as it does not depend on the magnitude of climate change and any feedbacks between climate and the ocean carbon cycle.

The increase in the seasonal difference of the surface ocean pCO_2 enhances in a substantial manner the effects of ocean acidification[4,26,27] on marine organisms, as they find themselves exposed earlier to higher levels of ocean acidification, possibly inducing the transition across critical thresholds, harmful to ocean ecosystems and fisheries, such as hypercapnia and low saturation states with regard to calcium carbonate ($CaCO_3$)[3,9,28]. Additionally, the observation-based trends are subject to substantial natural climate variability and the full extent of the seasonal pCO_2 difference is likely still masked[29]. This circumstance does not allow us to fully distinguish between decadal scale climate variability and anthropogenic trends, a difference that can only be resolved by long-term observational records. Our observation-based study however shows that anthropogenic

CO_2 emissions have already left a detectable imprint on the marine carbon cycle in the form of an increasing seasonal difference of the surface ocean CO_2 over the past decades.

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Contributions PL and NG designed the study with input from DCEB. NG and PL developed the theoretical framework and wrote the paper together with DCEB, IS and KDS. PL developed the neural network method and performed the analysis, assisted by NG, IS and KDS. DCEB led the SOCAT synthesis effort that underlies this work. All authors discussed the results and implications and commented on the manuscript at all stages.

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Methods

Calculation of the seasonal differences and trends To compute the winter-minus-summer differences of our $p\mathrm{CO}_2$ product, we first fit a 3rd order polynomial (to account for trends) and 4th order harmonic function (to reproduce the seasonality) to all data to reproduce the full seasonal cycle[30] :

$$\begin{aligned} f(t) &= a_1 + a_2 \cdot t + a_3 \cdot t^2 + a_4 \cdot \sin(2 \cdot \pi \cdot t/T) + \\ & \quad a_5 \cdot \cos(2 \cdot \pi \cdot t/T) + a_6 \cdot \sin(4 \cdot \pi \cdot t/T) + a_7 \cdot \cos(4 \cdot \pi \cdot t/T) \end{aligned} \tag{1}$$

where t is time in years and T is the period, chosen here as one year.

We recreate the seasonal cycle of a certain year by fitting equation (1) to every full analysis year as well as the year before and after that creating 3-year running timeseries. Years at the beginning or the end of our timeseries are reconstructed using the 2 following or proceeding years.

From the resulting harmonic function $f(t)$ segments, we calculate winter averages in the northern hemisphere as the mean of the months January, February and March and summer averages as the mean of the months July, August and September and vice versa for the southern hemisphere. The seasonal maximum minus seasonal minimum for a certain year is then calculated as the difference between winter-minus-summer means.

Trends in these winter-minus-summer differences are then calculated from the slope of the linear regression line fit to the 34-year timeseries and uncertainties are derived from the t-statistic of the regression (see below). We repeated our analysis without the quadratic ($a_3 \cdot t^2$) and linear terms ($a_2 \cdot t$) in equation (1) but find only small difference between the estimated cycles and seasonal trends (not shown).

Calculation of thermal and non-thermal components We split the observation-based $p\mathrm{CO}_2$ into its thermal (superscript th) and non-thermal (superscript ^{nt}) components, i.e., that part driven by the seasonal

variations in SST, and that part driven by the seasonal variations by all other factors, namely DIC, alkalinity, and salinity[18,19]. To compute the thermal component, we use the well established temperature sensitivity of CO_2 (γ_T) of 4.23%/°C[18]. This experimentally determined sensitivity may divert from the exact local sensitivity, but this error is very small and therefore not further considered. Furthermore, we perturb the annual mean pCO$_2$, $< p\mathrm{CO}_2 >_{annual}$, with the observed temperature anomalies, i.e., the differences between the measured SST and the long-term mean SST, (<SST>):

$$p\mathrm{CO}_2^{th} =< p\mathrm{CO}_2 >_{\mathrm{annual}} \cdot \exp(\gamma_T \cdot (\mathrm{SST} - < \mathrm{SST} >)) \tag{2}$$

The non-thermal component is computed by removing the temperature effect from the observation-based pCO$_2$, while normalizing the pCO$_2$ values to the long term mean SST, i.e., (<SST>)[19]

$$p\mathrm{CO}_2^{nt} = p\mathrm{CO}_2 \cdot \exp(\gamma_T \cdot (< \mathrm{SST} > - \mathrm{SST})) \tag{3}$$

Drivers of the seasonal pCO$_2$ difference trends To determine the potential drivers of the observed trends in the winter-minus-summer differences in pCO$_2$, we start with a decomposition of any variations in pCO$_2$ into their driving components, i.e., sea-surface temperature, SST, sea-surface salinity, S, dissolved inorganic carbon, DIC, and total alkalinity, Alk, thereby neglecting the very small contribution arising from variations in nutrients and other very minor drivers. Considering only the first-order terms of a Taylor expansion gives for any change in pCO$_2$, i.e., dpCO$_2$[6,18]:

$$\begin{aligned} dp\mathrm{CO}_2 \quad = \quad & \frac{\partial p\mathrm{CO}_2}{\partial \mathrm{SST}} \cdot d\mathrm{SST} + \frac{\partial p\mathrm{CO}_2}{\partial \mathrm{DIC}} \cdot d\mathrm{DIC} \\ & + \frac{\partial p\mathrm{CO}_2}{\partial \mathrm{Alk}} \cdot d\mathrm{Alk} + \frac{\partial p\mathrm{CO}_2}{\partial \mathrm{S}} \cdot d\mathrm{S} \end{aligned} \tag{4}$$

where the "d" denotes deviations of this property from some norm (with the assumption that d is small relative to mean value). Since variations in the freshwater input to the surface ocean affect not only salinity, but also DIC and Alk (with the latter two opposing each other), it is often more insightful to combine all terms affected by freshwater fluxes into one[31] leaving only the seasonal changes in salinity normalized DIC and Alk in the respective terms. The salinity normalization is given by sDIC = S/S_0 · DIC, sAlk = S/S_0· Alk, with S_0 representing the normalization salinity, here taken as the annual mean salinity. This gives:

$$\begin{aligned} dp\mathrm{CO_2} &= \frac{\partial p\mathrm{CO_2}}{\partial \mathrm{SST}} \cdot d\mathrm{SST} + \frac{\partial p\mathrm{CO_2}}{\partial \mathrm{DIC}} \cdot \mathrm{S/S_0} \cdot d\mathrm{sDIC} \\ &\quad + \frac{\partial p\mathrm{CO_2}}{\partial \mathrm{Alk}} \cdot \mathrm{S/S_0} \cdot d\mathrm{sAlk} + \frac{\partial p\mathrm{CO_2}}{\partial \mathrm{FW}} \cdot d\mathrm{FW} \end{aligned} \tag{5}$$

The partial derivatives with regard to any of the drivers, i.e., $\partial p\mathrm{CO_2}/\partial \mathrm{X}$ can be estimated from the $p\mathrm{CO_2}$ sensitivities, γ[6]. As we are considering deviations from the annual mean, S/S_0 is essentially equal to 1 and will be dropped subsequently. Taking advantage of this simplification, inserting these sensitivities and replacing "d" with the seasonal difference (Δ^{seas}) yields:

$$\begin{aligned} \Delta^{seas} p\mathrm{CO_2} &= \underbrace{\gamma_T \cdot p\mathrm{CO_2} \cdot \Delta^{seas}\mathrm{SST}}_{\text{sensitivity to temperature}} + \underbrace{\gamma_{DIC} \cdot \frac{p\mathrm{CO_2}}{\mathrm{DIC}} \cdot \Delta^{seas}\mathrm{sDIC}}_{\text{sensitivity to sDIC}} \\ &\quad + \underbrace{\gamma_{Alk} \cdot \frac{p\mathrm{CO_2}}{\mathrm{Alk}} \cdot \Delta^{seas}\mathrm{sAlk}}_{\text{sensitivity to sAlk}} + \underbrace{\gamma_{FW} \cdot \frac{p\mathrm{CO_2}}{\mathrm{FW}} \cdot \Delta^{seas}\mathrm{FW}}_{\text{sensitivity to freshwater}} \end{aligned} \tag{6}$$

where γ_T is the temperature sensitivity (see above), and where $\gamma_{DIC,Alk,FW}$ describe the dimensionless $p\mathrm{CO_2}$ sensitivities of the remaining terms, with γ_{DIC} usually referred to as the Revelle factor[6,32].

The first term of the right hand side of equation (6) represents the seasonal difference of the thermal component, while the remaining three terms together represent the seasonal difference of the non-thermal component. To simplify the further analysis, we take advantage of the observation that the contribution of sAlk

and FW to the seasonal cycle of $p\mathrm{CO}_2$ is generally much smaller than that of SST and sDIC[6]. This permits us to drop these terms subsequently. We expect this simplification to work well across the majority of the ocean basins, where the seasonal changes in sAlk are very small[33], but less so in high latitude and coastal ocean regions, where both the sAlk and the freshwater fluxes are larger. However, since we investigate zonal mean trends and no trends poleward of 65°N and 65°S, respectively, we expect our simplification to work very well. The non-thermal part of (6) thus reduces to a single component driven by sDIC:

$$\begin{aligned} \Delta^{seas} p\mathrm{CO}_2 &= \Delta^{seas} p\mathrm{CO}_2^{th} + \Delta^{seas} p\mathrm{CO}_2^{nt} \\ &\approx \gamma_T \cdot p\mathrm{CO}_2 \cdot \Delta^{seas}\mathrm{SST} + \gamma_{sDIC} \cdot \frac{p\mathrm{CO}_2}{\mathrm{DIC}} \cdot \Delta^{seas}\mathrm{sDIC} \end{aligned} \tag{7}$$

We next determine the temporal trends in these two components. Computing the temporal derivative of the seasonal difference we derive

$$\begin{aligned} \frac{d\Delta^{seas} p\mathrm{CO}_2}{dt} &= \gamma_T \cdot \frac{dp\mathrm{CO}_2}{dt} \cdot \Delta^{seas}\mathrm{SST} + \gamma_T \cdot p\mathrm{CO}_2 \cdot \frac{d\Delta^{seas}\mathrm{SST}}{dt} \\ &+ \frac{d\gamma_{DIC}}{dt} \cdot \frac{p\mathrm{CO}_2}{\mathrm{DIC}} \cdot \Delta^{seas}\mathrm{sDIC} \\ &+ \gamma_{DIC} \cdot \Delta^{seas}\mathrm{sDIC} \cdot \left(\frac{dp\mathrm{CO}_2/dt}{\mathrm{DIC}} - d\mathrm{DIC}/dt \cdot \frac{p\mathrm{CO}_2}{\mathrm{DIC}^2} \right) \\ &+ \gamma_{DIC} \cdot \frac{p\mathrm{CO}_2}{\mathrm{DIC}} \cdot \frac{d\Delta^{seas}\mathrm{sDIC}}{dt} \end{aligned} \tag{8}$$

Since $dp\mathrm{CO}_2$/dt and dDIC/dt are of similar magnitude (long term mean $dp\mathrm{CO}_2$/dt $\approx$ 1.5 μatm/yr), while $p\mathrm{CO}_2$/DIC $\ll$ 1, the second term inside the bracket is considerably smaller than the first term. Thus this equation can be simplified to:

$$\frac{d\Delta^{seas} p\mathrm{CO}_2}{dt} = \gamma_T \cdot \frac{dp\mathrm{CO}_2}{dt} \cdot \Delta^{seas}\mathrm{SST} + \gamma_T \cdot p\mathrm{CO}_2 \cdot \frac{d\Delta^{seas}\mathrm{SST}}{dt} \tag{9}$$

$$
\begin{aligned}
&+ \quad \frac{d\gamma_{DIC}}{dt} \cdot \frac{p\mathrm{CO}_2}{\mathrm{DIC}} \cdot \Delta^{seas}\mathrm{sDIC} + \gamma_{DIC} \cdot \frac{dp\mathrm{CO}_2}{dt} \frac{\Delta^{seas}\mathrm{sDIC}}{\mathrm{DIC}} \\
&+ \quad \gamma_{DIC} \cdot \frac{p\mathrm{CO}_2}{\mathrm{DIC}} \cdot \frac{d\Delta^{seas}\mathrm{sDIC}}{dt}
\end{aligned}
$$

and further

$$
\begin{aligned}
\frac{d\Delta^{seas}p\mathrm{CO}_2}{dt} \quad = \quad & \underbrace{\frac{dp\mathrm{CO}_2}{dt} \cdot (\gamma_T \cdot \Delta^{seas}\mathrm{SST} + \gamma_{DIC} \cdot \frac{\Delta^{seas}\mathrm{sDIC}}{\mathrm{DIC}})}_{\text{change in surface pCO}_2} \\
& + \quad \underbrace{\frac{d\gamma_{DIC}}{dt} \cdot \frac{p\mathrm{CO}_2}{\mathrm{DIC}} \cdot \Delta^{seas}\mathrm{sDIC}}_{\text{Revelle factor change}} \\
& + \quad \underbrace{\frac{d\Delta^{seas}\mathrm{sDIC}}{dt} \cdot \gamma_{DIC} \cdot \frac{p\mathrm{CO}_2}{\mathrm{DIC}} + \frac{d\Delta^{seas}\mathrm{SST}}{dt} \cdot \gamma_T \cdot p\mathrm{CO}_2}_{\text{seasonal difference change}}
\end{aligned}
\tag{10}
$$

Analyzing the terms that can drive a trend in the winter-minus-summer differences in $p\mathrm{CO}_2$ reveals that we have three sets of processes to consider. The first one is associated with the long-term change in surface ocean $p\mathrm{CO}_2$. This causes a trend simply by the seasonal variations in SST and sDIC acting on an increasing $p\mathrm{CO}_2$. The second process is a result of ocean acidification causing an increase in the Revelle factor. This term is, like the first one, directly tied to the increase in atmospheric CO_2 driving an uptake of anthropogenic CO_2 into the surface ocean. The third set of processes are associated with changes in the seasonal difference of the drivers, i.e., the $\Delta^{seas}\mathrm{X}$ terms owing e.g., to a change in ocean circulation/mixing or biological activity.

These terms can be grouped according whether they act on the thermal or non-thermal components. This gives for the thermal component:

$$
\frac{d\Delta^{seas}p\mathrm{CO}_2^{th}}{dt} = \underbrace{\gamma_T \cdot \frac{dp\mathrm{CO}_2}{dt} \cdot \Delta^{seas}\mathrm{SST}}_{\text{change in surface pCO}_2} + \underbrace{\gamma_T \cdot p\mathrm{CO}_2 \cdot \frac{d\Delta^{seas}\mathrm{SST}}{dt}}_{\text{seasonal difference change}} \tag{11}
$$

and for the non-thermal component, i.e., that driven by changes in sDIC:

$$\frac{d\Delta^{seas}pCO_2^{nt}}{dt} = \underbrace{\frac{d\gamma_{DIC}}{dt}\cdot\frac{pCO_2}{DIC}\cdot\Delta^{seas}sDIC}_{\text{Revelle factor change}} + \underbrace{\gamma_{DIC}\cdot\frac{dpCO_2}{dt}\frac{\Delta^{seas}sDIC}{DIC}}_{\text{change in surface } pCO_2} + \underbrace{\gamma_{DIC}\cdot\frac{pCO_2}{DIC}\cdot\frac{d\Delta^{seas}sDIC}{dt}}_{\text{seasonal difference change}} \quad (12)$$

Here we only quantify the impact of the first two sets of drivers, i.e., the surface ocean pCO_2 driven change and the change in the Revelle factor, as we currently lack good observations for the trends in $\Delta^{seas}X$ terms. Therefore, deviation between our observation-based CO_2 estimate and the theoretical framework can be partly explained by the third set of drivers (as well as by the neglected contributions from alkalinity and the freshwater balance).

We have already all the input data[34] to estimate the CO_2 effect for the thermal component (10), but we need to estimate the seasonal difference of sDIC required to estimate the CO_2 effect for the non-thermal component. We estimate this term, i.e., Δ^{seas} sDIC from pCO_2 using equation (8) and fields from the GLODAPv2 database[35,36]

$$\Delta^{seas}sDIC = (\Delta^{seas}pCO_2 - \gamma_T\cdot pCO_2\cdot\Delta^{seas}SST)\cdot\frac{DIC}{\gamma_{DIC}\cdot pCO_2} \quad (13)$$

and the change in the Revelle factor as[6]

$$\frac{d\gamma_{DIC}}{dt} = \frac{d\gamma_{DIC}}{dDIC}\cdot\frac{pCO_2/DIC}{\gamma_{DIC}}\cdot\frac{dpCO_2^{atm}}{dt} \quad (14)$$

using the dry air mixing ratio of atmospheric CO_2 (https://www.esrl.noaa.gov/gmd/ccgg/mbl/) with a long term mean $dpCO_2^{atm}$/dt of 1.7 μatm/yr, and by using the approximation[6]:

$$\gamma_{DIC} \approx \frac{3 \cdot \mathrm{Alk} \cdot \mathrm{DIC} - 2 \cdot \mathrm{DIC}^2}{(2\mathrm{DIC} - \mathrm{Alk}) \cdot (\mathrm{Alk} - \mathrm{DIC})} \tag{15}$$

we derive

$$\frac{d\gamma_{DIC}}{d\mathrm{DIC}} = \frac{\mathrm{Alk}^2 \cdot (4 \cdot \mathrm{DIC} - 3 \cdot \mathrm{Alk})}{(\mathrm{DIC} - \mathrm{Alk})^2 \cdot (2 \cdot \mathrm{DIC} - \mathrm{Alk})^2} \tag{16}$$

which yields the expected change in the non-thermal $p\mathrm{CO}_2$ seasonal difference in response to a change in Revelle factor plus the oceanic accumulation of anthropogenic CO_2. While the above approximation for the Revelle factor is far from being accurate, tests with a full model of the aqueous CO_2 system reveal that this approximation is reasonably close to the accurate results. We retain the approximation given its simplicity.

The results of equations 11 and 12 as well as their combination in equation 10 are displayed in Figure 4 of the main text as dashed green lines.

Uncertainty analysis Two main sources of uncertainty contribute to the uncertainty of the reported trend in the winter-minus-summer difference in surface ocean $p\mathrm{CO}_2$. The first source is associated with the uncertainty in the spatio-temporal interpolation of the observations, which directly affects the diagnosed magnitude of the seasonal cycle. The second source of uncertainty stems from the determination of the 34-year linear trend in the winter-minus-summer difference in the presence of interannual to decadal variations.

We use the surface ocean $p\mathrm{CO}_2$ residuals, computed as the difference between our interpolated $p\mathrm{CO}_2$ estimate minus the SOCATv4 gridded data[11] as the basis for computing the uncertainty for the winter-minus-summer difference. These residuals contain no significant temporal trends neither for the summer nor for the winter season (see supplement) and also do not depend on the data density. This permits us to assume that the error structure of our interpolated $p\mathrm{CO}_2$ is stationary and random in time, and thus not biasing the

determination of the linear trend. The substantial residuals (root mean squared error (RMSE) of a typical magnitude of 10-20 μatm) nevertheless imply a considerable uncertainty of the reconstructed seasonal cycle. But as we determine the mean seasonal cycle for large spatial regions, this uncertainty of the mean, i.e., the standard error (SE) goes down with one over the square root of the effective sample size N^{eff}, i.e.,

$$SE \approx \frac{RMSE}{\sqrt{N^{eff}}} \tag{17}$$

where N^{eff} represents the spatially decorrelated number of pCO$_2$ residuals per region. We calculate it from

$$N^{eff} \approx N \cdot \frac{1-r}{1+r} \tag{18}$$

where N is the total number of residuals per region and r is the lag 1 autocorrelation coefficient. We estimate r by randomly plotting the squared difference of the pCO$_2$ residuals as a function of haversine distance. We do this for five randomly chosen and non-repeating subsamples of 1000 residual pairs each, binning them into 300 km bins and calculating:

$$r = \frac{\sum_{d=1}^{n-1}(x_d - < x_{1:n-1} >)(x_{d+1} - < x_{2:n} >)}{(\sum_{d=1}^{n-1}(x_d - < x_{1:n-1} >)^2)^{1/2}(\sum_{d=1}^{n-1}(x_{d+1} - < x_{2:n} >)^2)^{1/2}} \tag{19}$$

where x is the residual difference squared of two locations separated by the distance d and $< x >$ represents the average squared distance. The standard error is computed for each analysis region.

To determine the uncertainty of the linear trend in the annual winter-minus-summer differences for each of the regions, we take the 95% confidence interval reported in the output of the least-squares regression fit in matlab. We test if the trends are significantly different from 0 using standard t-statistics.

Data availability Our pCO_2 analysis is based on measurements extracted from the Surface Ocean CO_2 Atlas (SOCATv4)[11] that is freely available via: https://www.socat.info. The neural network based interpolated sea surface pCO_2 product used in this study is freely accessible at the National Centers for Environmental Information via:

https://www.nodc.noaa.gov/ocads/oceans/SPCO2_1982_2015_ETH_SOM_FFN.html We further use bottle data from the Bermuda Atlantic Timeseries station (BATS)[16] and from the closely located Station S[15] to compute the surface ocean pCO_2 at these two sites near Bermuda. These data are available online via http://bats.bios.edu. For the Hawaiian Ocean Timeseries station (HOT)[14], we extracted the already computed pCO_2 data from http://hahana.soest.hawaii.edu/hot/products/HOT_surface_CO2.txt. Additionally, for our calculations, we use the gridded product from the Global Data Analysis Project version2 (GLODAPv2)[35,37] which can be freely accessed from https://www.nodc.noaa.gov/ocads/oceans/GLODAPv2/ and NOAA-ESRL marine boundary layer reference dry air mixing ratio of atmospheric CO_2 freely accessed from https://www.esrl.noaa.gov/gmd/ccgg/mbl/. Lastly we use the NOAA Optimum Interpolation (OI) version 2[34] sea surface temperature product, freely accisible via https://www.esrl.noaa.gov/psd/data/gridded/data.noaa.oisst.v2.html, for our calculations.

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Figure 1 Trends in the seasonal difference of sea surface pCO$_2$ from 1982 through 2015. (a) 10°N -40°N, (b) 40°N -65°N (c) 10°S -40°S and (d) 40°S -65°S. Trends are derived from the updated version of an observation-based sea surface pCO$_2$ product[13]. Black markers represent the seasonal maximum minus seasonal minimum for each year and the solid regression line represents the results of a linear least-squares regression. The shaded area represents the uncertainty of the interpolated pCO$_2$ product. The slope of the line represents the trends in seasonal variations, i.e., the trend in the seasonal maximum minus the seasonal minimum including its 95% confidence interval.

Figure 2 The changing seasonal sea surface pCO$_2$ cycle. (a) 10°N -40°N, (b) 40°N -65°N (c) 10°S -40°S and (d) 40°S -65°S. Each panel consists of a comparison of the mean seasonal cycle from 1985-1989 (dashed line) and 2010-2014 (solid line) including shaded uncertainty estimate. The mean seasonal cycles for each respective period are repeated and those of the southern hemisphere are shifted by 6 months to align the seasons. (e) The climatological mean winter-minus-summer difference for each 1°×1° pixel. Positive winter-minus-summer differences are marked in red, whereas negative winter-minus-summer differences are marked in blue.

Figure 3 Separation of the 5-year mean seasonal cycle of pCO$_2$ into its thermal and non-thermal components. (a) between 10°N -40°N and (b) between 40°S -65°S. Shown are the data for the period 1985-1989 (dashed lines) and 2010-2014 (solid lines). While in (a) the non-thermal component increases the seasonal winter-minus-summer difference

(marked in red with a plus sign highlighting the positive transition from winter maximum to summer minimum), the opposing thermal signal is stronger leading to an increase in the summer-minus-winter difference (marked in blue with a minus sign highlighting the negative transition from winter minimum to summer maximum). Vice versa, in (b) the non-thermal component dominates over the thermal component.

Figure 4 Regional and zonal mean trends in the winter-minus-summer difference. (a) total (black), (b) thermal (blue) and (c) non-thermal (red) seasonal $p\mathrm{CO}_2$ cycles visualized both geographically and as zonal means. Negative trends highlight an increasing summer maximum and decreasing winter minimum, whereas positive trends highlight increasing winter maxima and decreasing summer minima. The dashed green lines represent the expected increase based on our thermodynamic consideration considering only the effect of the increase in surface ocean CO_2 and changes in Revelle factor (see text). Stars further indicate the observation-based (gray, red, blue) winter-minus-summer trends from timeseries stations at Bermuda and Hawaii. Note that the latter are trends at a single location, while the trends shown based on the neural network based $p\mathrm{CO}_2$ analyses represent basin-scale or global zonal mean trends.