

## Author's Response

We thank referee#3 for reviewing the paper and for providing helpful comments which have improved the paper. We address point by point (answers in red) the concerns of the referee.

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### Major issues:

The authors calculate pCO<sub>2</sub> from DIC and TA data, which results in a nonequilibrium pCO<sub>2</sub>. This is then converted to a standard temperature and calculated back to TA and DIC assuming that pCO<sub>2</sub> is back to its initial value. As a major part of the pCO<sub>2</sub> disequilibrium in the ocean is associated to temperature changes wouldn't it be more useful to assume equilibrium with the atmosphere (at in-situ temperature as well as at standard temperature) when calculating the temperature effect on DIC?

We understand that the major part of the pCO<sub>2</sub> disequilibrium in the ocean is associated with temperature changes, with disequilibria able to persist for many months (e.g. Körtzinger et al. 2008) because of the long air-sea gas exchange equilibrium timescale of CO<sub>2</sub>. Previous studies such as Takahashi et al. (2014) revealed that in the real ocean, exact CO<sub>2</sub> equilibrium between the sea surface and the atmosphere above is hardly ever reached. Thus, the assumption of air-sea CO<sub>2</sub> equilibrium is a theoretical concept but is not often a practical reality. In fact we previously calculated temperature effects assuming air-sea equilibrium and obtained very similar results (the average absolute difference between temperature effects is about 20 μmol kg<sup>-1</sup>). However, for the reasons just stated, we prefer to calculate the effects from the starting point of the observed disequilibria.

The two major weaknesses raised during the first review iteration are still persisting, the normalization of DIC without a freshwater component and the disregard of seasonality.

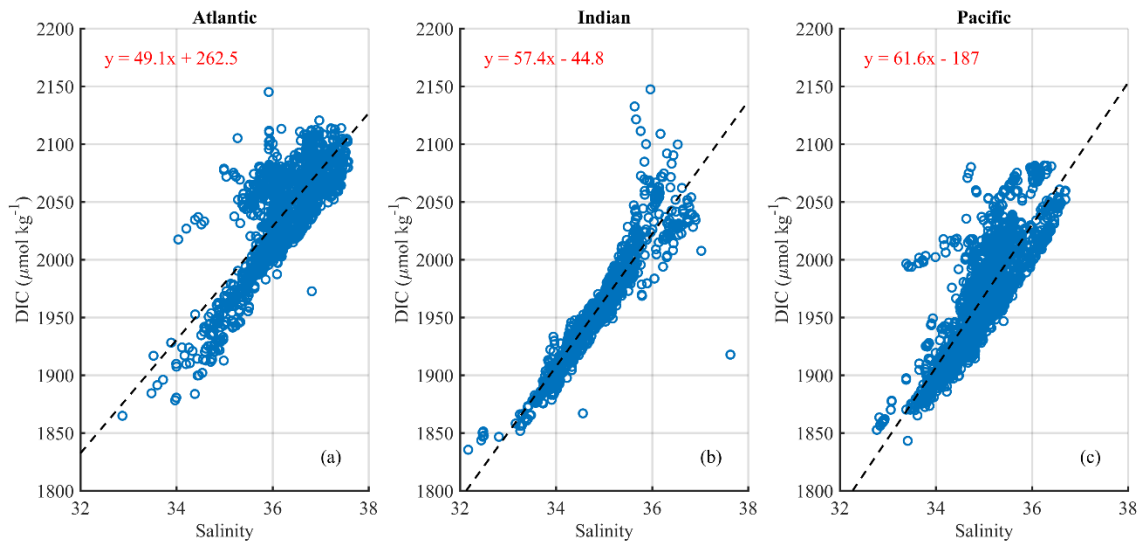
I am not convinced that excluding areas strongly influenced by riverine input is enough to solve the problem with the DIC normalization. Fitting the alkalinity against the salinity gives also in the open ocean a non-zero intercept that can vary from region to region. With the GLODAP v2 dataset there is an excellent database to calculate this intercept and its regional variations. It has at least to be discussed what influence this would have on the latitudinal gradients.

We agree with the referee that more details about salinity normalization should be stated. As a supplement to our response to the first referee (<https://www.biogeosciences-discuss.net/bg-2018-376/bg-2018-376-AC1-supplement.pdf>), which mainly focused on neglecting the influence of upwelling on DIC salinity normalization, we quantify here the impact of the 'non-zero intercept' issue raised by the referee.

Figure R1 shows the relationship between surface DIC and salinity. We selected data between 30°S and 30°N (i.e., the oligotrophic surface oceans) in order to avoid the perturbations of upwelling (this has been discussed in the response letter to

Referee#1, see link above) and biological activities on surface DIC. By fitting surface DIC against salinity, a non-zero intercept was found for each of the ocean basins, ranging from  $-187 \mu\text{mol kg}^{-1}$  (Pacific Ocean) to  $262 \mu\text{mol kg}^{-1}$  (Atlantic). By substituting the values into Equation  $n\text{DIC} = \frac{\text{DIC}^{\text{meas}} - \text{DIC}^{\text{S}=0}}{S^{\text{meas}}} \cdot S^{\text{ref}} + \text{DIC}^{\text{S}=0}$  (Friis et al., 2003), which can be further converted into  $n\text{DIC} = \frac{\text{DIC}^{\text{meas}}}{S^{\text{meas}}} \cdot S^{\text{ref}} + \text{DIC}^{\text{S}=0} \cdot \left(1 - \frac{S^{\text{ref}}}{S^{\text{meas}}}\right)$ , we found that the discrepancy (i.e., the term  $\text{DIC}^{\text{S}=0} \cdot \left(1 - \frac{S^{\text{ref}}}{S^{\text{meas}}}\right)$ ) between the Friis et al. (2013) method and our calculation (Equation 5 in the manuscript) only accounts for a difference ranging from  $-5 \mu\text{mol kg}^{-1}$  to  $7 \mu\text{mol kg}^{-1}$ , which is of similar magnitude to the uncertainty in DIC measurement and also much smaller than the phenomenon of interest (about  $200 \mu\text{mol kg}^{-1}$ ).

We have added some texts in Section 2.2 to discuss this influence.



10 Figure R1. Relationship between sea surface DIC and salinity in the Atlantic, Indian, and Pacific Oceans. The black dashed lines are best-fit linear regression lines.

Regarding the seasonality I do understand that there is not enough data in the Southern Ocean to resolve and discuss seasonalities. But nevertheless these influences need to be discussed.

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We agree that seasonality in surface DIC and nDIC at high latitudes must result in seasonal variation of the nDIC latitudinal gradient. We have added a new section 4.1.4 to discuss the issue raised by the referee.

The salinity normalization of phosphate, for example, can produce relatively high residual phosphate and thus ‘unused DIC’ during summer in regions with low salinities although these might experience phosphate limitation during summer.

20

We calculated ‘unused DIC’ based on the phosphate concentration without salinity normalization.

Also I am missing a notice that using phosphate concentrations as tracer for the influence of primary production is a simplification. C:P ratios are highly variable, not only, as mentioned, regionally, but also seasonally. Under phosphate limitation phytoplankton is still able to fix carbon, and release it as DOC.

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We thank the referee for pointing out the seasonal variation of marine C:N:P stoichiometry (e.g., Frigstad et al., 2011). However, since the seasonal variation of C:P is much smaller than the latitudinal variation of C:P, which we care about most in this context, and since there have been no studies investigating the seasonal variation of C:P over the global ocean, we decided to only consider its latitudinal variation in this study. We have modified the text in Section 2.4.2 to acknowledge the seasonal variation of C:P.

10

As a last major point, the manuscript is partly very difficult to read. The discussion of effects on DIC and nDIC is sometimes confusing and the reader might get lost which of both is discussed and why.

15 An English-speaking co-author has gone through the manuscript again in an attempt to improve readability. We have modified the text (e.g., revisions to the Introduction, Section 4.1 and Section 4.2) to try and make it as simple and understandable as possible given the complexity of the topic. We have added a short paragraph at the start of Section 4.1 to make it clear that the calculated effects are all based on nDIC.

20 **Minor issues:**

p. 10 l. 21: Here  $\text{DIC}_{\text{obs}}$  and  $\text{nDIC}_{\text{obs}}$  are discussed, right?

Yes, Section 3.1 described the distributions of  $\text{DIC}_{\text{obs}}$  and  $\text{nDIC}_{\text{obs}}$ . The heading of this section has been modified to “Spatial distributions of observed DIC and nDIC”.

25

p. 11 l. 12: Rephrase this sentence. It reads as if you have data at 70°S in the northern Atlantic.

Changed.

30 p. 12 l. 13: This part needs some changes. Most of the DIC variability is in fact not caused by changes in alkalinity but by dilution and evaporation and therefore is not existent anymore when discussing the nDIC. Alkalinity and DIC are just influenced in a similar manner.

We agree that both mechanisms (direct and indirect effects of evaporation and precipitation) must operate but argue that the latter must win out in the end. One of the novel contributions of our paper, we believe, is that the long-term (> 1 or 2 years) consequences of upwelling for DIC concentrations are dictated by the upwelling-induced changes to TA, rather than by the immediate upwelling-induced changes to nutrient or DIC concentrations (Section 4.2). By the same logic (Figure 6), changes to TA from other causes, including from evaporation or precipitation, should also eventually, because of gas exchange, determine changes to equilibrium DIC. Even in an imaginary scenario where evaporation and precipitation had no direct effect on the DIC concentration, but only on the TA concentration, then the processes would still end up altering the DIC concentration through the causal sequence: evaporation or precipitation → TA change → pCO<sub>2</sub> change → generation of air-sea CO<sub>2</sub> disequilibrium → air-sea exchange of CO<sub>2</sub> → DIC change until air-sea equilibrium is re-established. Figure 6 has been modified to acknowledge that evaporation / precipitation also has a direct impact on DIC.

p. 12 l. 25: TA and salinity are only in the open ocean highly correlated.

The phrase “surface ocean” has been changed to “surface open ocean” (as described in the methods section, our study is restricted to open ocean locations where the seafloor depth is greater than 200m).

p. 13 l. 9: nutrients, nDIC and nTA?

Changed.

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p. 13 l. 10: delete ‘then’

Changed.

25 p. 13 l. 30: change of nTA, or change of TA relative to the DIC changes.

Arrow ⑤ in Figure 6 is the difference between the original, pre-upwelling DIC and the final DIC after the long-term alkalinity effect has run to completion. The size of this difference is dictated by the TA (not nTA) change brought about by upwelling. Its magnitude is independent of the immediate DIC change brought about by upwelling (arrow ① in Figure 6). The text is correct as it stands and has not been changed.

30 p. 14 l. 22: this should also be a function of the chemical composition: how low is the iron concentration in the upwelled water in comparison to the other nutrients.

We relate the long-term effect of upwelling only to the surface TA and temperature, which together determine the surface DIC if the system is to be in equilibrium with the atmosphere. The iron concentration in the upwelled water was not considered in the manuscript because we used the changes in surface phosphate to calculate the effect of biological removal (i.e., if there was a complete absence of iron in the upwelled water, then biological uptake would not take place and phosphate concentrations would not alter from the original value).

p. 14 l. 23: delete ‘.’

Changed.

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Figure 1: increase the resolution of this picture. The description at the colour axis only says DIC.

Figure 1 will have the required resolution when published. In Word and pdf files the figures are automatically compressed, making the resolution of the figures seem not good enough. When the figures are taken from separate uploaded files, as they are for publication, then the issue does not occur. The colour axis description has been changed to “DIC or nDIC”.

15

Figure 2: Here a direct influence from evaporation & precipitation to DIC is missing. Also, in the description of the figure it is written about both DIC and nDIC. For which of these two is the diagram?

We added “also influence DIC” to the arrow from E. vs P. to TA. The diagram illustrates processes affecting DIC and we have changed the text to say this.

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Figure 5: The resolution of the picture should be improved. The map should be deleted from this picture. Show the location of the transects in Figure 3 instead. I think all figures should follow a common design when it comes to setup and fonts. Please change the design of this figure match the other figures. I don’t understand the use of the black contour lines. Either, choose them in a way that the cover the entire data range or delete them.

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Figure setup and fonts were changed. The black contours highlighted the values of the selected variable along the upwelling isopycnal but we have now removed the black contours to keep the figure simple and avoid confusing readers. However, we decided to leave the map in Figure 5 for two reasons: (1) adding the transects to Figure 3 will cover up the information which shows the sampling year; (2) we selected the three transects only for the calculation of upwelling effects, therefore there is no need to show the transects before Section 2.4.2.

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Figure 8: Are these the same transects as in figure 5?

Yes. All the associated calculations of upwelling effects are based on the three transects. We added one sentence in Section 3.3 as well as in the caption of Fig. 8 to state this.

Figure 9: Change the rotation of the longitudinal label to  $0^\circ$ .

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Changed.

## References

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**A marked-up manuscript version:**

# What drives the latitudinal gradient in open ocean surface dissolved inorganic carbon concentration?

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**Abstract.** Previous work has not led to a clear understanding of the causes of spatial pattern in global surface ocean DIC, which generally increases polewards. Here, we revisit this question by investigating the drivers of observed latitudinal gradients in surface salinity-normalized DIC (nDIC) using the Global Ocean Data Analysis Project Version 2 (GLODAPv2) database. We used the database to test three different hypotheses for the driver producing the observed increase in surface nDIC from low to high latitudes. These are: (1) sea surface temperature, through its effect on the CO<sub>2</sub> system equilibrium constants, (2) salinity-related total alkalinity (TA), and (3) high latitude upwelling of DIC- and TA-rich deep waters. We find that temperature and upwelling are the two major drivers. TA effects generally oppose the observed gradient, except where higher values are introduced in upwelled waters. Temperature-driven effects explains the majority of the surface nDIC latitudinal gradient (182 ~~out of~~ the 223  $\mu\text{mol kg}^{-1}$  increase from the tropics to the high-latitude Southern Ocean). Upwelling, which has not previously been considered as a major driver, additionally drives a substantial latitudinal gradient. Its immediate impact, prior to any induced air-sea CO<sub>2</sub> exchange, is to raise Southern Ocean nDIC by 20 220  $\mu\text{mol kg}^{-1}$  above the average low latitude value. However, this immediate effect is transitory. The long-term impact of upwelling (brought about by increasing TA), which would persist even if gas exchange were to return the surface ocean to the same CO<sub>2</sub> as without upwelling, is to increase nDIC by 74  $\mu\text{mol kg}^{-1}$  above the low latitude average.

## 1 Introduction

The ocean absorbs about one quarter of the anthropogenic CO<sub>2</sub> emitted every year (Le Quéré et al., 2018). It is the largest non-geological carbon reservoir (~38000 Gt C; Falkowski et al., 2000) ~~(Falkowski et al., 2000)~~, containing 50 times as much carbon as the pre-industrial atmosphere, and thereby plays an important role in modulating/modulates the Earth's climate system. Approximately 97% of the carbon in the oceanic carbon pool exists is in the form of dissolved inorganic



carbon (DIC) (~~Falkowski et al., 2000~~), which is the total concentration of ~~defined as the sum of the concentrations of~~ aqueous CO<sub>2</sub>, and bicarbonate and carbonate ions:

$$\text{DIC} = [\text{CO}_2^*] + [\text{HCO}_3^-] + [\text{CO}_3^{2-}] \quad (1)$$

where [CO<sub>2</sub>\*] refers to the sum of aqueous CO<sub>2</sub> and ~~true undissociated~~ carbonic acid (H<sub>2</sub>CO<sub>3</sub>), with the latter negligible (Zeebe and Wolf-Gladrow, 2001).

Understanding what controls the distribution of oceanic DIC ~~in seawater~~ is essential for quantifying understanding anthropogenic CO<sub>2</sub> invasion (e.g., Gruber, 1998; Humphreys et al., 2016; Lee et al., 2003; Sabine et al., 1999; Sabine et al., 2002; Vázquez-Rodríguez et al., 2009) and consequent ocean acidification (e.g., Doney et al., 2009; Orr et al., 2005).

~~Given that the surface ocean is where~~ As most marine organisms live in the sunlit surface ocean, ~~and that it is the part of the ocean that exchanges~~ where CO<sub>2</sub> exchange with the atmosphere happens, the controls on surface ocean DIC particularly merit investigation.

~~Many previous studies focused on the vertical rather than the horizontal distribution of DIC, aiming to understand the relative contributions of the different “carbon pumps” in controlling DIC variations throughout the water column (i.e., solubility pump, soft tissue pump, and carbonate pump, see details in Cameron et al., 2005; Gruber and Sarmiento, 2002; Toggweiler et al., 2003a; Toggweiler et al., 2003b). However, the solubility pump is so named because it is based on an assumption about the horizontal distribution of surface DIC—that DIC is high where new deep waters form at high latitudes, because of the effect of temperature on CO<sub>2</sub> solubility. Many previous studies focused on the vertical, rather than latitudinal, distribution of DIC. They investigated the contributions of the different “carbon pumps” – solubility pump, soft tissue pump, and carbonate pump (Cameron et al., 2005; Gruber and Sarmiento, 2002; Toggweiler et al., 2003a; Toggweiler et al., 2003b) – to the pattern of DIC with depth. The solubility pump is based on the assumption that, at high latitudes where deep waters form, DIC is high because the low water temperature increases CO<sub>2</sub> solubility. Lee et al. (2000) used this principle to predict salinity-normalized DIC (nDIC) from empirical functions of sea surface temperature and nitrate that varied seasonally and geographically.~~

~~Previous work on the horizontal distribution of surface DIC included development of an algorithm to approximately reproduce the global surface DIC distribution (Lee et al., 2000), in which salinity-normalized DIC (nDIC) was predicted from empirically derived functions of sea surface temperature and nitrate that varied seasonally and geographically. Key et al. (2004) depicted the global distribution of surface DIC based on using an earlier version (GLODAPv1) of the dataset than we used herein this study, noting that the surface DIC pattern is overall more similar to nutrients (including in the Southern Ocean, where both DIC and nutrients are enriched) than to salinity; – unlike total alkalinity (TA), whose pattern more closely resembles that of salinity (Fry et al., 2015). Using the data from the new GLODAPv2 database (Key et al., 2015; Olsen et al., 2016), surface DIC is confirmed here to have its highest values at high latitudes, like nutrients, and to~~

reach its lowest values ~~in the at low latitudes equatorial zone~~ in each basin (Fig. 1a; more details in Sect. 3.1). Earlier studies (Lee et al., 2000; Toggweiler et al., 2003a; Williams and Follows, 2011, Sect. 6.3 “What controls DIC in the surface ocean?”) suggested that temperature is of primary importance in regulating surface DIC (e.g., Lee et al., 2000; Toggweiler et al., 2003a; Williams and Follows, 2011; Humphreys, 2017); ~~under~~ Under this assumption, surface waters in cool regions at high latitudes should hold more DIC than surface waters in the warm regions at low latitudes. ~~(Williams and Follows, 2011, Toggweiler et al., 2003a). Temperature may also drive latitudinal differences in carbonate ion concentration and surface aragonite saturation state ( $\Omega_{\text{arag}}$ ) (Jiang et al., 2015; Orr et al., 2005), through its effect on CO<sub>2</sub> solubility and CO<sub>2</sub> system thermodynamics.~~

Unlike previous studies, Williams and Follows (2011) argued that another variable also exerts a secondary control on the surface DIC distribution: ~~at a given DIC, TA sets the equilibrium capacity for influences the~~ seawater to hold DIC in solution partial pressure of carbon dioxide (see also Omta et al., 2011; Humphreys et al., 2018), so higher surface TA values may lead to higher DIC and therefore air-sea gas exchange. Takahashi et al. (2014) ~~subsequently~~ explored the seasonal ~~climatological~~ distributions of climatological surface DIC using seawater  $p\text{CO}_2$  from the Lamont Doherty Earth Observatory (LDEO) database and TA estimated from ~~sea surface~~ salinity, qualitatively attributing seasonal differences (on a regional scale) to the greater upward mixing of high-CO<sub>2</sub> deep waters in winter and summer biological carbon drawdown. ~~Their study pointed out~~ They noted the great potential ~~of for~~ upwelling to alter surface DIC, but ~~it was~~ focused ~~more on controls on~~ DIC seasonality rather than ~~DIC its~~ spatial variability. In recent years, the global surface DIC database has ~~been~~ greatly expanded, culminating ~~now~~ in GLODAPv2 (Key et al., 2015; Olsen et al., 2016), but the ~~controls producing drivers of~~ the global spatial surface DIC distribution have not ~~yet~~ been quantitatively reassessed.

~~Many~~ The processes ~~are known to that~~ influence the ~~sea surface~~ distribution of surface DIC at the local scale. They can be divided ~~overall~~ into those which change DIC ~~concentration~~ by direct addition or removal (e.g., 1-3 of the following), and those which affect DIC indirectly. (e.g., 4-6 of the following) The direct processes include: (1) biological carbon assimilation during primary production and releasing during remineralization (Bozec et al., 2006; Clargo et al., 2015; Toggweiler et al., 2003b; Yasunaka et al., 2013); (2) transport of DIC-rich deep waters into the surface layer (Jiang et al., 2013; Lee et al., 2000); and (3) production and export of CaCO<sub>3</sub>; Indirect processes include: (4) seawater dilution ~~or~~ concentration due to precipitation ~~or~~ evaporation (Friis et al., 2003); (5) warming ~~or and~~ cooling, which alters ~~the~~ CO<sub>2</sub> solubility and induces air-sea gas exchange that acts to reduce air-sea CO<sub>2</sub> disequilibrium (Bozec et al., 2006; Toggweiler et al., 2003a; Williams and Follows, 2011); and (6) the above processes (1-4) through their impact on TA ~~or~~ - if high/low TA values are not matched by high/low DIC values then the resulting low/high seawater  $p\text{CO}_2$  stimulates CO<sub>2</sub> ingassing/outgassing until DIC matches TA (Humphreys et al., 2018, 2017). The effects of equilibrium processes (the effects through temperature and upwelled TA) change the surface ocean DIC at which air-sea CO<sub>2</sub> equilibrium occurs, so

these effects can persist beyond the air-sea CO<sub>2</sub> equilibrium timescale (months to a year; Jones et al., 2014). The effects of disequilibrium processes, such as direct DIC supply from upwelling, and biological uptake of DIC in response to upwelled nutrients (principally iron; Moore et al., 2016) can persist no longer than the CO<sub>2</sub> equilibrium timescale.

Our study ~~builds on~~ ~~differs from~~ previous work in several ways. Firstly, whereas many previous studies looked to understand the ~~vertical~~ DIC distribution, ~~here the our~~ target is to understand the ~~horizontal~~ ~~latitudinal~~ surface DIC distribution. Secondly, we ~~look to~~ identify the most important ~~processes~~, not just ~~the~~ ~~variables~~, driving the surface DIC distribution (Fig. 2). ~~Third, Another difference from previous studies is our we~~ use of a much larger observational global dataset – GLODAPv2.

We evaluated ~~the following~~ three main hypotheses as to which processes cause the increase in surface DIC and nDIC from low to high latitudes (Fig. 1):

- (1) latitudinal variation of solar heating via its effect on sea surface temperature, and hence CO<sub>2</sub> solubility;
- (2) evaporation and precipitation, through their effects on TA; and
- (3) upwelling and winter entrainment through the introduction of DIC- and TA-rich deep waters to the (sub)polar surface oceans, when coupled with iron limitation of biological uptake of DIC.

It is easier to constrain the dynamics of upwelling and quantify its impact on surface DIC in the Southern Ocean (where upwelling has been more comprehensively studied, e.g., Marshall and Speer, 2012; Morrison et al., 2015) than in the subarctic North Atlantic and North Pacific Oceans (where upwards transport occurs via deep mixing in the winter, combined with upwelling in the North Pacific). The Southern Ocean also plays a crucial role in the global overturning circulation (e.g., Marshall and Speer, 2012), and the global carbon cycle (Landschützer et al., 2015; Mikaloff-Fletcher, 2015). Therefore, we focused on the Southern Ocean for the evaluation of the third hypothesis. A novel conclusion of this study is that upwelling, whose global significance has previously been overlooked, is very important in shaping the spatial distribution of surface ocean DIC, in part because upwelling of TA changes equilibrium DIC.

~~The processes that we evaluated can be separated into two categories: equilibrium and disequilibrium processes (Fig. 2). The effects of equilibrium processes (the effects through temperature and upwelled TA) change the surface ocean DIC at which air sea CO<sub>2</sub> equilibrium would occur, and those effects can therefore persist beyond the air sea CO<sub>2</sub> equilibrium timescale (months to year, Jones et al., 2014). The effects of disequilibrium processes (the effects through direct supply of DIC from upwelling and the effects through biological uptake of DIC in response to upwelled nutrients (principally iron, Moore et al., 2016)), on the other hand, are likely to persist only over timescales shorter than the CO<sub>2</sub> equilibrium timescale. A novel conclusion of this study is that upwelling, whose global significance has previously been overlooked, is very~~

~~important in shaping the spatial distribution of surface ocean DIC, in part because upwelling of TA changes equilibrium DIC.~~

## 2 Methods

~~Data for this study were taken~~ We used data from GLODAPv2 (<https://www.node.noaa.gov/ocads/oceans/GLODAPv2/>) (Key et al., 2015; Olsen et al., 2016). This ~~product compilation contains~~ includes data from over 700 cruises conducted during the years ~~from~~ 1972- to 2013, with about a third collected since 2003, a large fraction (~~~34%~~) having taken place during the period 2003-2013. These data have ~~undergone~~ been subject to secondary quality control and ~~have been adjusted for consistency~~ subsequent adjustments (Key et al., 2015; Lauvset and Tanhua, 2015; Olsen et al., 2016). ~~We define the surface ocean as waters shallower than 30 m at latitudes greater than 30°, and shallower than 20 m at latitudes less than 30° (following e.g., Fry et al., 2015; Lee et al., 2006). Only open ocean data (water depth > 200 m) were included in this study (Fig. 3).~~

### 2.1. Data processing

~~We define the “surface” ocean as the uppermost waters shallower than 30 m at latitudes greater than 30°, and shallower than 20 m at latitudes less than 30° (following e.g., Fry et al., 2015; Lee et al., 2006). Only open ocean data (water depth > 200 m) were included in this study (Fig. 3).~~

We excluded regions ~~subjected to significant perturbations from~~ perturbed by river inputs in order to remove confounding factors affecting the latitudinal distributions of DIC and nDIC on smaller length scales than being investigated here. We excluded ~~all data from~~ the Arctic Ocean (> 65° N) (Fig. 3) because ~~they are~~ it is heavily influenced by river inputs (Fry et al., 2015; Jiang et al., 2014), all data from the Mediterranean Sea and the Red Sea because of its very high salinity (Jiang et al., 2014), as well as some data (those where S is less than 34) from other ocean areas: the Amazon River plume in the North Atlantic (5° N-10° N, > 45° W), the Ganges/Brahmaputra plume in the Bay of Bengal (> 5° N, 80-94° E) (both Fry et al., 2015) and the western North Atlantic margins (Cai et al., 2010). We also excluded ~~relatively small~~ low-latitude ocean areas affected by upwelling (*i.e.*, the eastern equatorial Pacific and northern Californian upwelling regions).

Because atmospheric CO<sub>2</sub> increased during the ~~time period~~ that the GLODAPv2 data was collected (1972-2013), DIC has also increased ~~in response~~ in surface ~~ocean~~ waters (Bates et al., 2014). To prevent ~~temporal~~ DIC ~~time~~ trends from generating artificial spatial variability, we normalized surface DIC to a reference year of 2005, ~~by~~ We assumed ~~assuming~~ that sea surface ~~seawater pCO<sub>2</sub> changes tracks~~ atmospheric ~~pCO<sub>2</sub> changes~~ (Feely, 2008; see also CO<sub>2</sub> Time Series in the North

Pacific at <https://pmel.noaa.gov/co2/file/CO2+time+series>). We first calculated the change ~~of in~~ atmospheric mole fraction of CO<sub>2</sub> ( $x\text{CO}_{2,\text{air}}$ ) from the reference year 2005:

$$\Delta x\text{CO}_{2,\text{air}} = x\text{CO}_{2,\text{air}}^t - x\text{CO}_{2,\text{air}}^{2005} \quad (2)$$

where the superscript “t” and “2005” refer to year, and the globally averaged atmospheric xCO<sub>2</sub> data can be found at <https://www.esrl.noaa.gov/gmd/ccgg/trends/> (neither spatial nor seasonal variability in atmospheric CO<sub>2</sub> is taken into account).

Then we converted  $\Delta x\text{CO}_{2,\text{air}}$  into  $\Delta p\text{CO}_{2,\text{air}}$  (Takahashi et al., 2009) just above the sea surface, using in-situ humidity data. It is then assumed that  $\Delta p\text{CO}_{2,\text{sw}}$ , representing the change of sea surface pCO<sub>2</sub> relative to the year 2005, is equal to  $\Delta p\text{CO}_{2,\text{air}}$ .

10 Therefore the sea surface pCO<sub>2</sub> normalized to year 2005 was calculated as:

$$p\text{CO}_{2,\text{sw}}^{2005} = p\text{CO}_{2,\text{sw}}^t - \Delta p\text{CO}_{2,\text{sw}} \quad (3)$$

where  $p\text{CO}_{2,\text{sw}}^t$  was calculated from in-situ DIC, TA, temperature and salinity ~~using through~~ CO<sub>2</sub>SYS [v1.1](#) (van Heuven et al., 2011; dissociation constants used are described in Sect. 2.3).

Since the anthropogenic CO<sub>2</sub> perturbation does not change TA, DIC normalized to ~~the year~~ 2005 was calculated with inputs   
15 of in-situ TA and  $p\text{CO}_{2,\text{sw}}^{2005}$  using CO<sub>2</sub>SYS (van Heuven et al., 2011):

$$\text{DIC}^{2005} = f(T_{\text{in-situ}}, S_{\text{in-situ}}, \text{TA}_{\text{in-situ}}, p\text{CO}_{2,\text{sw}}^{2005}) \quad (4)$$

The concentration of DIC hereinafter refers to ~~the~~ DIC normalized to ~~the~~ year 2005.

## 2.2 Salinity normalization

Salinity normalization was used to correct for the influence of precipitation and evaporation in the open ocean (Postma,   
20 1964). Data were normalized to a reference salinity of 35 using a standard procedure:

$$nX = X_{\text{obs}} \times 35/S_{\text{obs}} \quad (5)$$

where nX refers to the normalized variable, X<sub>obs</sub> is the observed value of the variable, and S<sub>obs</sub> is the observed salinity.

~~It should be acknowledged that straightforward salinity normalization (Eq. 5) We acknowledge that this approach (Eq. 5) can create artificial variance in DIC distribution (Friis et al., 2003) because it ignores the influences of riverine input and upwelling from below the lysocline. To this end, we excluded the major regions affected by river inputs (see text in Sect. 2.1). In addition, we found that the influence of upwelling from below the lysocline on salinity normalization of DIC is negligible (i.e., it can only account for up to 4 μmol kg<sup>-1</sup> artificial change in DIC) compared with the DIC latitudinal gradient of about 200 μmol kg<sup>-1</sup> that we seek to explain in this study. We avoided the riverine problem by excluding affected regions (Section 2.1). We found that correcting for the “non-zero intercept” of DIC/salinity plots in different ocean~~

basins has negligible influence on salinity normalization of DIC, accounting for at most  $7 \mu\text{mol kg}^{-1}$  change in DIC; upwelling from below the thermocline also has negligible influence, accounting for at most  $4 \mu\text{mol kg}^{-1}$  change in DIC. These are small compared to the DIC latitudinal gradient of about  $200 \mu\text{mol kg}^{-1}$  that we investigate here.

## 5 2.3 Carbonate chemistry

Carbonate system variables were calculated from DIC and TA using version 1.1 of CO<sub>2</sub>SYS for MATLAB (van Heuven et al., 2011). The dissociation constants for carbonic acid and bisulfate were taken from Lueker et al. (2000) and Dickson (1990); respectively, and the total borate-salinity relationship from Lee et al. (2010).

### 2.4. Calculation of DIC and nDIC latitudinal gradients

10 The magnitude of the latitudinal gradient depends on the time of year. It is calculated because DIC values are higher in winter at high latitudes. The seasonal amplitude of surface nDIC varies over the global open ocean. It is generally small at low latitudes:  $\sim 20 \mu\text{mol kg}^{-1}$  in the subtropical Pacific Ocean (Keeling et al., 2004) and  $\sim 40 \mu\text{mol kg}^{-1}$  in the subtropical Atlantic Ocean (Bates et al., 1996). It is much larger at some (but not all) high-latitude locations:  $\sim 113 \mu\text{mol kg}^{-1}$  in the northwestern Pacific Ocean (Kawakami et al., 2007) and  $\sim 60 \mu\text{mol kg}^{-1}$  in the subarctic northeast Pacific Ocean (Wong et al., 2002), but only  $\sim 25 \mu\text{mol kg}^{-1}$  at the KERFIX site in the Southern Ocean (Louanchi et al., 1999). Because most ship-collected data (as contained in GLODAPv2) is collected in spring or summer months, the latitudinal gradients averaged across the whole year will be larger in some locations than presented here, and the magnitudes of the latitudinal gradients presented here should be considered as lower estimates. For instance, the observed nDIC difference ( $\Delta\text{nDIC}$ ) between the North Pacific ( $40^\circ\text{N} - 60^\circ\text{N}$ ) and low latitudes ( $30^\circ\text{S} - 30^\circ\text{N}$ ) is  $171 \mu\text{mol kg}^{-1}$  when calculated from summer data only,

15  $224 \mu\text{mol kg}^{-1}$  when calculated from winter data only. For the Southern Ocean,  $\Delta\text{nDIC}$  is  $214 \mu\text{mol kg}^{-1}$  in summer and  $240 \mu\text{mol kg}^{-1}$  in winter. This sensitivity to time of year should be noted but is not considered further here because it is relatively small compared to the overall magnitude of  $\Delta\text{nDIC}$ .

### 2.4.5 Calculations of the effects of various processes on DIC

25 The second hypothesis (evaporation and precipitation through their effects on TA) was evaluated by salinity normalization (Eq. 5). The methods for calculating the impacts of the other two processes on the surface DIC concentration are now explained. The effect of upwelling is evaluated in the Southern Ocean, from both short- and long-term perspectives. In addition, we also quantify the effect of iron limitation, which would potentially affect the observed (n)DIC distribution.

### 2.45.1 SST-driven effect

Generally, the temperature effect on the carbonate system has two aspects. Firstly, when water temperature increases, the equilibrium equilibria between carbonate species (Eq. 6) shifts to the left, towards increasing the aqueous CO<sub>2</sub> and carbonate ion concentrations (Dickson and Millero, 1987):



Secondly, CO<sub>2</sub> solubility is reduced-lower at higher temperatures and vice versa (Weiss, 1974). Neither effect alters DIC directly, but both lead to changes the seawater in pCO<sub>2</sub>. A larger proportion of DIC exists as aqueous CO<sub>2</sub> at higher temperatures and the ratio of pCO<sub>2</sub> to [CO<sub>2</sub>] also increases as solubility decreases (Eq. 7, Henry's Law):

$$p\text{CO}_2 = [\text{CO}_2]/K_H \quad (7)$$

10 where K<sub>H</sub> is the Henry's constant (solubility) for CO<sub>2</sub>, one of the CO<sub>2</sub>-system equilibrium constants.

Both effects thus tend to increase sea surface pCO<sub>2</sub> as sea water warms, potentially elevating it to values above the atmospheric pCO<sub>2</sub> and thereby stimulating increasing the net air-to-sea CO<sub>2</sub> gas exchange flux; the induced outgassing of CO<sub>2</sub> reduces sea surface pCO<sub>2</sub> and DIC as it shifts the system towards air-sea CO<sub>2</sub> equilibrium. Therefore, for an open ocean system with gas exchange in contact with the atmosphere, SST can control the DIC distribution has the potential to

15 be controlled by the SST distribution, and this can by itself produce DIC latitudinal variations.

To examine the magnitude of the expected temperature-induced DIC changes, we chose the low-latitude area as the reference, then removed the latitudinal SST variation and recalculated the open ocean surface DIC everywhere for a constant SST of 27°C (the mean sea surface temperature in the subtropics from 30° S- to 30° N). To do this wWe first calculated the in-situ pCO<sub>2</sub> from observed SST, SSS (sea surface salinity), TA and DIC using CO<sub>2</sub>SYS. The calculated

20 pCO<sub>2</sub> from TA and DIC has been found to agree well with the measured pCO<sub>2</sub> by Takahashi et al. (2014) (root mean square deviation of ± 6.8 µatm, Takahashi et al., 2014). We then altered the sea surface temperature from its in-situ value to 27°C, which would change the solubility of CO<sub>2</sub> and induce air-sea CO<sub>2</sub> gas exchange. Then air-sea CO<sub>2</sub> gas exchange (which does not change TA) was assumed to proceed until pCO<sub>2</sub> was back to the same level as before resetting the temperature. Next, we used CO<sub>2</sub>SYS to calculate DIC<sub>SST=27</sub> based on an input temperature of 27°C, observed salinity and TA, and the

25 in-situ pCO<sub>2</sub> calculated as above. DIC<sub>SST=27</sub> thus represents temperature-normalized DIC, and should exhibit the same spatial variability as DIC except that the temperature-induced component of the variability has been removed. Finally, the difference between observed DIC and DIC<sub>SST=27</sub> gives the DIC variation attributed to temperature variation:

$$\Delta\text{DIC}_{\text{temp}} = \text{DIC}_{\text{obs}} - \text{DIC}_{\text{SST=27}} \quad (8)$$

The same procedure was followed for calculating ΔnDIC<sub>temp</sub>:

$$30 \quad \underline{\Delta n\text{DIC}_{\text{temp}} = n\text{DIC}_{\text{obs}} - n\text{DIC}_{\text{SST=27}}} \quad (9)$$

## 2.45.2. Upwelled DIC-driven effect (short-term effect of upwelling)

Upwelling of DIC-rich subsurface waters ~~is capable of increasing~~can increase the surface DIC. The largest upwelling (~~in terms of flow rate~~)flux anywhere in the world takes place in the Southern Ocean (Talley, 2013): the upwelling there is made up of 18 Sv (Sverdrup, 1 Sv = 10<sup>6</sup> m<sup>3</sup>/s) of NADW (North Atlantic Deep Water), 11 Sv of IDW (Indian Deep Water), and 9 Sv of PDW (Pacific Deep Water). Subsurface waters in the Southern Ocean ~~are considered to~~ upwell along the neutral density isopycnals of 27.6 kg m<sup>-3</sup>, 27.9 kg m<sup>-3</sup> and 27.9 kg m<sup>-3</sup> in the southern Atlantic, Indian and Pacific Oceans, respectively (Ferrari et al., 2014; Lumpkin and Speer, 2007; Marshall and Speer, 2012; Talley, 2013).

Upwelling occurs within the Antarctic Circumpolar Current (ACC) ~~at those latitudes~~ where the wind stress is greatest (Morrison et al., 2015). ~~As~~ As the upwelled water subsequently advects away, the effects of upwelling on DIC are transported to nearby locations. Therefore, instead of a direct supply from deep to surface locations such as L3 (Fig. 4), DIC is ~~assumed to be~~ brought to the subsurface primarily along isopycnals (shown in Fig. 4 as the black curve to L1), finally reaching the surface at L2, ~~the zone in which upwelling occurs~~. Then, the upwelled ~~subsurface~~ waters s with enriched DIC, TA and nutrients feeds both branches of the Southern Ocean overturning ~~circulation in the Southern Ocean~~. One branch is transported northwards via Ekman transport from L2 to L3 ~~to join the upper branch~~, as shown by the black arrow towards the equator, and the other ~~one~~ is recycled ~~back through the lower branch~~ to form Antarctic Bottom Water (AABW) (Talley, 2013). ~~Potential~~ The effects of upwelling on sea surface temperature are ~~not considered here, but are likely to be small~~ negligible and not considered here, because both deep water and high-latitude surface waters have similarly low temperatures ~~are cold~~.

We first consider the increase in DIC induced by the upwelling of deep water with high DIC concentrations. While some of the initial increase is usually removed shortly afterwards by biological export fueled by the nutrients brought up at the same time, excess DIC remains if the subsequent biological removal of DIC does not match the initial increase. Phosphorus has the simplest nutrient behavior in the ocean with only one significant source to the ocean as a whole (river input) and one major sink (organic matter burial) (Ruttenberg, 2003; Tyrrell, 1999). In this study, the salinity-normalized phosphate (nPhos) concentration was used as a proxy for calculating how much salinity-normalized DIC (nDIC) was upwelled along with it and not yet removed again by biological uptake of phosphate and DIC. We used salinity-normalized concentrations to correct for the influence of precipitation (rainfall) that dilutes DIC and phosphate concentrations in proportion to the effects on salinity (Eq. 5). In this calculation, it was assumed that the only external source of phosphate to surface waters is from upwelling and the only subsequent loss is through export of organic matter, leading to the equation:

$$n\text{Phos}_{\text{surf}} = n\text{Phos}_{\text{supply}} - \text{NCP}/R_{\text{C:P}} \quad (910)$$



where the subscript ‘supply’ indicates the end-member concentration of deep water supplied along the upwelling isopycnals (i.e., the value at L1 in Fig. 4), and the subscript ‘surf’ indicates the surface water concentration at some later time. NCP refers to the total time-integrated net community production (uptake and export by biology) in carbon units, and  $R_{C:P}$  is the Redfield ratio of carbon to phosphorus.  $R_{C:P}$  is given the standard value of 106:1 (Redfield, 1963), except for the cold nutrient-rich high-latitude region in the Southern Ocean (south of 45° S), where  $R_{C:P}$  is given a lower value, of 78:1 (Martiny et al., 2013). We only considered the spatial variation of  $R_{C:P}$  in this study.  $R_{C:P}$  has seasonal variation as well (e.g., Frigstad et al., 2011), but this is much smaller than its latitudinal variation.  $nPhos_{surf}$  refers to the observed surface value of  $nPhos$  at some location distant from where upwelling occurs.

Another possible process involved in the change of DIC during its upwelling and subsequent advection is calcium carbonate ( $CaCO_3$ ) precipitation and dissolution (Balch et al., 2016), which alters DIC and TA with a ratio of 1:2. In order to quantify the magnitude of this process, we used  $Alk^*$  (Fry et al., 2015) as an indicator, which is capable of diagnosing  $CaCO_3$  cycling in the context of the large-scale ocean circulation (see more details on  $Alk^*$  distribution in Fig. 10). The change in  $Alk^*$  concentrations between its supplied and surface end-members is attributed to  $CaCO_3$  precipitation/dissolution and assimilation of inorganic nutrients by primary production (Brewer and Goldman, 1976):

$$15 \quad Alk_{surf}^* = Alk_{supply}^* - \Delta Alk_{CaCO_3}^* - NCP/R_{DIC:TA} \quad (411)$$

$$Alk^* = \frac{Alk_m - Alk_r + 1.36 \times NO_3^-}{S} \times 35 + Alk_r - 2300 \mu mol \text{ kg}^{-1} \quad (412)$$

where  $R_{DIC:TA}$  is the relative ratio of -106/17 between changes in DIC and TA during primary production (Wolf-Gladrow et al., 2007).  $Alk_m$  is the measured TA,  $Alk_r$  is the riverine TA end-member (zero in the Southern Ocean), and  $2300 \mu mol \text{ kg}^{-1}$  is the average TA in the low-latitude surface oceans.  $Alk_{surf}^*$  is calculated by Eq. 412.

20 Assuming the carbon source is from upwelled  $CO_2$ -rich deep waters and carbon sinks are from organic matter export (NCP) and  $CaCO_3$  cycling, then:

$$nDIC_{surf} = nDIC_{supply} - NCP - 0.5 \times \Delta Alk_{CaCO_3}^* \quad (413)$$

Three hydrographic sections, one in each of the Indian (I95E), Pacific (P150W), and Atlantic (A25W) Oceans, were used to determine the different supply concentrations ( $nPhos_{supply}$ ,  $Alk_{supply}^*$  and  $nDIC_{supply}$ ) for each basin (see Fig. 5c inset; ~~values here are expressed as mean  $\pm$  standard error of the mean~~). In the Indian Ocean,  $nPhos_{supply}$ ,  $Alk_{supply}^*$  and  $nDIC_{supply}$  along the  $27.9 \text{ kg m}^{-3}$  isopycnal are  $2.29 \pm 0.01 \mu mol \text{ kg}^{-1}$ ,  $109.4 \pm 1.0 \mu mol \text{ kg}^{-1}$ , and  $2273.1 \pm 1.1 \mu mol \text{ kg}^{-1}$ , respectively (values here are expressed as mean  $\pm$  standard error of the mean), as it approaches the surface. In the Pacific Ocean,  $nPhos_{supply}$ ,  $Alk_{supply}^*$  and  $nDIC_{supply}$  along the  $27.9 \text{ kg m}^{-3}$  isopycnal are  $2.32 \pm 0.01 \mu mol \text{ kg}^{-1}$ ,  $108.1 \pm 1.9 \mu mol \text{ kg}^{-1}$ , and  $2277.2 \pm 1.8 \mu mol \text{ kg}^{-1}$ , respectively. In the Atlantic Ocean,  $nPhos_{supply}$ ,  $Alk_{supply}^*$  and  $nDIC_{supply}$  along the  $27.6 \text{ kg m}^{-3}$  isopycnal are  $2.28 \pm 0.01 \mu mol \text{ kg}^{-1}$ ,  $103.5 \pm 1.1 \mu mol \text{ kg}^{-1}$ , and  $2254.6 \pm 1.3 \mu mol \text{ kg}^{-1}$ , respectively (Fig. 5).

Since  $n\text{Phos}_{\text{surf}}$  tends to decrease to zero upon moving northwards, due to biological uptake,  $n\text{DIC}_{\text{surf}}$  has a relatively constant value in the subtropical regions (data not shown), where is not influenced by upwelling in the Southern Ocean. Because of this, the potential effect of upwelling on surface  $n\text{DIC}$ , is calculated as the excess in  $n\text{DIC}_{\text{surf}}$  compared to the subtropical average value (30° S-30° N):

$$5 \quad \Delta n\text{DIC}_{\text{upw\_st}} = n\text{DIC}_{\text{surf}} - \overline{n\text{DIC}_{\text{surf}} (30^\circ \text{ S}-30^\circ \text{ N})} \quad (4314)$$

### 2.45.3. Upwelled TA-driven effect (long-term effect of upwelling)

Some effects of upwelling on DIC are temporary, becoming overridden later by gas-exchange. In contrast, the effect of upwelled TA persists because it changes the equilibrium DIC with respect to gas exchange ( $\text{DIC}_{\text{eq}}$ ) (discussed also in Sect. 4.1.3). Upwelling of high-TA water has a long-lasting effect on DIC because, if all else remains constant, an increase in TA decreases the fraction of DIC that exists as  $\text{CO}_2$  molecules. The resulting decrease in  $\text{CO}_2$  concentration lowers seawater partial pressure of  $\text{CO}_2$  ( $p\text{CO}_2$ ), having the potential to lower seawater  $p\text{CO}_2$  to below atmospheric values which in turn drives an influx of  $\text{CO}_2$  from the atmosphere, raising DIC (Humphreys et al., 2017). The effects of upwelling are complex because they consist of both direct and indirect effects on DIC (Fig. 2), lasting over both short (when DIC is altered but  $\text{DIC}_{\text{eq}}$  is not) and long (when  $\text{DIC}_{\text{eq}}$  is altered) timescales. The different effects and the meanings of the terms used here are illustrated in Fig. 6.

The calculation of the long-term effect of upwelling through upwelled TA in the Southern Ocean (i.e., the difference between  $\text{DIC}_3$  and  $\text{DIC}_0$  in Fig. 6) was achieved through five steps:

(1) calculation of TA in the Southern Ocean with the upwelling effect subtracted,  $\text{TA}_{\text{nonupw}}$ :

$$20 \quad \text{TA}_{\text{nonupw}} = \text{TA}_{\text{obs}} - (\text{Alk}^* \times S_{\text{obs}} / 35) \quad (4415)$$

where  $\text{TA}_{\text{obs}}$  is the observed in-situ TA, and  $\text{Alk}^*$  is the TA tracer (Fry et al., 2015) revealing excess TA supplied by the large-scale ocean circulation (upwelling in the Southern Ocean), as well as removal by calcification and export (Eq. 4412). Since  $\text{Alk}^*$  is a salinity-normalized concept, it is necessary to restore it to the in-situ salinity before subtracting it from the in-situ TA.

(2) calculation of in-situ sea surface  $p\text{CO}_2$ , following the same method as described in Sect. 2.4.2.

25 (3) calculation of DIC with the effect of upwelled TA subtracted. We calculated  $\text{DIC}_{\text{nonupw}}$  using  $\text{CO}_2\text{SYS}$  with inputs of  $\text{TA}_{\text{nonupw}}$  and in situ  $p\text{CO}_2$ , SST and salinity.

(4) salinity-normalization for consistency with other calculated effects.

(5) finally, the long-term effect of upwelling through the upwelled TA and the subsequent air-sea gas exchange is calculated as:

$$30 \quad \Delta n\text{DIC}_{\text{upw\_lt}} = n\text{DIC}_{\text{obs}} - n\text{DIC}_{\text{nonupw}} \quad (4516)$$

where  $\Delta n\text{DIC}_{\text{upw\_lt}}$  corresponds to the magnitude of ⑤ in Fig. 6.

#### 2.45.4. Iron-driven effect

The iron limitation-driven DIC differences ( $\Delta\text{DIC}_{\text{Fe}}$ ) relate to the concepts of “unused nutrient” and associated “unused DIC”, which can be thought of as the amounts of macro-nutrients and DIC that are left behind after iron limitation brings an end to biological uptake, in those regions where iron is the limiting nutrient. Iron limitation alters the impact of upwelling. In locations experiencing upwelling but where nitrate is the proximate limiting nutrient, then the quantity of upwelled DIC might more or less be balanced by the quantity of subsequently exported DIC (fueled by the upwelled nitrate). In the Southern Ocean, however, the two appear not to be close to balance, even before considering iron limitation. According to the calculations in Sect. 2.4.2, the ratio of the excess upwelled nDIC against nPhos is around  $250:2.3 \approx 109:1$  for the Southern Ocean, considerably exceeding the low C:P (average  $\approx 80:1$ ) of organic matter in the region (Martiny et al, 2013). So even if all upwelled phosphate were to be used up and then exported in biomass in conjunction with carbon, a considerable surplus of DIC would be left behind. A lack of iron in surface waters, however, leads to even more upwelled DIC being left behind after the end of blooms induced by the upwelled nutrients.

We used phosphate as the “unused nutrient” from which to calculate “unused DIC”.

For each  $1^\circ \times 1^\circ$  grid in the surface open ocean, the unused phosphate was taken from its annual minimum concentration based on the monthly data in World Ocean Atlas 2013 version 2 (WOA 2013: <https://www.nodc.noaa.gov/OC5/woa13/>, Boyer et al., 2013). The unused phosphate was then converted into unused DIC based on a C:P ratio of 106:1 (Redfield, 1963) for most of the global ocean, except in the warm nutrient-depleted low-latitude gyres, warm nutrient-rich equatorial upwelling regions, and cold nutrient-rich high-latitude regions. The C:P ratios used for these three regions were 195:1, 137:1, and 78:1, respectively (Martiny et al., 2013).

The amount of unused DIC was therefore calculated as:

$$\text{unused DIC} = \text{unused phosphate} \times R_{\text{C:P}} \quad (1617)$$

#### 2.56. Uncertainty estimation

~~In this study, uncertainties in calculated the effects of different drivers (e.g.,  $\Delta n\text{DIC}_{\text{temp}}$ ,  $\Delta n\text{DIC}_{\text{upw\_st}}$ ,  $\Delta n\text{DIC}_{\text{upw\_lt}}$ ) were~~ determined by a Monte Carlo approach (following e.g., Juranek et al., 2009; Ribas-Ribas et al., 2014). For example, the uncertainty of  $\Delta n\text{DIC}_{\text{temp}}$  was calculated as follows: (1) given that  $\Delta n\text{DIC}_{\text{temp}}$  is the difference between  $n\text{DIC}_{\text{obs}}$  and  $n\text{DIC}_{\text{SST}=27}$  (Eq. 9), its uncertainty is propagated from the uncertainties of both  $n\text{DIC}_{\text{obs}}$  and  $n\text{DIC}_{\text{SST}=27}$ , where the uncertainty of  $n\text{DIC}_{\text{obs}}$  is  $5 \mu\text{mol kg}^{-1}$  (Table 2), and the uncertainty of  $n\text{DIC}_{\text{SST}=27}$  was determined by a Monte Carlo approach; (2) for calculation of the uncertainty of  $n\text{DIC}_{\text{SST}=27}$  (see its function in Table 1), we first calculated artificial

random errors (normally distributed according to the central limit theorem, with a mean of zero and a standard deviation equal to the accuracy/uncertainty of measurement) using a random number generator. Then, new carbonate system variable values (the original ones plus the randomly generated errors) were input into the CO<sub>2</sub>SYS program (Van Heuven et al., 2011) to calculate new nDIC<sub>SST=27</sub> values. By doing this 1000 times, we obtained a set of 1000 different values for every single data point in the dataset. We used the standard deviations of these sets to characterize their individual uncertainties. The overall uncertainty of nDIC<sub>SST=27</sub> was 6.4 μmol kg<sup>-1</sup>; (3) by applying the same Monte Carlo method, but to calculate the uncertainty propagated through Eq. (9), we then calculated the uncertainty of ΔnDIC<sub>temp</sub> to be 8.0 μmol kg<sup>-1</sup> (Table 2). ~~This Monte Carlo approach has been used previously (e.g., Juranek et al., 2009; Ribas Ribas et al., 2014) to propagate uncertainties involving CO<sub>2</sub> system calculations.~~

## 10 3. Results

### 3.1. Spatial distributions of observed DIC and nDIC

Surface observations reveal values of DIC across the global ocean ranging from less than 1850 μmol kg<sup>-1</sup> in the tropics to more than 2200 μmol kg<sup>-1</sup> in the high latitudes (Fig. 1a). To first order, surface DIC increases polewards, being positively correlated with absolute latitude (Spearman's rank correlation coefficient  $\rho = 0.71$  for the global oceans, Table 3). Spatially, it is monotonically inversely related to sea surface temperature ( $\rho = -0.78$ , Table 3), with DIC being highest where the surface ocean is coolest. Another conspicuous feature of surface DIC is the higher values (by ~100 μmol kg<sup>-1</sup>) in the tropical and subtropical Atlantic Ocean relative to the same latitudes in the Pacific and Indian Oceans (Fig. 1a), as attributed to the transport of water vapor from the Atlantic to the Pacific (Broecker, 1989). This is not considered further here because our main purpose is to explain the sizeable observed latitudinal gradients in DIC (on average 153 μmol kg<sup>-1</sup> higher in the Southern Ocean than at low latitudes, for instance) and nDIC (on average 223 μmol kg<sup>-1</sup> higher in the Southern Ocean than at low latitudes).

Salinity-normalized DIC (nDIC) increases towards the poles in all three ocean basins (Fig. 1b), although less strongly in the North Atlantic. The surface nDIC correlates more tightly with latitude and SST than does DIC, yielding a positive correlation with absolute latitude and a negative correlation with SST ( $\rho = 0.86$  and  $-0.94$  respectively for the global ocean, Table 3).

The distributions of surface DIC and particularly nDIC also show modest regional maxima in the eastern equatorial Pacific, the Arabian Sea, and the eastern boundaries of the Pacific and Atlantic Ocean basins, presumably as a result of upwelling (Capone and Hutchins, 2013; Chavez and Messié, 2009; Millero et al., 1998; Murray et al., 1994).

### 3.2. SST-driven effect in the global surface ocean

The differences between the latitudinal patterns of  $\text{DIC}_{\text{obs}}$  and  $\text{DIC}_{\text{SST}=27}$  are shown in Fig. 7. As expected,  $\text{DIC}_{\text{SST}=27}$  agrees well with  $\text{DIC}_{\text{obs}}$  in the subtropics where SST is close to  $27^\circ\text{C}$ ; the differences become larger with increasing latitude and decreasing SST (Fig. 7a-c). Correcting for salinity variations (Fig. 7d-f) greatly reduces the variability in DIC at low

5 latitudes:  $n\text{DIC}_{\text{obs}}$  is fairly constant at  $\sim 1970 \mu\text{mol kg}^{-1}$  in the subtropics.  $\Delta n\text{DIC}_{\text{temp}}$  (Eq. 8 but for  $n\text{DIC}$ ), the temperature-driven  $\text{CO}_2$  gas exchange effect on surface  $n\text{DIC}$ , increases sharply with latitude (Fig. 7g-i), reaching  $-220 \mu\text{mol kg}^{-1}$  at  $70^\circ\text{S}$ , and  $\sim 200 \mu\text{mol kg}^{-1}$  at  $60^\circ\text{N}$  in the northern part of the Atlantic and Pacific Oceans, and  $\sim 220 \mu\text{mol kg}^{-1}$  at  $70^\circ\text{S}$  in the Southern Ocean. The average  $\Delta n\text{DIC}_{\text{temp}}$  in the Southern Ocean is with an average value of  $182 \mu\text{mol kg}^{-1}$  in the Southern Ocean, which is large enough to account by itself for most - but not all - of the  $n\text{DIC}$  latitudinal gradient of  $223 \mu\text{mol kg}^{-1}$  (2193-1970  $\mu\text{mol kg}^{-1}$ ).

The estimated overall uncertainty of SST-driven effect on surface  $n\text{DIC}$  (Table 2) ranges from 5 to  $8 \mu\text{mol kg}^{-1}$ , which is of comparable magnitude to the uncertainty of DIC normalized to 2005, and much smaller than the large latitudinal variations of  $\Delta n\text{DIC}_{\text{temp}}$ .

### 3.3. Upwelling-driven effects in the Southern Ocean

15 The upwelling-driven effects in the Southern Ocean calculated from both short- and long-term perspectives are shown in Fig. 8. The values were calculated from data collected along selected transects in each of the Atlantic, Indian, and Pacific sectors.

$\Delta n\text{DIC}_{\text{upw\_st}}$  increases polewards (Fig. 8a-c), with the same trends as surface phosphate (not shown), because it is calculated from phosphate. It can be seen that surface  $n\text{DIC}$  is potentially elevated dramatically by the Southern Ocean upwelling.

20 The effect is of larger magnitude (average of  $220 \mu\text{mol kg}^{-1}$  in the Southern Ocean) than that calculated for  $\Delta n\text{DIC}_{\text{temp}}$  (Fig. 7g-i).

Fig. 8d-f show the long-term effect of upwelling, which are controlled by the concentration of TA in the upwelled water (how much upwelling increases surface TA values by). The average magnitude of  $\Delta n\text{DIC}_{\text{upw\_lt}}$  is around  $74 \mu\text{mol kg}^{-1}$  for the Southern Ocean.

25 The estimated overall uncertainty of upwelling-driven effects on surface  $n\text{DIC}$  (Table 2) ranges from 5 to  $9 \mu\text{mol kg}^{-1}$ , close to the uncertainty of DIC normalized to 2005, and much smaller than the large latitudinal variations of  $\Delta n\text{DIC}_{\text{upw\_st}}$  and  $\Delta n\text{DIC}_{\text{upw\_lt}}$ .

### 3.4. Iron-driven effect in the global surface ocean

As shown in Fig. 9,  $\Delta\text{DIC}_{\text{Fe}}$  is close to zero except in the classic HNLC regions (i.e., the North Pacific, the equatorial Pacific, and the Southern Ocean, Moore et al., 2013). There is also some residual nitrate during most summers in the Iceland and Irminger Basins of the North Atlantic due to the seasonal iron limitation there (Nielsdóttir et al., 2009). The surface Southern Ocean south of 40° S has the largest unused DIC ( $\Delta\text{DIC}_{\text{Fe}}$  of up to 180  $\mu\text{mol kg}^{-1}$ , average of 120  $\mu\text{mol kg}^{-1}$ ), followed by the North Pacific 40° N-65° N ( $\Delta\text{DIC}_{\text{Fe}}$  of up to 120  $\mu\text{mol kg}^{-1}$ , average of 75  $\mu\text{mol kg}^{-1}$ ) and the equatorial Pacific (average of 35  $\mu\text{mol kg}^{-1}$ ). It is negligible elsewhere in the tropics and subtropics.

## 4. Discussion

### 4.1. Factors controlling the surface nDIC latitudinal variation

10 For each of comparison, all effects discussed in this section are effects on nDIC rather than on DIC

#### 4.1.1. Effect of SST variation in the global surface ocean

The previously accepted explanation for higher DIC at high latitudes is that cooler SSTs there increase  $\text{CO}_2$  solubility, resulting in a higher equilibrium DIC (Toggweiler et al., 2003a; Williams and Follows, 2011). Our results support an important role for SST, but also that other processes contribute significantly.

15 Our analysis concludes that the latitudinal gradient in temperature is capable of raising nDIC by about 180  $\mu\text{mol kg}^{-1}$  in the Southern Ocean, or in other words of explaining about four-fifths of the observed gradient of 223  $\mu\text{mol kg}^{-1}$ . SST variation is thus able to explain most of the observed pattern.

#### 4.1.2. Effect of TA distribution in the global surface ocean

20 A second factor that has been proposed as influential in driving spatial variations in the concentration of DIC in the surface ocean is TA (Williams and Follows, 2011). Our analysis supports this contention, although we note that the effect of TA is most prominent at low latitudes. Large differences in DIC are observed between the subtropical gyres, where values are relatively high, and the vicinity of the equator, where values are relatively low (Fig. 1a). These differences in DIC are driven initially by the direct effects of evaporation and precipitation on ~~TA~~DIC. However, direct effects of evaporation and precipitation on TA also lead to indirect effects on DIC because of the influence of TA on the value of DIC required for gas exchange equilibrium with a given atmospheric  $\text{CO}_2$  level, which then drive differences in DIC because of the influence of TA on DIC at gas exchange equilibrium with a given atmospheric  $\text{CO}_2$  level. The indirect effects will dominate

over longer timescales (see section 4.2 and Figure 6). The role of TA explains the much clearer relationship between latitude and nDIC than between latitude and DIC (Fig. 1, Table 3); normalizing DIC to salinity is almost the same as normalizing DIC to TA, because salinity and TA are highly correlated in the surface open ocean. As a result, the effect of TA on DIC is counteracted by salinity normalization, with the pattern in nDIC (Fig. 1b) then revealing more clearly how other factors impact DIC.

The latitudinal pattern in TA is not the dominant driver of the DIC trend, because TA values are generally lower at high latitudes (where precipitation often exceeds evaporation) than they are at low latitudes (where evaporation often exceeds precipitation). However, TA is also biologically cycled and thus not perfectly correlated to salinity (Fry et al., 2015) and the presence of excess TA in deep water upwelled at high latitudes does contribute to the DIC trend.

#### 4.1.3. Effect of upwelling in the Southern Ocean

Although not traditionally considered as a factor, our analyses show that upwelling is important in driving the latitudinal gradient in DIC. Upwelling of DIC by itself is capable of producing an nDIC latitudinal gradient of  $220 \mu\text{mol kg}^{-1}$  in the Southern Ocean, even higher than the effect of temperature (Fig. 8a-c, Table 4). However, the contribution of upwelling is reduced by about two thirds if only the long-term effect through upwelled TA is considered (see Fig. 6 for definitions of terms).

Deep water usually has higher concentrations of nutrients, nDIC and nTA than does surface water. Introduction of deep water into the surface mixed layer therefore usually stimulates increases in these concentrations, with three main consequences for DIC (Fig. 6), as follows. (A) If the upwelled water has higher DIC than the surface, then the upwelling can cause an immediate initial increase in DIC; (B) additional nutrients stimulate phytoplankton blooms until the proximate limiting nutrient runs out, leading to a reduction in DIC over timescales of days to weeks (or months if, for instance, the upwelling occurs at high latitudes during winter when phytoplankton cannot bloom); (C) finally, air-sea gas exchange tends to remove any upwelling-induced air-sea  $\text{CO}_2$  disequilibrium over a period of months to a year (Jones et al., 2014), although the full equilibrium is seldom achieved across the global surface ocean (Takahashi et al., 2014).

The upwelling effects in Fig. 8 are calculations based on phosphate and TA concentrations, taking into account both the amount upwelled, and the amount subsequently removed by biology. They therefore correspond to the sum of the direct upwelling effect (① in Fig. 6) and the indirect upwelling effect through supplied nutrients (② in Fig. 6). There are two reasons why the initial amount of upwelled DIC considerably exceeds the amount of DIC subsequently taken up by phytoplankton growth fueled by the upwelled nutrients (why ① > ②) in the Southern Ocean.

Firstly, iron is typically much scarcer in deep waters than are macronutrients, relative to phytoplankton need (Moore, 2016).

Regions like the Southern Ocean that are strongly influenced by upwelling are for this reason often iron-limited (Moore,

2016), leading to large amounts of ‘unused DIC’ (order of  $120 \mu\text{mol kg}^{-1}$  in the Southern Ocean - Fig. 9) accompanying unused macronutrients. This scarcity of iron also leads to muted seasonal cycles of DIC (Merlivat et al., 2015) and thus year-round persistence of unused DIC. Secondly, as described in Sect. 2.4.5, the higher C:P ratio of supply ( $\sim 109:1$ ) compared to removal ( $\sim 80:1$ ) implies a considerable surplus of DIC even without iron limitation.

5 The upwelling effects shown in Fig. 8a-c are however relatively short-term, and are expected to be overridden by air-sea gas exchange within months (Jones et al., 2014). They are thus likely to be most significant in the vicinity of where upwelling takes place (Morrison et al., 2015). For effects that may persist further away from locations of upwelling, it is important to consider also the long-term effect (⑤ in Fig. 6), the magnitude of which is dictated mainly by the change ~~in~~ of TA brought about by upwelling. The level of TA in upwelled water ( $\sim 2315$ ,  $2340$ , and  $2337 \mu\text{mol kg}^{-1}$  in the Atlantic, Indian and Pacific sectors of Southern Ocean, respectively; calculated according to the same method as in Sect. 2.4.3) are higher than the typical levels of TA in the surface waters of the high latitude Southern Ocean ( $\sim 2300$ ,  $2289$ , and  $2288 \mu\text{mol kg}^{-1}$  in the Atlantic, Indian and Pacific sectors, respectively). The increase in TA brought about by upwelling corresponds to a long-term upwelling effect on nDIC of about  $74 \mu\text{mol kg}^{-1}$  (Fig. 8d-f) in the Southern Ocean.

Our results show that upwelling in the Southern Ocean can, by itself, generate high-latitude nDIC values that are around 15  $220 \mu\text{mol kg}^{-1}$  greater than subtropical values. We emphasize that there is, in addition, a sizeable long-term effect of upwelling (forcing nDIC values to be around  $74 \mu\text{mol kg}^{-1}$  higher than they would be otherwise). Contrary to what might typically be assumed, the long-term effects of upwelling are dictated by the amounts of TA upwelled, and not by the amounts of DIC or nutrients.

#### 4.2. A new understanding of the controls on the surface DIC distribution

20 ~~The analyses presented above show that our understanding of the causes of latitudinal gradient in DIC needs to be revised. Our analysis revises the prevailing paradigm of the causes of the latitudinal gradient in surface DIC. Whereas it was considered previously that~~ Previously, the latitudinal gradient ~~was thought to be in DIC is~~ completely explained by ~~the effect of~~ sea surface temperature ~~on CO<sub>2</sub> solubility variation, but here~~ we have shown that upwelling is also an important contributor ~~to it, based on an evaluation in the Southern Ocean~~. DIC and nDIC would still be elevated at high latitudes 25 even ~~without any temperature effect, if there was no temperature effect (even if CO<sub>2</sub> solubility was completely unaffected by SST). The incomplete previous view of what drives the DIC latitudinal gradients should be replaced by this more nuanced view in which upwelling is also seen to contribute.~~

~~It seems that n~~ Neither temperature ~~variation patterns~~ nor upwelling are responsible for all of the observed large latitudinal gradients in DIC and nDIC (for instance, the  ~~$\sim 223$~~   $\sim 220$   $\mu\text{mol kg}^{-1}$  difference in nDIC between low-latitudes and the Southern 30 Ocean), but rather ~~that~~ they are jointly responsible. There is an apparent contradiction because both  $\Delta\text{nDIC}_{\text{temp}}$  and



$\Delta nDIC_{upw\_st}$  appear to account for more than 80% of the nDIC latitudinal gradient. While both processes are capable individually of raising  $nDIC$  by 182 and 220  $\mu mol\ kg^{-1}$  in the Southern Ocean, acting together they raise it by only 223  $\mu mol\ kg^{-1}$  instead of 400  $\mu mol\ kg^{-1}$ . An obvious explanation of this apparent paradox is that when we consider upwelling effects, we should consider not only its short-term effect through supplying DIC and nutrients ((1) + (2) in Fig. 6), but also

5 its long-term effect with gas exchange with the atmosphere ((5) = (1) + (2) + (3) in Fig. 6), the amount of which is a function of the amount of upwelled TA (which, together with temperature, controls the equilibrium DIC). The sum of the SST-driven effect and the long-term effect of upwelling approximately equals the nDIC latitudinal gradient (Table 4).

On the global scale, therefore, the ultimate controls on the surface DIC and nDIC latitudinal gradients are the spatial patterns of SST and upwelling, and the chemical composition of the upwelled water.

#### 10 4.3. Importance of upwelling confirmed by the North Atlantic

From inspection of the global nDIC distribution (Fig. 1b), it can be seen that nDIC increases with latitude in all basins, but, as shown in Table 4, does so less strongly in the North Atlantic (difference between high latitudes and low latitudes of 114  $\mu mol\ kg^{-1}$ ) than in the North Pacific (difference of 192  $\mu mol\ kg^{-1}$ ). Although the latitudinal temperature gradient is less pronounced in the North Atlantic, this is not enough to explain the variation in gradients between the two basins: the

15 average temperature of the high-latitude North Atlantic is 12.4°C and of the high-latitude North Pacific is 9.5°C, which can explain about 20  $\mu mol\ kg^{-1}$  of variation between the two nDIC gradients but cannot explain the observed 78  $\mu mol\ kg^{-1}$  variation (Table 4).

The reason for the discrepancy is that the Southern Ocean and the North Pacific experience elevations in values due to inputs of deep water whereas the North Atlantic does not. Upwelling occurs in the Southern Ocean and entrainment due to

20 deep winter mixing occurs in the subarctic North Pacific (Mecking et al., 2008; Ohno et al., 2009) where it entrains waters high in both TA (Fry et al., 2016) and DIC. While deep winter mixing also occurs in the high latitude North Atlantic (de Boyer Montégut et al., 2004), the entrained waters left the surface relatively recently and hence there is little accumulated remineralized DIC and TA in the deep water that is reintroduced to the surface. For this reason, winter entrainment produces little increase in surface nDIC in the North Atlantic. This makes the North Atlantic useful in discriminating between the

25 two effects because, uniquely out of the three regions, only the SST effect operates there. As expected, the SST effect is able to completely account for the observed nDIC gradient in the North Atlantic, whereas it cannot in the other two regions (columns 2 and 3 of Table 4). The North Atlantic confirms the important contribution of upwelling to latitudinal gradients, while also showing that latitudinal gradients occur in the absence of upwelling.

#### 4.4. Comparison of nDIC distribution to Alk\* and nutrients

Fig. 10 shows a comparison between the patterns of nDIC, the TA tracer Alk\* (Eq. 11, Fry et al., 2015) and salinity-normalized nutrients. The similarities and differences in distributions of Alk\* and nutrients have previously been discussed by Fry et al. (2015). Here we extend the comparison to also include nDIC. All exhibit low and fairly constant values at low latitudes. This is primarily due to biological uptake and restricted supply from subsurface waters, for most variables, but is primarily due to fairly uniform high temperatures for nDIC. All increase polewards due to upwelling/entrainment (also SST for nDIC), exhibiting maxima at high latitudes in the Southern Ocean and North Pacific. All exhibit a more modest increase in the North Atlantic than in the North Pacific, because the deep water formed relatively recently. The coincident increases in nDIC and nitrate in the north Indian Ocean and equatorial Pacific Ocean are not matched by increases in either Alk\* or silicate, most probably due to the source waters for the upwelling coming from depths that are shallower than the dissolution depths of calcium carbonate and opal (Fry et al., 2015; Schlitzer, 2000). There are differences in the latitudes at which the different parameters start to increase on a transect from the equator towards Antarctica, reflecting the different processes involved. Surface nDIC is the first to start increasing, under the influence of SST (rows 3 and 4 of Fig. 7), at around 20° S in the Atlantic and Pacific Oceans and 25° S in the Indian Ocean. Alk\* and nitrate, on the other hand, do not start to increase until about 40° S in the Atlantic and Indian Oceans and about 30° S in the Pacific Ocean. Silicate does not increase in concentration until about 50° S, for reasons that are still debated (Assmy et al., 2013; Holzer et al., 2014; Vance et al., 2017).

#### 4.5. Implications for the future CO<sub>2</sub> sink under climate change

It is widely understood that global warming may alter the spatial distribution and intensity of upwelling in the ocean (Bakun, 1990; McGregor et al., 2007; Wang et al., 2015). It could either increase it on average, due to higher average wind speeds in a warmer, more energetic atmosphere (Bakun, 1990; Wang et al., 2015), or decrease it on average, due to enhanced stratification as the temperature differential between surface and deep waters is increased (Barton et al., 2013; Sarmiento et al., 2004b). Furthermore, it is widely understood that an increase in upwelling would lead to an increase in the amount of CO<sub>2</sub> outgassed from the ocean, as larger quantities of CO<sub>2</sub>-rich deep water are brought up to the surface and their CO<sub>2</sub> vented to the atmosphere (Evans et al., 2015; Marinov et al., 2006; Morrison et al., 2015). However, we have identified an additional effect here. Changes in upwelling would alter the distribution of carbon in the surface ocean not only through the supply of CO<sub>2</sub>, but also through the supply of TA which determines the eventual surface carbonate system equilibrium with the same atmospheric *p*CO<sub>2</sub> (Humphreys et al., 2018). That is to say, the impact of changes in upwelling on the ocean's carbon source/sink strength depends not only on the DIC content of the upwelled water but also on its TA

content. Ocean carbon cycle models should include these additional consequences if they are to make accurate predictions about the impacts of global warming on future carbon cycling. They should include the several routes identified here by which upwelling affects surface DIC: through upwelling of DIC, through upwelling of nutrients, and through upwelling of TA.

## 5 5. Conclusions

We investigated the global surface DIC and nDIC distributions in order to explain the large differences between high latitude (especially Southern Ocean) and low-latitude regions. This issue has been addressed in previous studies and here we revisited it using new analytical approaches that lead to new findings. We considered three drivers for how the phenomenon could be explained: (1) sea surface temperature variations through their effect on CO<sub>2</sub> system equilibrium constants, (2) salinity-related TA variations through their effect on *p*CO<sub>2</sub>, and (3) upwelling in the subpolar oceans. Our analyses confirmed that temperature plays a dominant role through its effect on solubility, and is able to explain a large fraction of the surface nDIC latitudinal gradient (182 μmol kg<sup>-1</sup> out of 223 μmol kg<sup>-1</sup> in the high-latitude Southern Ocean). Variations in TA associated with evaporation and precipitation are unable to explain higher DIC concentrations at higher latitudes, because alone they would drive the opposite DIC pattern. Their role is therefore to reduce the magnitude of the polewards gradient in DIC. Upwelling, whose role in driving the large-scale spatial patterns has not previously been appreciated, accounts for a sizeable component of the surface nDIC latitudinal gradient (on average 220 μmol kg<sup>-1</sup> in the Southern Ocean). Its importance is magnified by the iron limitation that frequently occurs in upwelling areas, leaving behind residual upwelled excess DIC and macronutrients that cannot be utilized by biology. We emphasize that the upwelling of TA alongside DIC generates a prolonged effect that persists beyond CO<sub>2</sub> gas exchange re-equilibration timescales. The long-term effect of upwelling (74 μmol kg<sup>-1</sup> in the Southern Ocean) helps explain the shortfall between the observed nDIC latitudinal gradient (223 μmol kg<sup>-1</sup>) and the magnitude of the temperature-driven effect (182 μmol kg<sup>-1</sup>). On the global scale, we conclude that no single mechanism accounts for the full amplitude of surface DIC latitudinal variations but that temperature and the long-term effect of upwelling, in that order, are the two major drivers.

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**Table 1.**

**Definitions of Subscripts and Main Terms used in the Text.** X represents any variable involved in the calculations. The program CO<sub>2</sub>SYS was used to calculate values under different conditions.

<i>Subscript</i>	<i>Meaning</i>
<i>Referring to (n)DIC values at a particular location</i>	
X <sub>supply</sub>	Value at depth, along isopycnals that upwell at this location
X <sub>surf</sub>	Predicted value in the surface layer
X <sub>obs</sub>	Observed value at this surface location
<i>Referring to predicted (n)DIC values under different conditions</i>	
X <sub>SST=27</sub>	Predicted value with sea surface temperature changed to 27°C
X <sub>nonupw</sub>	Predicted value with upwelled TA subtracted
<i>Referring to changes in (n)DIC values because of processes</i>	
ΔX <sub>Fe</sub>	Effect of iron limitation (biological drawdown that is prevented)
ΔX <sub>temp</sub>	Effect of sea surface temperature variations
ΔX <sub>upw_st</sub>	Short-term effect of upwelling, through upwelled DIC
ΔX <sub>upw_lt</sub>	Long-term effect of upwelling, through upwelled TA
<i>Carbonate variables used to calculate predicted DIC values with CO<sub>2</sub>SYS</i>	
DIC <sub>SST=27</sub> = f(T <sub>SST=27</sub> , S <sub>in-situ</sub> , TA <sub>in-situ</sub> , pCO <sub>2,in-situ</sub> )	DIC <sub>SST=27</sub> is a function of in-situ S, TA, and pCO <sub>2</sub> , and SST at 27°C
DIC <sub>nonupw</sub> = f(T <sub>in-situ</sub> , S <sub>in-situ</sub> , TA <sub>nonupw</sub> , pCO <sub>2,in-situ</sub> )	DIC <sub>nonupw</sub> is a function of in-situ SST, S, and pCO <sub>2</sub> , and pre-upwelling TA

**Table 2.****Uncertainties for variables in this study.**

<i>Initial Variable</i>	<i>Uncertainty</i>	<i>Reference</i>
Salinity	0.005	Olsen et al. (2016)
Phosphate	0.05 $\mu\text{mol kg}^{-1}$	Olsen et al. (2016)
DIC	4 $\mu\text{mol kg}^{-1}$	Olsen et al. (2016)
TA	6 $\mu\text{mol kg}^{-1}$	Olsen et al. (2016)
$p\text{CO}_2$	6.8 $\mu\text{atm}$	Takahashi et al. (2014)
DIC normalized to 2005	5.0 $\mu\text{mol kg}^{-1}$	derived in this study <sup>a</sup>
Alk <sup>*</sup>	6.1 $\mu\text{mol kg}^{-1}$	Modified from Fry et al. (2015)
$n\text{PhOS}_{\text{supply}}$ , Alk <sup>*</sup> <sub>supply</sub> , $n\text{DIC}_{\text{supply}}$	See text in Sect. 2.4.3	derived in this study <sup>b</sup>
<i>Calculated Propagated Uncertainties</i>		
$\Delta n\text{DIC}_{\text{temp}}$	8.0 $\mu\text{mol kg}^{-1}$	derived in this study
$\Delta n\text{DIC}_{\text{upw\_st}}$	5.4 $\mu\text{mol kg}^{-1}$	derived in this study
$\Delta n\text{DIC}_{\text{upw\_lt}}$	8.9 $\mu\text{mol kg}^{-1}$	derived in this study

<sup>a</sup>the uncertainty of DIC normalized to 2005 was primarily propagated from TA and  $p\text{CO}_{2,\text{sw}}^{2005}$ . The uncertainty of  $p\text{CO}_{2,\text{sw}}^{2005}$  was calculated from error propagation (Fornasini, 2008), to be 0.17  $\mu\text{atm}$ .

5 <sup>b</sup>the uncertainties for variables with subscript “supply” were from their standard error of the mean.

**Table 3.****Global and Regional Correlations Between DIC, nDIC and SST, Latitude.**

Ocean	Region	DIC vs. Lat		nDIC vs. Lat		DIC vs. SST		nDIC vs. SST	
		$\rho^a$	N <sup>b</sup>	$\rho$	N	$\rho$	N	$\rho$	N
Global		<b>0.71</b>	14228	<b>0.86</b>	14228	<b>-0.78</b>	14228	<b>-0.94</b>	14228
Southern Ocean	S of 40° S	<b>0.79</b>	3061	<b>0.81</b>	3061	<b>-0.93</b>	3061	<b>-0.95</b>	3061
North Atlantic	N of 40° N	<b>0.30</b>	1640	<b>0.58</b>	1640	<b>-0.34</b>	1640	<b>-0.78</b>	1640
North Pacific	N of 40° N	<b>0.02</b>	1601	<b>0.34</b>	1601	<b>-0.78</b>	1601	<b>-0.87</b>	1601

<sup>a</sup>the Spearman's rank correlation coefficient, for assessing monotonic relationships (there is a non-linear relationship between SST and CO<sub>2</sub> solubility). Statistically significant correlations are shown in bold.

5 <sup>b</sup>The number of data points from that area that were used in calculating the correlations.

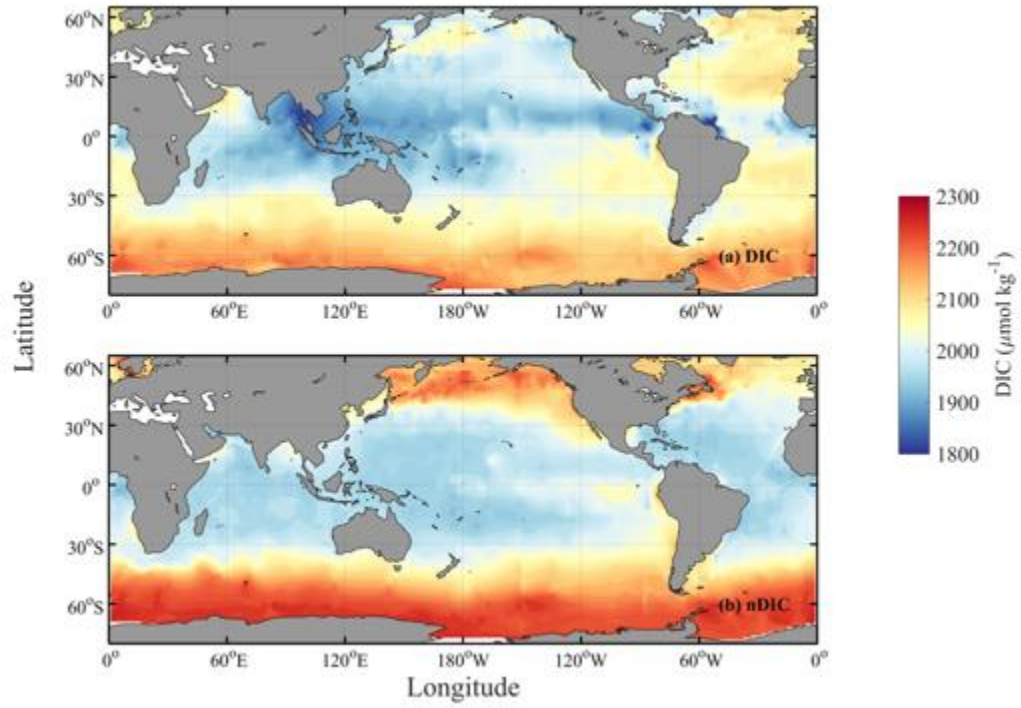


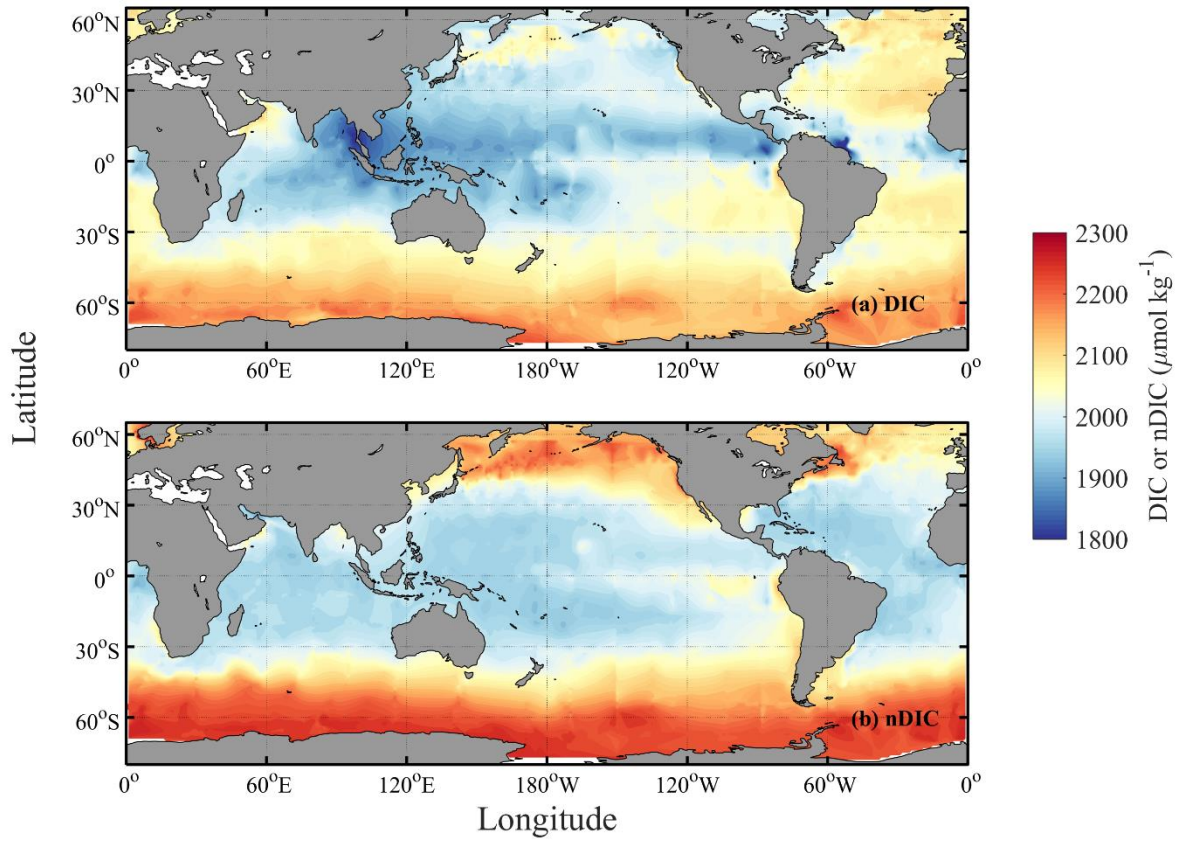
**Table 4.**

**Summary of nDIC Differences Between Low and High Latitudes.** Each  $\Delta$ nDIC value is the amount by which the annual average nDIC value for the high latitude region exceeds the annual average value for the low latitudes (30° S to 30° N). %'s in brackets represent the ratio to the observed nDIC difference in column 2. n.c. = not calculated.

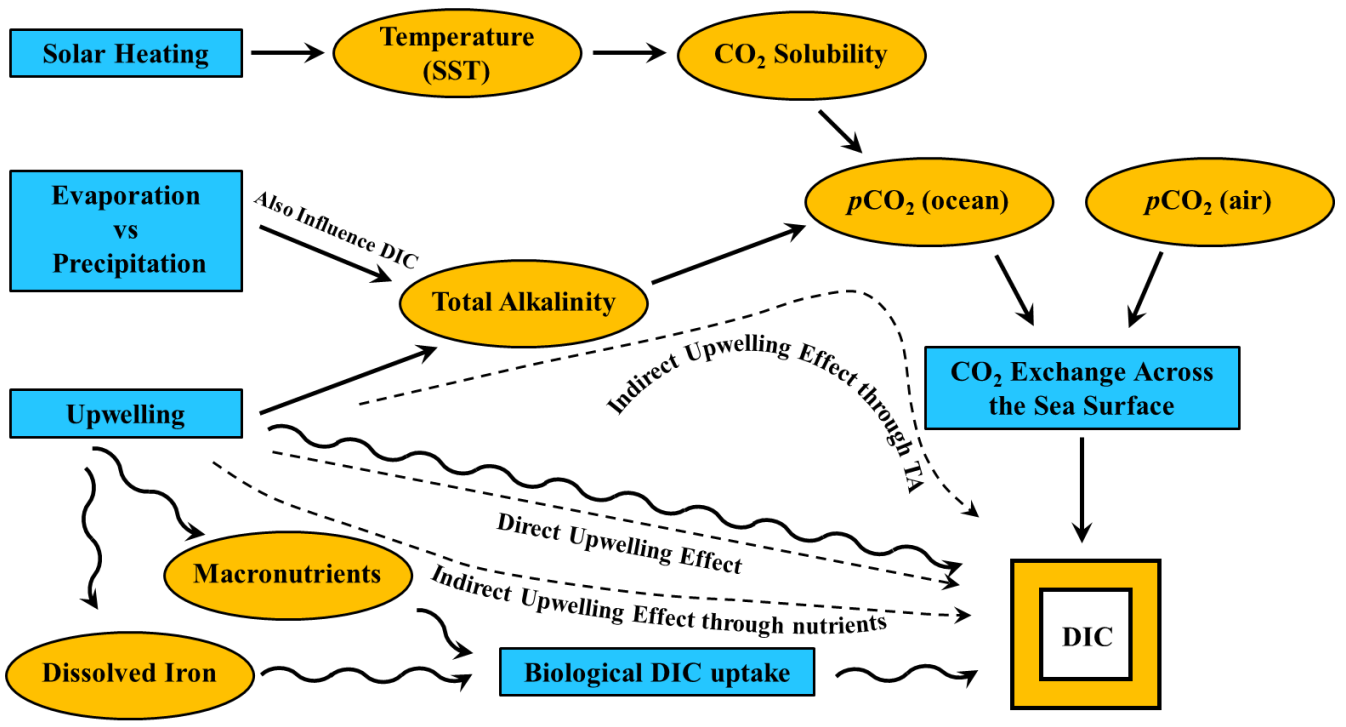
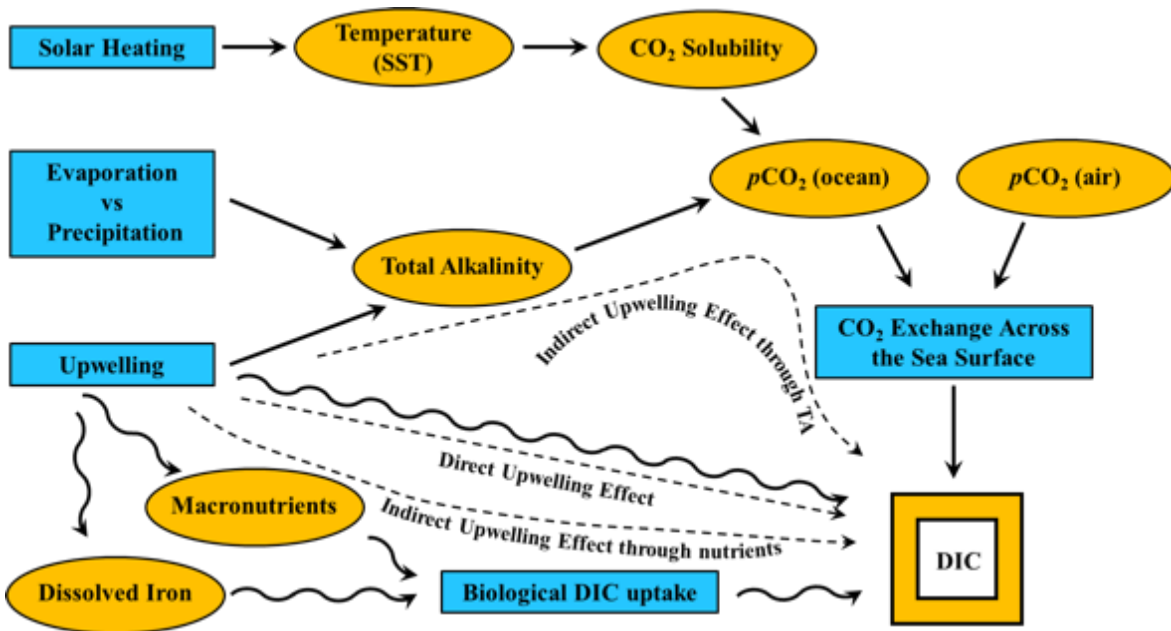
Region <sup>a</sup>	Observed $\Delta$ nDIC ( $\mu\text{mol kg}^{-1}$ )	$\Delta$ nDIC <sub>temp</sub> ( $\mu\text{mol kg}^{-1}$ )	$\Delta$ nDIC <sub>upw_st</sub> ( $\mu\text{mol kg}^{-1}$ )	$\Delta$ nDIC <sub>upw_lt</sub> ( $\mu\text{mol kg}^{-1}$ )
Southern Ocean	223	182 (82%)	220 (98%)	74 (33%)
North Atlantic	114	122 (107%)	n.c.	n.c.
North Pacific	192	137 (71%)	n.c.	n.c.

5 <sup>a</sup>The regions are defined as follows: North Atlantic: 40° N - 60° N; North Pacific: 40° N - 60° N; Southern Ocean: S of 40° S.

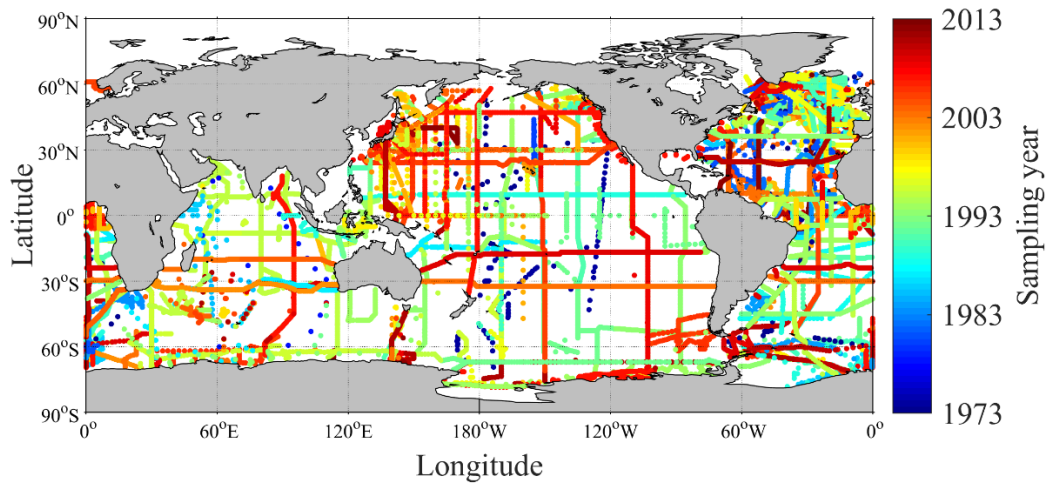




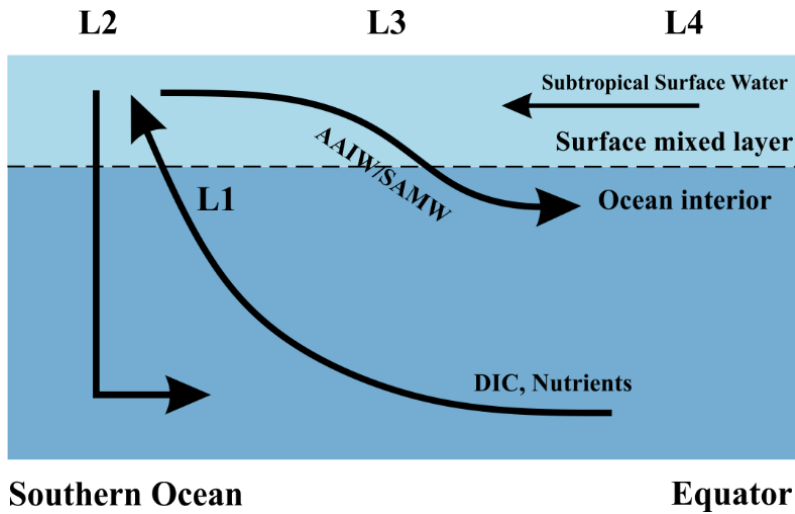
**Figure 1. Spatial distributions of DIC and nDIC.** (a) DIC (normalized to year 2005), (b) salinity-normalized DIC (nDIC, DIC normalized to reference year of 2005 and salinity of 35) in the surface global ocean. The latitudinal trends are clear, particularly for nDIC.



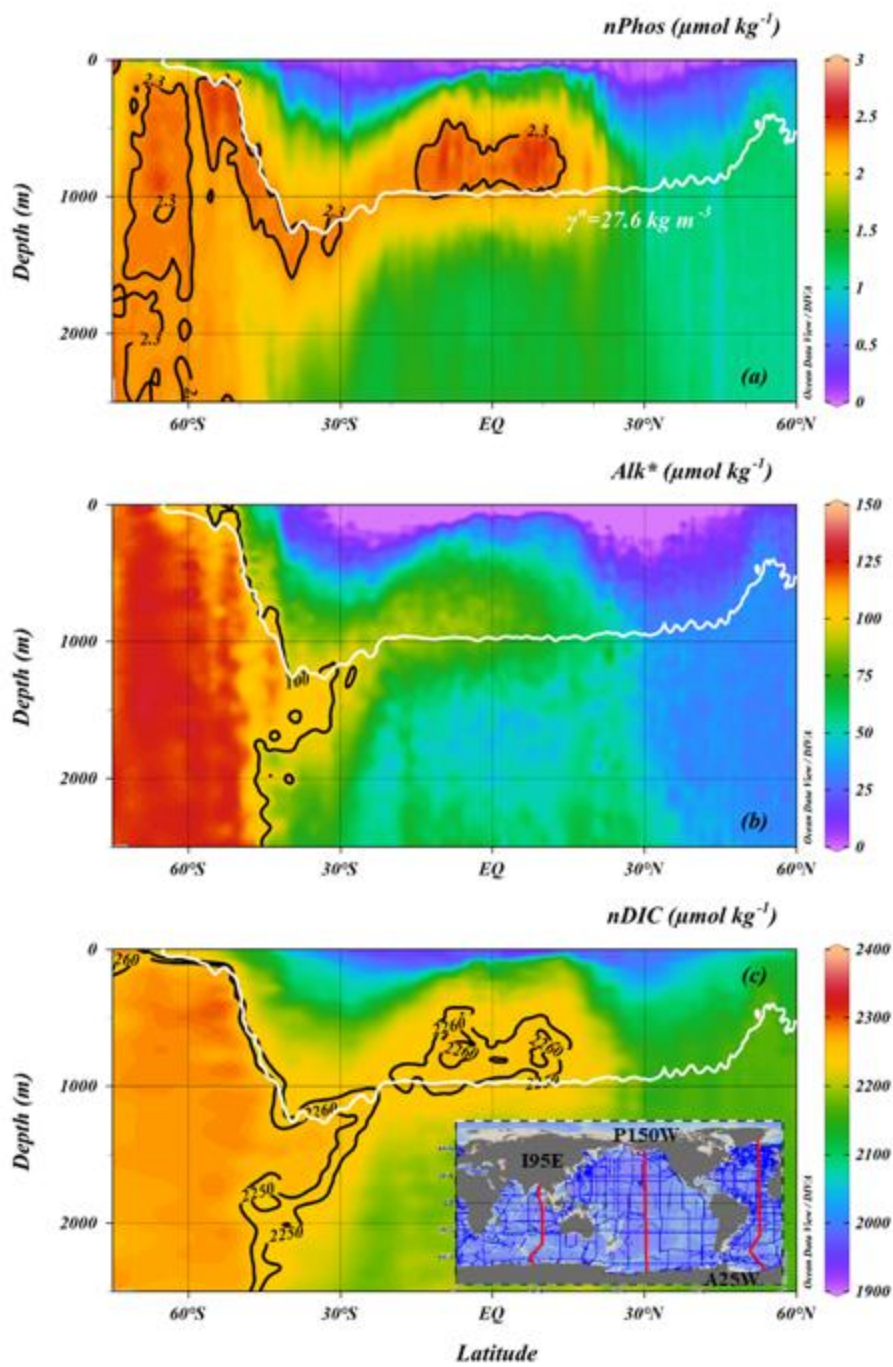
**Figure 2. Major controls on surface DIC.** Schematic showing the main processes exerting an influence over the concentration of DIC in the global surface ocean (producing variation with latitude). Blue shapes are processes and orange shapes are variables. Straight solid arrows represent equilibrium processes regulating  $\Delta$ DIC in the long-term and wavy solid arrows represent disequilibrium processes regulating  $\Delta$ DIC in the short-term. In the manuscript, we evaluate the upwelling effect on surface DIC in the Southern Ocean. Dashed arrows with text denote the three different ways that upwelling affects DIC: the direct effect through upwelled DIC; the indirect effect through upwelled nutrients which stimulates biological removal of DIC; and the indirect effect through upwelled TA in changing the equilibrium DIC with the atmosphere.



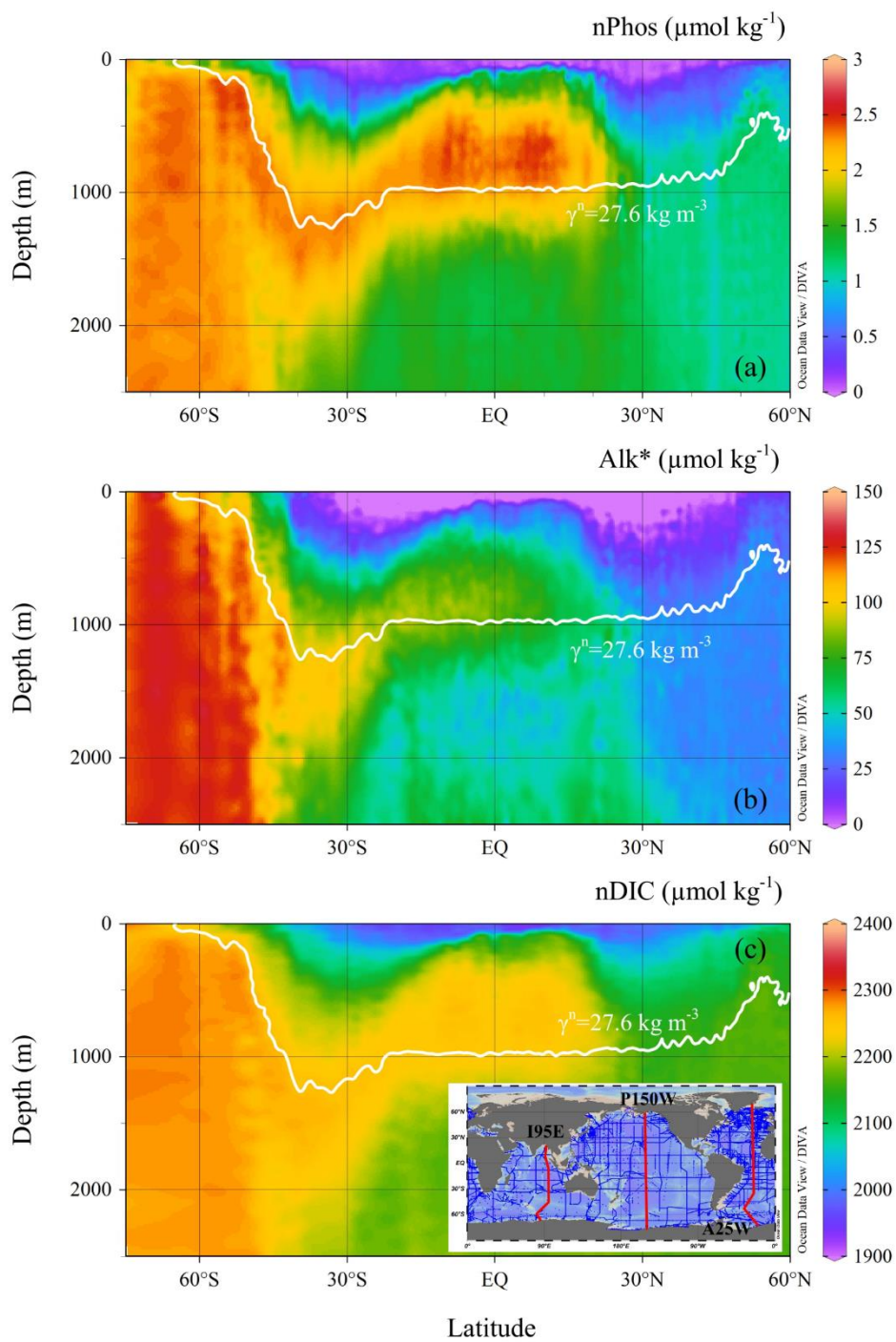
**Figure 3. Spatial and temporal distribution of GLODAPv2 sampling stations used for this study.**



**Figure 4.** A schematic illustrating locations of interest and assumed major flow paths in the Southern Ocean. Black arrows represent the flow directions of water masses. The lower curved arrow denotes upwelling of deep water along isopycnal surfaces, and the upper curved arrow denotes subduction to form Subantarctic Mode Water (SAMW) and Antarctic Intermediate Water (AAIW). L1: upwelling water below the mixed layer, prior to any influence of surface processes; L2: sea surface within the core of the Southern Ocean upwelling south of 50° S (Morrison et al., 2015); L3: sea surface from 30° S to 50° S; L4: sea surface north of 30° S which experiences no direct effects from upwelling in the Southern Ocean.

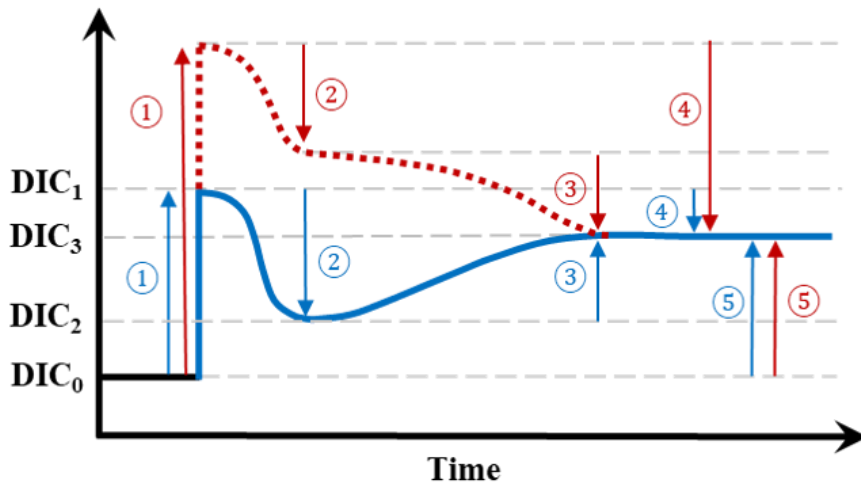




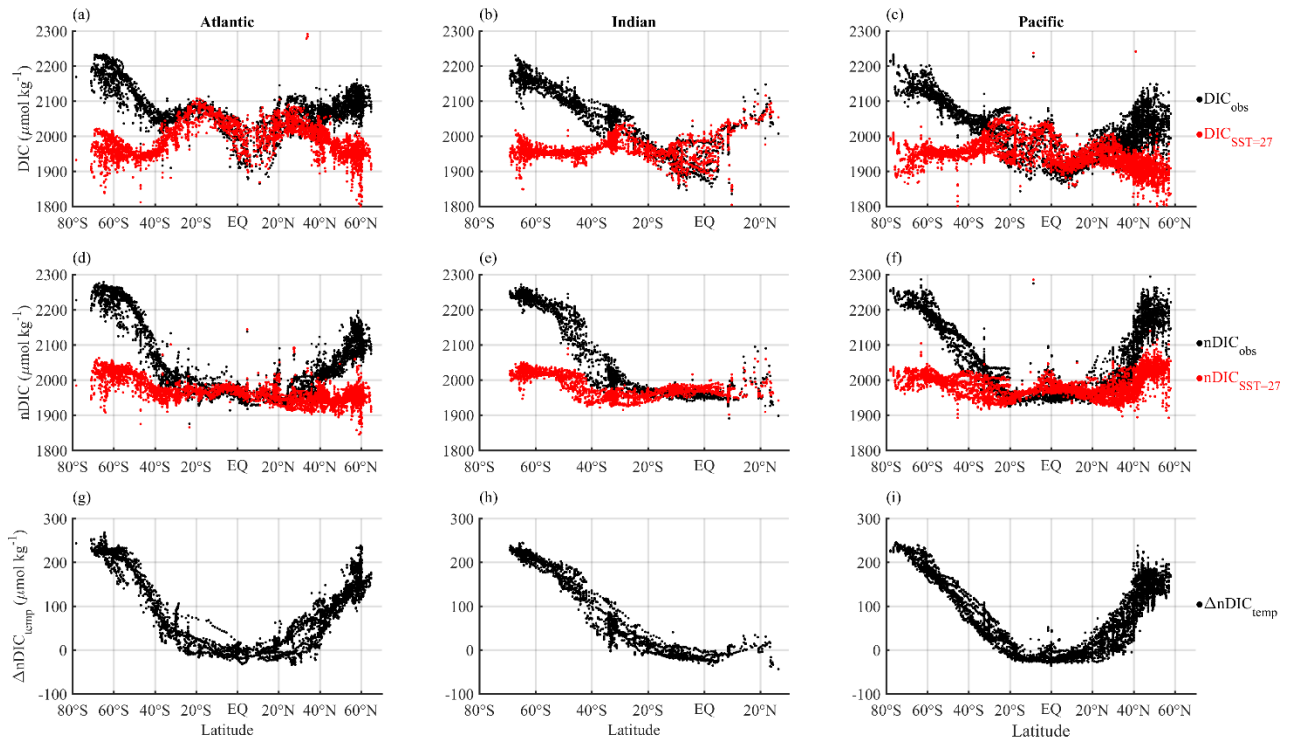


**Figure 5. Vertical distributions of (a) nPhos, (b) Alk<sup>\*</sup> and (c) nDIC along the Atlantic Ocean section.** The Indian and Pacific sections are not shown. The selected Atlantic section (A25W) is shown as the red line on the right-hand side of the inset. The neutral density isopycnal along which upwelling occurs is indicated by the white contour, ~~with the black contours referring to the observed variable, which is characterized by neutral density of 27.6 kg m<sup>-3</sup> in the Atlantic sector of the~~ Southern Ocean.

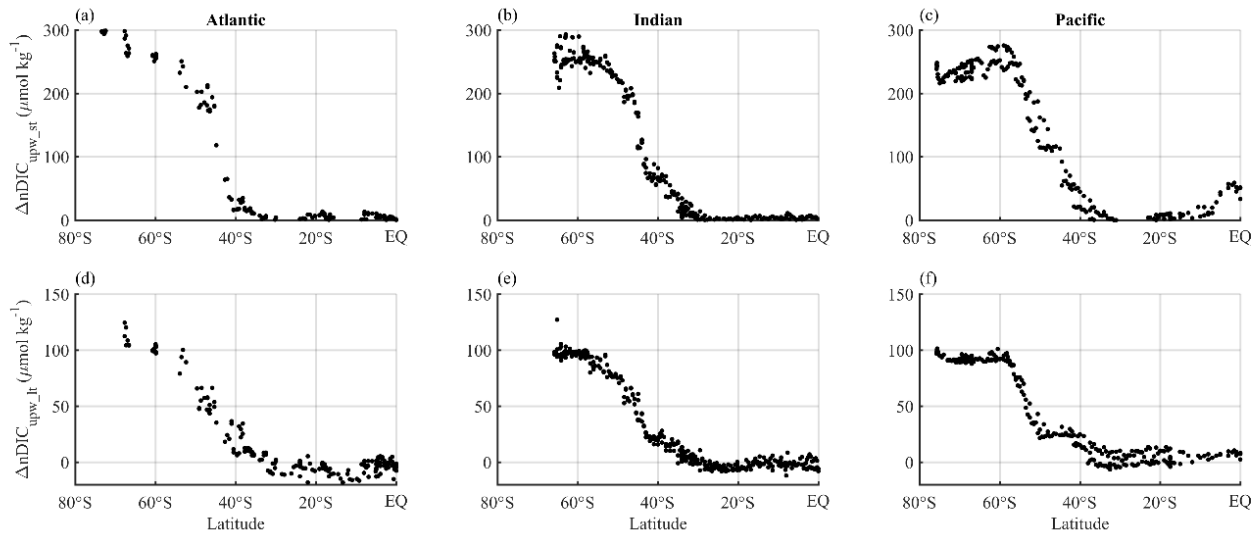
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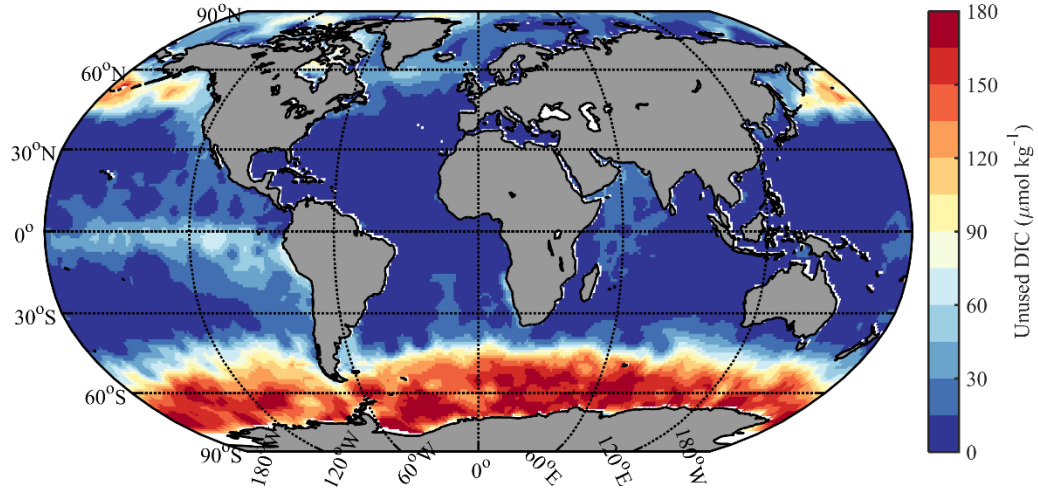
**Figure 6. A schematic illustrating the various effects of upwelling on surface DIC.** Numbers represent processes changing surface DIC, arrows point in the direction of change: ①: the *direct effect* of upwelling which elevates surface DIC from  $DIC_0$  to  $DIC_1$ ; ②: the DIC uptake by biology supported by upwelled nutrients, dropping DIC from  $DIC_1$  to  $DIC_2$ . The processes of ① and ② make up the *short-term effect* of upwelling (i.e., difference between  $DIC_2$  and  $DIC_0$ ); ③: the change brought about by air-sea  $CO_2$  gas exchange which continues towards the equilibrium with the atmosphere ( $DIC_3$ , whose level is determined by the amount of upwelled TA as well as by temperature); ④: the combination of both ② and ③ makes up the total *indirect effect* of upwelling (the difference between  $DIC_3$  and  $DIC_1$ ); ⑤: the *long-term* impact of upwelling on the level of surface DIC is the difference between  $DIC_3$  and  $DIC_0$ . Blue and red indicate two scenarios with different amounts of upwelled DIC relative to upwelled TA, but the same amounts of upwelled TA. Blue is for upwelled water with a deficit in additional DIC relative to additional TA whereas red is for an excess in DIC relative to TA.

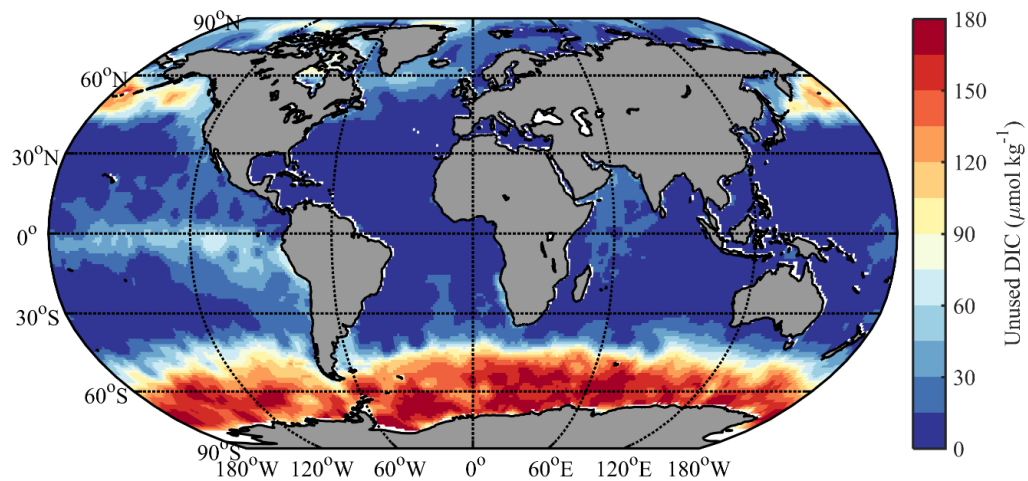


**Figure 7. Latitudinal distributions of calculated temperature effect on surface DIC.** Different columns show different basins (Atlantic, Indian and Pacific) and different rows show different calculated DIC variables. a, b and c show the observed surface DIC (black) and predicted DIC at SST of 27°C (red). d, e and f show the observed surface nDIC (black) and predicted nDIC at SST of 27°C (red). g, h and i show  $\Delta n\text{DIC}_{\text{temp}}$ , where  $n\text{DIC}_{\text{SST}=27}$  is subtracted from  $n\text{DIC}_{\text{obs}}$  to obtain the calculated temperature effect.

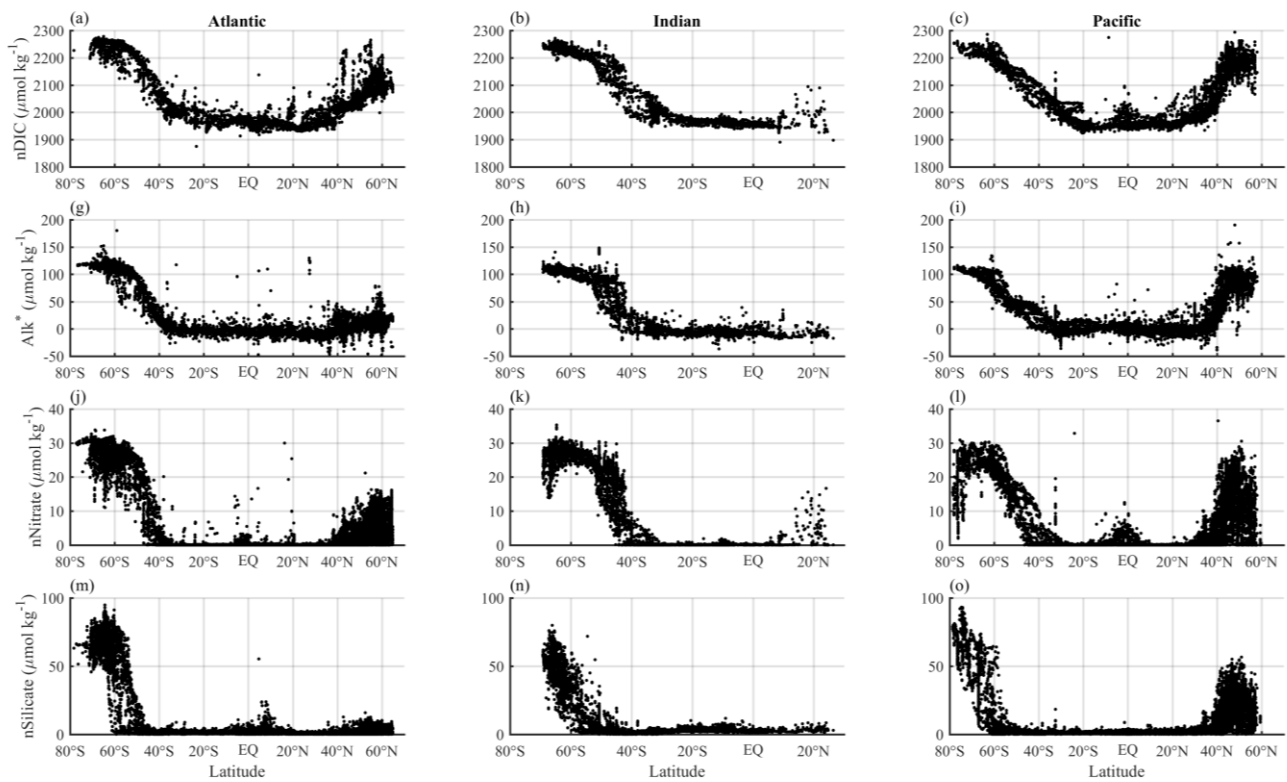


**Figure 8. Latitudinal distributions of calculated upwelling effects on surface nDIC.** Different columns show different sectors in ocean basins (Atlantic, Indian and Pacific) and different rows show different calculated effects on surface DIC. a, b and c show the short-term effect of upwelling ( $\Delta n\text{DIC}_{\text{upw\_st}}$ ), which is driven by the direct supply of DIC from deep water and subsequent change by biology in the Southern Ocean. d, e and f show the long-term effect of upwelling ( $\Delta n\text{DIC}_{\text{upw\_lr}}$ ), which is the difference between the observed nDIC value (determined mainly by the amount of upwelled TA, as well as by SST) and pre-upwelling nDIC value. The results were calculated from the three selected transects defined in Section 2.5.2.





**Figure 9.** Calculated potential impact of iron limitation on surface DIC. Different colors correspond to different amounts of “unused DIC”, calculated by Redfield ratio from observed residual phosphate.



**Figure 10. Latitudinal distributions of sea surface nDIC, Alk\* (Fry *et al.*, 2015), salinity-normalized nitrate and silicate in each ocean basin.**