

Response to anonymous Referee #2

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I would, on the behalf of all co-authors on this manuscript, like to thank the reviewer for a very thorough and thoughtful review. The comments regarding our description of a changing CO₂ chemistry in the ocean were in particular welcome, and revising these sections have led to a more readable manuscript. Below each comment from the reviewer has been restated, with my response below in bold font. All technical comments not specifically answered here have all been revised according to the reviewer's suggestions. Please also refer to the tracked changes version of the resubmitted manuscript for details on how the revisions were implemented.

Specific comments:

(1) P15553, L1-3; "This causes a decrease...": This will be wrong. First, the increase in surface fCO₂ leads to the decrease in surface ocean pH. Second, the increase in Revelle factor causes a slower decrease in surface ocean pH for a given increase in surface fCO₂ from the addition of CO₂ while it causes a faster decrease in surface ocean pH for a given increase in surface DIC from the addition of CO₂. Please check. **The reviewer is correct in this description of effects of changing the Revelle factor. We agree that this entire section was poorly worded, and our use of the term "buffer capacity" did not help the readability. The entire section has been revised for clarity, and we are confident it is now correct.**

(2) P15553, L11-14; "Thus local and...pH trends. The complex spatial variability...this hypothesis.": The hypothesis authors propose is unclear and needs more explanations. Do authors suggest that biological production and calcification cause the decoupling of fCO₂ and pH variations? In carbonate system, it is evident that the increase (decrease) in fCO₂ and decrease (increase) in pH at in-situ temperature nearly couples to each other, as the changes in pH and aqueous CO₂ concentration are thermodynamically coupled. The relationship between pH and aqueous CO₂ concentration (or pCO₂) changes with pH (or Revelle factor) but can be approximated by a linear function within the range of pH change of about 0.1, which is the range of pH variation in each biome described in this manuscript, under the contemporary

oceanic surface water condition. **The hypothesis is that local and regional changes in the ratio of DIC to alkalinity drive local and regional changes in the Revelle (buffer) factor. An altered spatial pattern of DIC:alkalinity is what will lead to a decoupling of the pH and fCO₂ trends, causing greater spatial variability in the pH trends. The text has been rewritten to clarify this hypothesis.**

(3) P15554, L22-25 and Figures 2-4: In Figures 2-4, the number of data points used to analyze the pH trend is significantly smaller than those used to analyze the fCO₂ trend. For example, in NPSTPS, the number of data point is N=299 for fCO₂ and N=246 for pH. Are these differences attributable to the “data fall outside the valid range for input data for the Lee et al (2006)”? These large differences might cause a significant inconsistency between the trends in pH and fCO₂. **Yes, these differences are due to the calculation of alkalinity. It is also true that this affects the trends, but it only significantly affects the results in the 1980s and is the reason so many biomes has no pH-trend for the period 1981-2011. The fCO₂ trends presented in Table 1 have all been calculated using only data that have a calculated pH value. A sentence has been added reflecting this. The figures showing the fCO₂ trends have been removed (at the suggestion of reviewer #1).**

(4) P15556, L15-17 and L20-21; “The pH change expected from...”: In principle, it is impossible to change pH and fCO₂ without changing either of alkalinity, DIC, SST, or salinity. I think DIC and Revelle factor are allowed to change here. **The reviewer is completely correct. We have revised this section to make our method and our assumptions more clear to the reader.**

(5) P15561, L16-18; ”In the biomes...”: I am not clear what is meant in this sentence. Discussion here should be improved. **This section has been simplified and revised for clarity. Some more discussion has been added for background.**

(6) P15563, L8-9: Decadal variation is not visible in Table 1. **This section has been revised for clarity. See also our response to reviewer #1 regarding this section (3.4).**

(7) Table 2 and 3: Unit should be specified. **Done**

(8) Both “carbon chemistry“ and “carbonate chemistry” have been used in the text to refer to the chemical equilibria of carbonate species in seawater. I would suggest using “chemistry of carbonate system” as used in P15550, L24 or just “carbonate system”, because “carbon chemistry” sounds to designate the chemistry of carbon compounds in general. **The text has been revised and we now consistently use the term “CO₂ chemistry”. See also the reply to reviewer #1 who had the same concern.**

Sincerely,

Siv K. Lauvset, on behalf of all authors