

Thank you for your careful reading and comments. We have replied your comments as follows. We show your comments and our replies in black and blue texts, respectively.

---

Referee #2 comments

This is my second time to read this paper. The concept of studying the coastal ocean acidification have great merit. I can tell that this version was improved especially by stressing the water depth issue. However, there are still some unclear things that need further clarification from authors.

1). First, the study ignores some basic inorganic carbon concepts. For example, the CO<sub>2</sub> system is the dominant acid-base buffer, the statement from Lines 148 to 153 and Lines 337 to 346 in session 4.3 is not accurate. K<sub>water</sub> does vary but variations of K<sub>1</sub> and K<sub>2</sub> values of CO<sub>2</sub> dissolution should be greater. The description of solubility pump was also inaccurate in Line 345. I still have difficulty in understanding why the authors assumed the T<sub>max</sub> (or T<sub>min</sub>) matched with pH<sub>min</sub> (or pH<sub>max</sub>). If you check your reference Bate et al 2014 (Fig. 2g), the summer pH was not consistently lower than the values in other seasons. Can the authors draw a scatter plot to show the relation between pH values and temperature? If there was strong negative correlation, it can clear this concern. Again, the authors should also remember that pH<sub>min</sub> may come from subsurface water (T<sub>min</sub>) whenever interpreting the pH<sub>min</sub> trend and should avoid using summer pH<sub>insitu</sub> to represent pH<sub>min</sub> (Line 437).

①> First, the study ignores some basic inorganic carbon concepts. For example, the CO<sub>2</sub> system is the dominant acid-base buffer, the statement from Lines 148 to 153 and Lines 337 to 346 in session 4.3 is not accurate. K<sub>water</sub> does vary but variations of K<sub>1</sub> and K<sub>2</sub> values of CO<sub>2</sub> dissolution should be greater. The description of solubility pump was also inaccurate in Line 345.

We corrected wrong descriptions about ocean carbon chemistry in the text (lines 148-153 and line 337-346 in the previous manuscript).

②> I still have difficulty in understanding why the authors assumed the T<sub>max</sub> (or T<sub>min</sub>) matched with pH<sub>min</sub> (or pH<sub>max</sub>). If you check your reference Bate et al 2014 (Fig. 2g), the summer pH was not consistently lower than the values in other seasons. Can the authors draw a scatter plot to show the relation between pH values and temperature? If there was strong negative correlation, it can clear this concern. Again, the authors should also remember that pH<sub>min</sub> may come from subsurface water (T<sub>min</sub>) whenever interpreting the pH<sub>min</sub> trend and should avoid using summer pH<sub>insitu</sub> to represent pH<sub>min</sub> (Line 437).

We checked the pH time-series of Fig.2 in Bates et al. (2014). Most of the stations except for three

sites in Iceland and Irminger Seas, Munida show common seasonal variations with Tmax (Tmin) matched with pHmin (pHmax), which supports our assumptions. To be limited in the discussion for the north hemisphere, the Iceland and Irminger Seas are located in higher latitudes, being largely influenced by biological processes, so such situations in summer are more affected by DIC decrease of the biological processes than by the seasonal variability of temperature. To describe this issue more clearly, we added the words “low- and mid-latitudes of the north hemisphere in open oceans (Bates et al. 2014)” in Section 2.1 in the present manuscript.

③ > Can the authors draw a scatter plot to show the relation between pH values and temperature? If there was strong negative correlation, it can clear this concern. Again, the authors should also remember that pHmin may come from subsurface water (Tmin) whenever interpreting the pHmin trend and should avoid using summer pHinsitu to represent pHmin (Line 437).

Since WPLC dataset only contains yearly minimum and maximum pH, we cannot plot seasonal variation of pH vs that of water temperature in a station. Instead of that, we plot the relationship between pH and temperature among the stations in each year as partly shown in Fig. A below. The relationship between pHmax and Tmin (red points) basically tends to show negative correlation, while the relationship between pHmin and Tmax (blue points) show no clear correlation. The pHmax supposed to be observed in the surface water in winter was more affected by temperature than by the other processes including biological ones. In contrast, pHmin data includes the data measured in both surface and subsurface waters, and could result in more complicated relations.

Note that the published data resolution of  $\pm 0.1$  in pH is not much sufficient for well resolving the pH fluctuation ranges of the pHmax and pHmin subcategories. However, the differences between pHmax and pHmin are resolved by this resolution and ranges of Tmax and Tmin groups are well different from each other. These features are not inconsistent with our assumption.

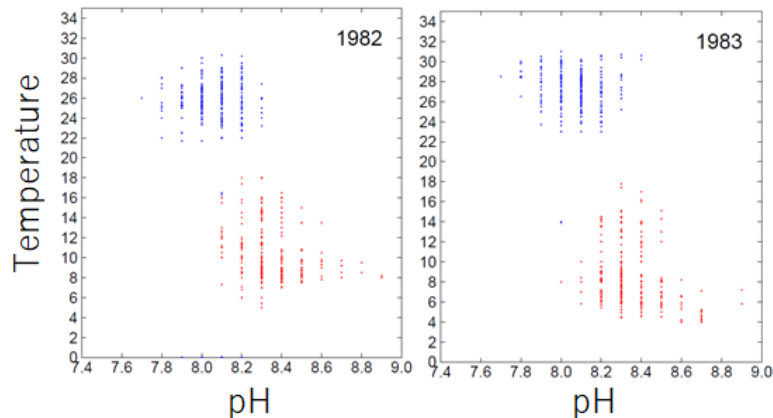


Fig. A Examples of the relationship between pH and temperature in 1982 and 1983.

2). As the authors also agree with that removing the sites with high variability (from 1127 to 1289) actually removed a big fraction of sites that have large local process (for example, heavy phytoplankton bloom, or freshwater discharge change). With this data treatment, the discussion part (in my opinion) is circular logic because the only process left is global process (warming and anthropogenic CO<sub>2</sub> intrusion) after removing the local process. Then the authors discussed and found that global process can explain the pH trends. Even though the authors want to keep the current strategy, this impact of QC process 3 should be included in discussion part to remind the readers.

The sites with high variability due to large local processes should be removed by the three-step quality control procedures. Therefore following from your suggestion, we have added the reminder sentences about our QC process 3 in Section 4.3 as below, before the discussion of the global effects in Section 4.4. It is very helpful to make the descriptions more readable. Thank you for your suggestion.

3). Lines 348-353, the explanation is still very weak. Do you divide the water column (inside one site) into autotrophic or heterotrophic by depth: surface is autotrophic, while subsurface is heterotrophic? Your definition of autotrophic and heterotrophic condition should be included in text. Otherwise, the readers will feel puzzled as what happened to me when trying to understand why the negative correlation between pH and TN trends can show the metabolic state.

Sorry for unclear definition of the term autotrophic/heterotrophic. We consider two situations in autotrophic/heterotrophic separation. In most coastal stations with low terrestrial input, surface water becomes autotrophic while subsurface water becomes heterotrophic. In estuary waters and waters near urbanized area with high terrestrial input, however, decomposition of terrestrial POC often overcomes local primary production, and as this result, both surface and subsurface waters become

heterotrophic (e.g., Kubo et al., 2017). If we assume that input of terrestrial POC varies in proportion to that of terrestrial N, we can expect that most of these stations show heterotrophic response against N variation. We add some description about autotrophic/heterotrophic separation in Japanese coastal waters in Lines 363-371.

4). Discussion part did not really discuss what processes should be responsible for the observed basification rate (~25% sites).

Figure 14 suggests that at least several WPCL stations are located in heterotrophic waters (i.e., estuary or water near urbanized area), and decreased TN input have caused basification is such stations. For other stations, however, we must agree that we had not yet make fully convincing explanation for the basification. One possible cause is change in residence time of coastal water (e.g., artificial change of closure rate of an inlet), although we have no hydrography data to probe this assumption so far. We add these discussions in line 473-483.

5). Fig. 8 is one of the key take home figures. Can you change the color map order? I mean to put the insignificant trends in the first layers, and largest trend on the top. The insignificant trends should also be marked in the figure. The authors should also note in the MS that XX% of the trends are insignificant.

The color map order in Fig. 8 was changed to put the significant trends on the top. Also we have added the histogram with the sites having significant correlations for the calculated regression lines as reference. The percentage of the trends in Fig. 8 was not attached because it is likely to emphasize the significance and insignificance of pH trends. We would like to treat pH trends equally. Readers can see the degree of the number of significant trends in Fig. 7.

6). Equation 3 is wrong. Please check through whether your pH<sub>25</sub> was correctly calculated.

Yes, this part was a mistake. We confirmed that the mistake was just only the description. All of the pH<sub>25</sub> values has been correctly calculated in the previous manuscript. We corrected the description in the present version. Thank you very much for your indication.

7). Lines 316, how did you get the percentage of 10 m? If you can pinpoint the 10 m depth value, why do not you remove this data and see what the trend is?

The WPCL dataset does not discriminate between surface (0.5–2 m) and subsurface (10 m) data when

calculating the annual maximum and minimum  $\text{pH}_{\text{insitu}}$ , although monitoring depths were fixed throughout the monitoring period at all the sites. But the WPCL dataset provides the temperature data with the observed depth. Therefore, assuming that pH values were measured at the same depth as temperature, we estimated the percentage possibility that samples were collected at 10 m depth for the quality-controlled datasets with 1481, 1127, and 289 sites.

Actually our previous manuscript did not provide such information in detail. We have improved the part in Section 4.2 by describing the additional information. Thank you for your comment.

### **Some minor comments**

1). Line 34, what is the “large deviation”?

We have added the numerals to represent “large deviation” in the present abstract, as follows.

>The annual maximum  $\text{pH}_{\text{insitu}}$  decreased at an average rate of  $-0.0024 \text{ yr}^{-1}$ , with relatively large deviations ( $0.0042 \text{ yr}^{-1}$ ) from the average value.

2). Lines 194-198, not only biological process, other physical process may be more severe in summer than winter.

We have added the sentence “other physical processes” as follows in Section 2.2.

>The absolute values of the correlation coefficients for the  $\text{pH}_{\text{insitu}}$  were slightly lower than those for temperature for each corresponding pair of sites (Figs. 4 and 5), and might reflect the fact that  $\text{pH}_{\text{insitu}}$ , but not the water temperature, is subjected to strong forcing by coastal biological processes and other severe physical processes in summer, which causes the  $\text{pH}_{\text{insitu}}$  to vary on the short-term.

3). Lines 201- 203, unclear. The purpose of Fig. 5 is unclear. A good correlation should also be there if you paired  $\text{pH}_{\text{min}}$  and  $\text{pH}_{\text{max}}$  or whatever two parameters.

We have added the purpose of Fig.5 in Section 2.2, as follows.

>In the final step of the quality check procedure (step 3), we removed all the time sequences with weak and insignificant correlations for temperature and  $\text{pH}_{\text{insitu}}$  (Fig. 5), since we considered that the monitoring sites having both significant correlations for water temperature and  $\text{pH}_{\text{insitu}}$  were reliable.

After this final step, 289 sites remained.

Since the explanation of Fig. 5 has been added, we did not put additional figures in this manuscript.

4). Lines 422-423, unclear. Where did the  $-0.0003 \text{ yr}^{-1}$  or  $-0.0005 \text{ yr}^{-1}$  come from? Fig. 7 e-f has no this information.

We checked this parts and realized that only “ $-0.0005 \text{ yr}^{-1}$ ” was actually a mistake. We have fixed the part as follows.

>The differences between the  $\text{pH}_{\text{insitu}}$  and the corresponding  $\text{pH}_{25}$  trends in summer ( $-0.0004 \text{ yr}^{-1}$ ) and winter ( $-0.0010 \text{ yr}^{-1}$ ) can be partly explained by the difference between the decrease in the pH trends in summer ( $-0.0003 \text{ yr}^{-1}$ ) and winter ( $-0.0008 \text{ yr}^{-1}$ ) arising from the thermal effects.

The numbers of “ $-0.0003 \text{ yr}^{-1}$ ” and “ $-0.0008 \text{ yr}^{-1}$ ” was calculated from change from 8.0150 to 8.0147 in summer and from 8.2598 to 8.2560, which indicates the difference between in the pH trends in summer ( $8.0150-8.0147=-0.0003$ ) and winter from ( $8.2568-8.2560=-0.0008$ ), arising from the thermal effects. “Fig. 7 e-f” was removed in this sentence. Thank you for your careful checking.

5). Linea 60-61, Please change into another easy way to understand the sentence. The same with Lines 77-78. Please check through the MS to avoid this kind of description.

We have improved this part. As for the description of Line 77-78 in the previous manuscript, we guess that you feel the words “mesohaline” and “polyhaline”. We checked Duart et al. (2013) and actually they used those words. It means that it would be proper for their environments. We have still used the words in the present manuscript.

7). Line 446, pH insitu TREND.

We have corrected this part. Thank you for your indication.

6). Line 447, how many sites were heterotrophic based on your definition?

The scatter plots in Fig 14 are very sparse. If we try to count the number of the sites clearly showing heterotrophic, the number could be 8 points as marked in blue dotted circles in Fig. B (new Fig.14). We recognized that the all sites were measured in the same bay, Shimotsu Bay, Wakayama Prefecture. The bay seemed to change volumes of the terrestrial input during the monitoring period and could result in the negative correlation of the whole distributions. By considering this result, we have

removed Table 3 in the previous manuscript and changed the description in Section 4.4.2 of the discussion about the relations between TN and the  $\text{pH}_{\text{insitu}}$  trends shown in Fig. 14.

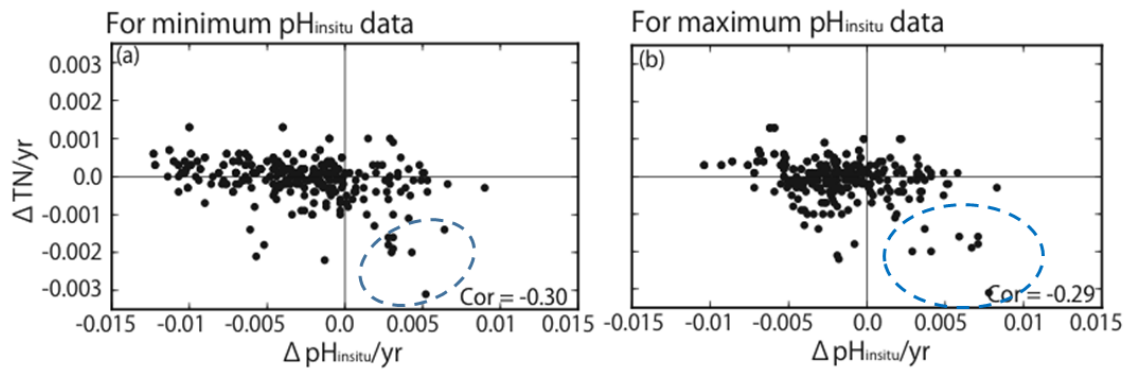


Fig. B Same as Fig. 14 in the revised manuscript