Dear the editor, Dr. Jean-Pierre Gattuso

Thank you for your comments. You picked up three points for our previous manuscript. We have revised it according to them. Below we show your comments and our replies in black and blue colored texts, respectively. We also inform you that we have attached the revised article marked up in blue, in the author's response. You and reviewers can see which parts in the revised manuscript has been improved.

1) Methods should be provided comprehensively, especially because some of the literature cited is in Japanese.

We have added the additional information on type of electrode used, buffers, method of calculation, time between sampling and measurement, and several other issues in the revised manuscript (Section 2.1).

2) Lumping together surface and 10 m data could very problematic in shallow estuaries as pH can be vastly different, even 10 m apart.

The sampling depth at each site was fixed throughout the monitoring period, and the trend at each site was calculated from the data measured at the same sampling depth during the period. In addition, we examined at what depths our monitoring data were sampled and described the results in Section 2.1 in the revised manuscript.

The information of the sampling depths in $\mathrm{pH}_{\mathrm{insitu}}$ is not available at each site in the data downloaded from the NIES website (www.nies.go.jp/igreen; http://www.nies.go.jp/igreen/md_down.html), but the information of the sampling depths is available for temperature. Assuming that pH values were measured at the depth as same as temperature, we estimated percentages of the possible deepest sampling depths for the quality-controlled datasets with 1481, 1127, and 289 sites. The percentages of the possible deepest sampling depth of 10 m are 13%, 13%, and 15% in the cases of 1481, 1127, and 289 sites, respectively.

We must agree that our annual minimum pH data contain both surface and subsurface water measurements, while annual maximum pH data hardly include subsurface data. However, we keep our results of the annual minimum pH in the revised manuscript, because the fact that the annual minimum pH data show a similar trend with that of the annual maximum pH suggests, by itself, that the long-term changes of the biological process must not be the main cause of the observed trends, as this process may affect pH in an opposite way between surface and subsurface waters. We consider that the global phenomena such as the atmospheric CO_2 increase and the relevant warming would be related to the observed trends of annual maximum and minimum pH.

We have added a new section (4.3) for the relevant discussion and modified the abstract based on the additional discussion. Thank you for your critical notice.

3) pH scale: I understand that early monitoring was on the pH scale but I wonder why it is still measured this way. Both a glass electrode (NBS) and spectophotometric (Mosley et al., 2004, Marine Chemistry) method could be used during 1 year and then all measurements subsequently made on the total scale. If the spectrophotometric method cannot be used, one could also calibrate the glass electrode using seawater buffers, although it would less satisfactory.

One of the reasons of this situation is that monitoring for water quality control (the WPCL program) and for global warming studies are controlled by completely different departments within Japan Ministry of Environment, and hence people of the WPCL program had not realized that their data are also meaningful to assess ocean acidification, until we did this study. We have already contacted people of the WPCL program and started negotiations to add total-scale pH measurement at (at least) some WPCL sites. We consider that publication of this paper will definitely support this process.

We believe that all the comments have been well addressed in the revised manuscript. We look forward to your positive decision.

Sincerely yours,

Miho Ishizu, Yasumasa Miyazawa, Tomohiko Tsunoda, Tsuneo Ono

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

In this study, the authors estimated the long-term trends of pH in Japanese coastal waters from 1978 to 2009. In 70 to 75 % of the monitored sites, they found acidification trends while they obtained basification trends in 25 to 30 % of the sites. The authors tried to interpret the spatio-temporal patterns in pH based on the in situ pH, temperature and total nitrogen data. The paper's idea is very important taking into consideration the increasing need of a continuous OA monitoring, particularly in coastal areas where OA effects on marine ecosystems could be exacerbated due to local pressures. However, I do have some major concerns about the pH data and the methodology used to get it:

1) Are the authors calibrating the glass electrode with TRIS solutions for seawater measurements? I'm not against NBS standard buffers for experimental essays or to check the in situ variations of pH in coastal stations to assess the pollution there or whatsoever, but pH potentiometric measurements with NBS calibrations are strictly not recommended for seawater monitoring, particularly for long term surveys (climatic survey) where the pH uncertainty should be around 0.003 pH unit. Moreover, this technique's results are not comparable with the ones adopted for seawater elsewhere and mentioned in the entire text (i.e. Bates et al., 2014, etc.). Please check the following useful links for the recommended strategies to better study the OA in open and coastal areas for long or short periods: - http://goa-on.org/documents/general/GOA-ON_Implementation_Strategy.pdf - http://goa-on.org/resources/sdg_14.3.1_indicator.php

Thank you for your information of the links to the monitoring strategies recently recommended by GOA-ON for studying the ocean acidification. We understand that the NBS standard buffers are not appropriate for long-term monitoring focusing on climatic survey as we described in the introduction (Chapter 1). However, widespread use of seawater-scale pH buffers started in 1994 (Dickson and Goyet, 1994). This means that we MUST use NBS-scaled pH data if we want to analyze interannual variation of pH with time scale longer than 25 years.

The WPCL program fully realizes uncertainty of their seawater pH data, and this is because they set their permissible range of pH data as 0.1 pH. This precision is, of course, far insufficient to assess a temporal trend of a single station. We, therefore, focus on statistical characteristics of all derived trends instead of assessing each single trend. We demonstrate in section 4.1 that even if each trend at each site involves non-negligible measurement error, evaluation of whole statistical characteristics of the population (group) is feasible. To clarify the punch-line of our study, we have added new descriptions, especially in Section 4.3.

In summary, we propose here one practicable way to extract some meaningful information from past NBS-scale pH datasets. We believe this approach more useful than just revoking all past NBS-scale pH data.

2) The authors did not explain why they calculated trends for minimum and maximum pH values? Why didn't you calculate the trends based on the annual average pH instead of doing it for the minimum and maximum values separately?

As described in Section 2.1, the WPCL pH dataset contains only the annual minimum and maximum pH data without any information of the detailed measurement time. We assume that basically the annual minimum and maximum represents summer pH of 10m water and winter pH of surface water, respectively. As these two valuables represent pH trends at different water depths, we did not calculate average of these values. Moreover, the situation would be different at each site in summer and winter; therefore we calculated trends for minimum and maximum pH values separately. For example, in summer, biological activity would be more active but in winter, winter mixing would be more active. Such situation should be totally different from each other. Our analysis results of thermal effects on the trends are consistent with our assumption that annual minimum and maximum pH were measured in summer and winter, respectively. In the present version, we added the discussion of "Effects of sampling depth" in Section 4.2.

3) The authors are relying on this methodology: ISO10523 (https://www.iso.org/standard/51994.html) mentioned in P7, L135. This method is adopted mainly for freshwater measurements. Could you please provide more information about the JIS Z8802 standard protocol (2011). It is apparently accredited in JIS list (file:///C:/Users/user/Downloads/jis-japanese-industrial-standards.pdf; p397) but I couldn't find its details.

ISO10523 is the methodology mainly adopted for freshwater measurements, but as we mentioned in the reply 1), this method had been adopted also for seawater measurement until 1994. JIS Z8802 is Japanese standard protocol that is formally compatible with ISO 10523. WPCL adopted this methodology for seawater pH measurement as it has launched in 1970, and they had not changed the methodology to maintain continuity in the measurements.

We are now proposing to Japan Ministry of Environment to add pH measurements with present standard methods (ca. Dickson, Sabine and Christian 2007) in some coastal stations, but so far, only available dataset is the presented ISO10523-based dataset.

One of the reasons why it is still measured by this way is because the monitoring for water quality control (the WPCL program) and for global warming studies are controlled by completely different departments within Japan Ministry of Environment, and hence people of the WPCL program had not realized that their data are also meaningful to assess ocean acidification, until we did this study. We have already contacted people of the WPCL program and started negotiations to add total-scale pH measurement at (at least) some WPCL sites. We consider that publication of this paper will definitely support this process.

4) Any inter-calibration essays have been conducted to compare the pH results between the licensed operators/ labs?

We have found no information about inter-calibration essays between the licensed operators/labs in the WPCL program. We suppose that inter-calibration essays have not been conducted. To check the data quality by ourselves, we compared the pH trends measured by different licensed operators (see Section 2.2 and Fig. 6) and processed the data selection by the multi-step quality checking procedure.

5) How did you correlate the pH trends to biological processes? Did you check the correlation between pH and biological parameters measured in parallel at the monitored sites?

Since the pH data under the WPCL program were measured for monitoring the pollution control, the proper biological parameters were basically not included in the targets of the monitoring. Only the Total Nitrogen (TN) data are available from the data archive for a period from 1981 to 1995. We thus use them for the relevant discussion in section 4.2.2.

As mentioned by other reviewer, pH minimum substantially represents summer pH of subsurface (10m) water, so this trend shows negative correlation with that of TN. This relationship had partly offset anthropogenic-CO_2 induced pH decrease, because TN loadings to Japan coastal waters had significantly decreased in recent years.

6) How the dominance of heterotrophs or autotrophs might affect the pH in coastal waters? How did you related these to your data? Based on what have you suggested that these waters are oligotrophic? Many statements through the text are so weak and need to be better justified.

To consider possible causes leading to contrast in acidification and basification trends among the sites, we assume that the eutrophication enhances acidification (basification) in the heterotrophic (autotrophic) sites (Duarte et al. 2013). We show that the assumption is partly confirmed by checking the negative correlation between pH and TN trends (Table 3). Figure 14 also indicates that some of the sites involve combination of negative (positive) pH and positive (negative) TN trends, suggesting the heterotrophic condition at the sites. The autotrophic condition is suggested by the sites shown in the second and fourths quadrants (Fig.14). We have modified the relevant descriptions to more elucidate this point.

Figures: The style of many figures is very confusing, also their captions! For Fig. 6 for example, the same-color lines indicate the pH values taken for the same place and the same operator, but one for the annual maximum and another one for the annual minimum pH? This was understood from the Fig. 6 caption, but not from the text. Please rephrase.

We have added some explanations about the annual and minimum pH_{insitu} data in caption of Fig. 6. The captions of other figures were also reconsidered. Thank you for your indication.

Tables: Table 2: How significant were these correlations? Why you didn't present this table the way you did in Table 3?

We have added information about the significance in Table 2.

Replies for the specific comments in your attached document.

L81-L85: Too much info. about only one region "Chesapeake Bay, US".

This part has been removed in the present version.

L310-L312: This sentence is very confusing!

We have revised this part in the present manuscript as follows.

"In the waters where primary productivity is predominantly to organic decomposition (i.e., autotrophic water), N increase will enhance primary production and hence decrease DIC, causing basification. In the adjoining waters of this autotrophic watermass, however, N increase will arise increase of POC transport from the autotrophic watermass, and this leads increase of POC-decomposed DIC (i.e., hetelotrophic water) and cause acidification (e.g., Sunda and Cai 2012; Duarte et al. 2013).”

L314-L317: These statements need to be related to your data.

This part is surely related to the analyses focusing on the distributions of the whole pH trends. We modified it to more emphasize our viewpoint.

L332-L335: In the rest of this section, you have assessed the thermal effect on pH trends by normalizing the pHinsitu to pH25. How did you test the second assumption related to the coastal carbon cycle?

We have modified the relevant description to clarify the logical structure. We first examine the thermal effect (D (T)) targeting the whole populations of pH_{insitu} trends, and then check ocean acidification effect (DIC (AirCO2)) for the populations of pH_{25} trends after normalization. Variability inside of the trend populations comes from the regional differences in the trends, which would be affected by other factors.

L356: The captions in your figures need to be clearer, so each color should be better assigned to a specific parameter. Also, please replace "deg" for Temperature by "°C".

The captions in the all figures were reconsidered, being improved. We have replaced “deg. C” for "°C" in the present manuscript.

L380-L381: Mixing the values/trends of both minimum pHinsitu and maximum pHinsitu is very confusing through the entire text. This needs to be improved.

We have unified to use the ‘trends’ in the present manuscript.

L407-409 how the dominance of heterotrophs or autotrophs might affect the pH in coastal waters? How did you relate these to your data? Based on what have you suggested that these waters are oligotrophic? These statements are so weak and need to be better justified.

We simply speculate possible existence of the heterotrophic and autotrophic sites according to Figure 14. Also see our reply comment to item 6).

L414 : This is weird. Do you mean oligotrophic and eutrophic waters?

We mean heterotrophic and autotrophic conditions for categorization of each site. A heterotrophic site shows a negative (positive) pH trend by responding to an eutrophication (oligotrophication) trend, and vice versa for an autotrophic site.

L451: I think you mean the trophic state index of the waters.

Yes, our categorization of heterotrophic/autotrophic sites is based on basically same terminology.

Please also note the supplement to this comment:
://www https.biogeosciences-discuss.net/bg-2019-150/bg-2019-150-RC1-supplement.pdf

Thank you for careful checking of our manuscript. It was very helpful for improving the description.

Interactive comment on Biogeosciences Discuss., https://doi.org/10.5194/bg-2019-150, 2019.

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

This study determined the long-term trends (from 1978 to 2009) of pH in Japanese coastal waters. They found that both positive and negative pH trends distributed along Japanese coasts. Majority sites have decreasing trends, which is consistent with open ocean. The authors then discussed the impact of warming on the spatial distribution of pH trend and speculated the potential impacts from other processes. Overall, this study presented a very good dataset, but I have less confidence in the methodology in order to derive a robust story.

1) After finishing this MS, I am still not sure what kind of data this study used. First, this study did not show any information about salinity, so, the dataset was from freshwater, brackish water or sea water? Can you give more information about the pH measurement? Was pH measured under in-situ temperature, or the samples were taken back to lab and measured at 25_C? Where did pH minimum or maximum come from? It seems like the min or max values were from entire water column in each site based on Lines 137-138 "NIES gathered all pH data measured at each site and calculated annual minimum and maximum pH". However, the respiration was more powerful in decreasing pH comparing to anthropogenic CO2 intrusion (Cai et al, 2011), so, the pHmin generally came from bottom water, while the maximum came from surface water (without considering other local processes). In other words, pHmax and pHmin totally represented the values from different water depth, so, all trend interpretation should be related to the water sources.

   We have modified the relevant descriptions to include more detailed information about the measurements by the WPCL program.

   >The dataset was from freshwater, brackish water or sea water?

   Over 90 % of the data comes from coastal sea water, while less than 10 % comes from estuary. We carefully extracted the measurements in sea water from the whole data archive. To clear this matter, we have changed the term "coastal waters" to "coastal seawater" in the present manuscript. Readers also can get the information of latitude and longitude, measured pH data, in the supplement data.

   >Was pH measured under in-situ temperature, or the samples were taken back to lab and measured at 25_C?

   The pH was measured under in-situ temperature. We have analyzed pH_{insitu}, which means pH at ambient sea surface temperature. We have added the word, "in-situ" to clarify this point in the revised version, as follows:

   "Similarly, analysis of the Hawaii Ocean Time-series (HOT) (Dore et al. 2009) and the Bermuda Atlantic Time Series (BATS) (Bates 2007) showed that pH at ambient (in-situ) sea surface temperature (pH_{insitu}) decreased by 0.0019±0.0002 and 0.0017±0.0001 yr^{-1} from 1988 to 2007 and from 1983 to 2005, respectively."

   >Where did pH minimum or maximum come from? It seems like the min or max values were from entire water column in each site based on Lines 137-138 "NIES gathered all pH data measured at each site and calculated annual minimum and maximum pH". However, the respiration was more powerful in decreasing pH comparing to anthropogenic CO2 intrusion (Cai et al, 2011), so, the pHmin generally came from bottom water, while the maximum came

from surface water (without considering other local processes). In orther words, pHmax and pHmin totally represented the values from different water depth, so, all trend interpretation should be related to the water sources.

As we mentioned in Section 2.1, “at each basic survey, water samples were collected at several depths (0.5 and 2.0 m) below the surface for all sites, and 10 m where bottom depth was more than this) for times a day to cover diurnal variation.

The sampling depth was each site was fixed throughout the monitoring period, and the trend at each site was calculated from the data measured at the same sampling depth during the period. In addition, we examined at what depths our monitoring data were sampled and described the results in Section 2.1 in the revised manuscript.

The information of the sampling depths in pH_{insitu} is not available at each site in the data downloaded from the NIES website, but the information of the sampling depths is available from temperature. Assuming that pH values were measured at the depth as same as temperature, we estimated percentages of possible deepest sampling depths for the quality-controlled datasets with 1481, 1127, and 289 sits. The percentages of the possible deepest sampling depth of 10m are 13%, 13% and 15% in the case of 1481, 1127 and 289 sites, respectively.

We speculate that some monitoring sites should be forced by biological activity, but some not. The situations depend on time and place. To remove extreme data that would be affected by significant biological activity, we adopted the strict quality control procedures and then used the time-series of pH that passed the strict criteria in the analysis. We consider that the remaining moderated max-min variations in the data after QCs were mainly caused by the seasonal variations of the thermal effect (D(T)).

However we must agree that our annual minimum pH data contain both surface and subsurface water measurements, while annual maximum pH data hardly include subsurface. We keep our results of the annual minimum pH data in the revised manuscript, since the fact that annual minimum pH data show a similar trend with that of the annual maximum pH suggests, by itself, that the long-term changes of the biological process must not be the main cause of the observed trends, as this process may affect pH in an opposite way between surface and subsurface waters. We consider that the global phenomena such as the atmospheric CO2 increase and the relevant warming would be related to the observed trends of annual maximum and minimum pH.

We have added a new section (4.3) for the relevant discussion and modified the abstract based on the additional discussion. Thank you for your critical notice.

2) I am not sure how pHmin or pHmax could be representative of the average pH situation in specific year. This min or max values have a good chance to be affected by extreme events, for example, phytoplankton blooms or heavy flooding events. I am not sure whether the trends or pHmin or pHmax can represent the overall pH change rates in that sites. However, it did represent the variation of pH in each year. Did the authors find the difference between pHmax and pHmin change (increase of decrease) over time? This examination can also help derive useful information about CO2 chemistry data change over time, because extreme values matter. Here are a few references the authors may need.

Fassbender, A. J., K. B. Rodgers, H. I. Palevsky, and C. L. Sabine (2018), Seasonal Asymmetry in the Evolution of Surface Ocean pCO2 and pH Thermodynamic Drivers and the Influence on Sea-Air CO2 Flux, Global Biogeochemical Cycles, 32(10), 1476-1497.

Landschützer, P., N. Gruber, D. C. E. Bakker, I. Stemmler, and K. D. Six (2018), Strengthening seasonal marine CO2 variations due to increasing atmospheric CO2, Nature Climate Change.

Thank you for your suggestions. But we have avoided analyzing the difference between pHmax and pHmin because differencing two variables could lead to enhancement of data errors. As for the references mentioned above, those references could be useful to mention seasonal variability of pCO_2 and the asymmetric response of air-sea CO_2 flux. We have added relevant new descriptions in Section 4.3.1.

3) The authors did a lot of work in quality control by step 1, 2, 3. In my opinion, step 1 is strict enough. Removing the outlier points instead of entire time sequence can keep all 1481 sites. I do not agree with "step 3" to get rid of "random errors". The authors removed the time sequences whose pHinsitu stdev> the average stdev of 1127 sites. With this process, you actually removed all the sites that have a high stdev, which may have nothing to do with random errors. For the sites with a strong biological activity, or a site that is easy affected by the river discharge, they all have large stdev. However, this is their feature, but not caused by "error". With this operation (step 3, and extra step from Line 197-198), you have already excluded all the sites that were affected by B(T,N) and Alk (S). Thus, only the sites with mild hydrological or biological variation, and strong thermal impact were left. I fail to see why this process was included in this manuscript.

We keep this form by using the quality control (QC) steps 1, 2 and 3, because if the data include a lot of processes resulting in the large variability deviating from the average pH, the discussion would be more complicated. So it would be better to keep this form and to use the data only with moderated variations.

We partly agree with your comment: "you have already excluded all the sites that were affected by B(T, N) and Alk (S)". But even after applying the all QC steps, the trend distributions involve some variability around the averages (Figs.7e and 7f). We thus discuss possible variations which might be caused by area-dependent B(T, N) and Alk (S) effects. Since there is no available data of ALK(S) for checking our data, we do not much discuss the influence of Alk (S) (Section 4.3). As for the discussion of B(T, N), we suggest coastal acidification/basification mechanisms by comparing the pH trends and TN ones (Section 4.3.2 and Fig. 14).

4) I have difficulty in understanding why the authors compared pHmin with Tmax or pHmax with Tmin across the maintext. Line 142 "the pH values were lowest in summer and highest in winter".

To make readers more clearly understand this thermal effect, we have added a term "$H_2O \leftrightarrow H^+ + OH^-$" in this part as follows.

"Previous studies have reported negative correlations between seasonal variations in pH and water temperature, mainly because of changes in the dissociation constant in dissociation equilibrium ($H_2O \leftrightarrow H^+ + OH^-$).”

5) My concern is the pH value was also impacted by biological activities (photosynthesis and respiration). Thus, high temperature in summer cannot guarantee low surface pH, when the photosynthesis was very strong. Please check through the maintext.

The QC steps effectively work to exclude pHmax and pHmin data caused by the extreme biological effects. Max-min pH variations in the remaining data after QCs are considered to be caused by mainly the seasonal thermal effect as explained above. In the revised manuscript, we have improved the descriptions to clarify our viewpoint about the possible influences on the $\mathrm{pH_{insitu}}$ trends in coastal sea waters (Section 4.3, 4.3.1, 4.3.2).

6) Based on the 289 sites, the authors derived two sets of pH trends: -0.0014±0.0033 and -0.0024±0.0042 yr-1 for pHmin and pHmax, respectively. Are these two trends significant different? A paired t-test is needed here.

Yes, the two trend averages are significantly different from each other. We have added the results of the paired-t test and demonstrated whether these trends significantly different or not in Section 4.1. That suggests the seasonal difference in the thermal effects on $\mathrm{pH_{insitu}}$ trends (Section 4.3.1). We have added this information in the revised manuscript as well.

7) Fig. 7 included all the trends across the 289 sites, both significant and insignificant. Can you only include the significant trends? What is the average value of significant trends? Based on the discussion in section 4.1, the threshold of significant pH trend (caused by measurement precision only) is _0.002 yr-1. Other variation of pH (i.e. caused by local processes), should also impact the detection of significant trends. This can be further examined by previous comment (#5).

If we select the significant trends alone, the trends with larger magnitudes remain. A left panel of Fig. R1 shows a histogram of some pH trends including both statistically significant and insignificant values, representing a shape like the normally distribution with a negative shift. A right panel shows a histogram of selected trends with statistical significance for ΔpH not zero, indicating that the trends with $\mathit{\Delta pH} \fallingdotseq 0$ disappear in this case and the relatively large negative pH trends tend to remain in the distribution. These figures illustrate dominant existence of the negative trends, but this procedure results in deleting the trends of monitoring sites with $\Delta\mathrm{pH} \fallingdotseq 0$, which must actually exist.

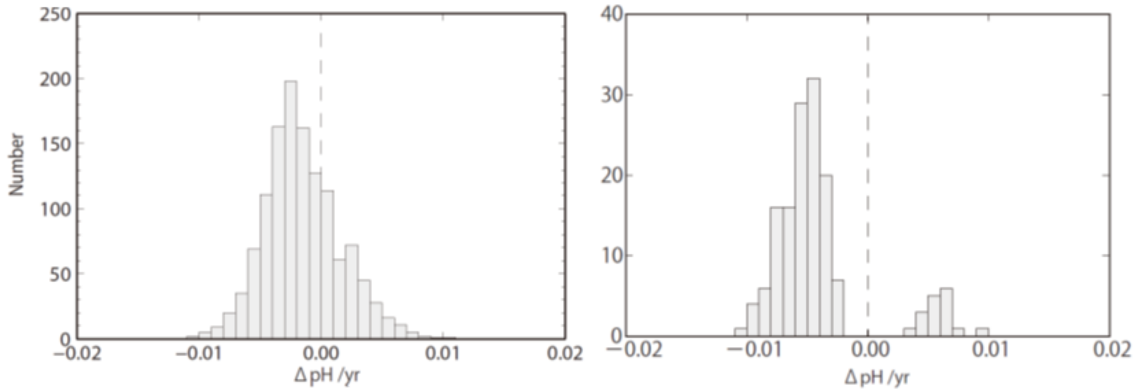

Figure R1. Examples of histograms of pH trends both with and without statistical significance for ΔpH not zero (left) and only with significance (right).

8) The discussion between pH change and heterotrophic or autotrophic is very weak. In addition, I still think the 289 sites have already excluded the stations that have strong biological activities.

   We suggest possible heterotrophic/ autotrophic pH responses to eutrophication/ oligotrophication inferred by relations between the pH and TN trends shown in Fig.14. The negative correlation evaluated from Fig.14 further suggests dominance of the heterotrophic sites. Since we consider that this result alone is not sufficient for rigid confirmation of our consideration, we modified abstract and relevant descriptions. However, we still note that variability remaining in the pH trends after the QC steps suggests coastal acidification/basification processes, which could be affected by the heterotrophic/ autotrophic conditions of each site.

9) Do the salinity or water discharge change support the conclusion in Line 431?

   Salinity data and/or information of water discharges suitable for cross-comparison with the pH data used in this study are not available. It is difficult to directly answer this question.

Replies for your comments in the manuscripts:
>There is also some unclear description in maintext, figure caption, and legend. 1.

1. It should be 289 sites (under current version) in the abstract, but not 1481.

   Following your comment, we have changed the part from "1481 monitoring sites" to "289 monitoring sites".

2. How did you get the mean value in Line 165? Average of pHmin and pHmax?

   For clarification, we have improved this part with the words "average of minimum and maximum pH_{insitu}", as follows, in the present manuscript (Section 2.1).
   "We then excluded time sequences with outliers, defined as sites with data points
   that were more than three standard deviations from the average of minimum and

maximum pH_{insitu} for each year."

3. Lines 206-211, what is the "standard deviations of pHinsitu trends"? The legend and caption of figure 6 is very confusion. A comment here (in my opinion), this MS studied the trend instead of absolute value. So, the site crosscheck may have very minor impact on the final results.

   We have modified legend and captions of Fig. 6. At first, we have used the thin and bold lines to discern minimum and maximum pH_{insitu} data at each monitoring station in this figure. We have added the relevant description to the caption.

4. Lines 232 to 235, the reference here reported pH25, so this comparison should be moved to later section.

   We moved this part to the later section (Section 4.2.1).

5. Lines 319-321, I have difficulty in understanding "both DIC (B (T, N)) and Alk (S) are difficult to have general trends that covered all monitoring sites, because factors that control these variables have no mutual trends all over the Japan coast".

   The overall pH_{insitu} trends shown in Fig.7 were governed by the thermal effect, D(T), and ocean acidification, DIC(Air CO2), because the all monitoring sites are equally affected by these global factors. In contrast, DIC (B(T, N)) and Alk (S) were significantly affected by local situations, depending on regions. The variability among the pH_{insitu} trends, which are characterized by standard deviations of the trends populations, were caused by such local factors. To emphasize this viewpoint, we have modified abstract and all relevant descriptions.

6. Lines 321- 324, why did "same trend of B (T, N) leads opposite trends of DIC (B (T, N)) between autotrophic and heterotrophic ocean"? How do you define the "autotrophic and heterotrophic" here?

   We consider that the heterotrophic conditions are defined as the increasing (decreasing) response of DIC to eutrophication (oligotrophication), and vice versa for the autotrophic condition. For example, the sites shown in the second quadrant of Fig.14 means negative pH trends (increasing DIC) involve positive TN trends (eutrophication) at those sites. But we realize that this consideration is based on some assumptions that not well confirmed by only the available data from the WPCL data archive. We modified abstract and conclusion for clarification.

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6. Line365, a typo? from 8.2565 to 8.2560?

   Yes, this is a typo. We have corrected as 8.2565 to 8.2560. Thank you for your careful notice.

7. Line 384-396, how would the previous studies relate to your results? Some more in-depth discussion is needed here.

   Here we discuss possible regional differences in the pH trends caused by the localized biological factors. We have simplified the descriptions to emphasize this point.

8. Fig. 3. Red and blue colors indicated the annual MAXIMUM and MINIMUM pHinsitu data.

   That is correct. We have modified it.

9. Fig. 9, there is no “black and red shading” as said in captain.

   We have deleted it. Thank you for your indication. We have again carefully checked the all captions to remove such incorrect descriptions.