

Responses to comments from Reviewer 1

Thank you for thorough and constructive comments. Based on the comments from you and another reviewer, we have revised the manuscript as follows:

>P2, L63-65: You are mentioning that “The observed range of pH trend within the Japanese coast corresponds to 85% of that observed in 83 coastal systems in the world (Carstensen and Duarte, 2019), and this result suggests that the Japanese coastal area can function as the “sample shelf” of the coastal environment for the entire world.” I guess you mean that “The observed range of pH trend within the Japanese coast is in harmony with 85% of the ones observed in 83 coastal systems around the world (Carstensen and Duarte, 2019)”. Right? But this does not mean that the Japanese coastal area can represent THE world’s coastal environment. However, it can be considered as one of the coastal areas that can be used as a “sample shelf” or “coastal ocean acidification test area”.

In this sentence we meant to point out that the observed range of pH trend within the Japanese coast is equivalent to 85% of that observed in 83 coastal systems in the world. Also, I agree with your comment that Japanese coasts will be useful as a sample shelf of coastal acidification but not that of coastal environment itself. I modified the words in this paragraph to reflect your comments (lines 62-64).

>In P2, L60-63, you have mentioned that “The Japan Ministry of the Environment (MOE) conducts regular pH monitoring at over 2,000 coastal stations around Japan from the early 1980s until the present, and the obtained data showed significant variability in the multi-decadal pH trend from -0.012 y^{-1} to $+0.009 \text{ y}^{-1}$ among the stations” and in P3, L76-81, you mentioned the 5 stations without justifying the reason you chose them among 2000 stations! It would be very useful to do so before describing each station in details.

Monitoring sites in this study were not selected from the MOE stations but newly launched by two recent pH monitoring programs (Study of Biological Effects of Acidification and Hypoxia (BEACH) and Ocean Acidification Adaptation Project (OAAP)). The reason for determination of monitoring locations differed by program: in BEACH, two stations were set to represent "natural" state of Japanese coastal environment with relatively low anthropogenic nutrient loadings. In OAAP, three stations were selected from major farming areas of Pacific oyster. We add this information in the revised text (lines 79-82).

>Also, the time range of data is different between these 5 stations. Why didn’t you choose the ones that have exactly the same time range of beginning and end of measurements to

>facilitate the inter-comparison?

As these stations were launched in order by two different programs, we couldn't start/end years of these stations. Please extenuate that this is the first synthesis effort of OA monitoring stations operated by different founders / programs.

>P4, L111: You've mentioned that "All sensors were replaced every 2 months". Despite
>calibration? Why?

Several sensors including pH has only three-months lifetime of their batteries, and hence we changed all sensors every two months for safety. In the revised manuscript we add this information (Line 116).

>P4: You didn't mention the precision and accuracy of measurements for any parameter.
>Why? This is crucial to add.

Precision of each parameter is added in the revised text. (Lines 108-110).

>P4: You used CO2sys Excel v2.1 (Lewis and Wallace, 1998) but the correct reference should
>be (Pierrot and Wallace, 2006): Pierrot, D.E. Lewis, and D.W.R. Wallace. 2006. MS Excel
>Program Developed for CO2 System Calculations. ORNL/CDIAC-105a. Carbon Dioxide
>Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy,
>Oak Ridge, Tennessee. doi: 10.3334/CDIAC/otg.CO2SYS_XLS_CDIAC105a ; Index of
>/ftp/co2sys/CO2SYS_calc_XLS_v2.1 (lbl.gov).

Thank you for your comment. I modified the reference.

>P4: Also, you didn't mention the uncertainty of your carbonate system measurements. This
>can be easily calculated through the same CO2sys Excel with the new features added by Orr
>et al. (2018).

Measurement precision of DIC and Talk reported from JAMSTEC was added in Line 129.
Uncertainty of carbon-derived parameters ($p\text{CO}_2$ and Ω_{ara}) are function of T, S, and carbon parameters (pH and Talk in this case), and estimated uncertainty of these parameters calculated by CO2sys were added in Section 3.4 (Lines 391-400).

>You use the term "eutrophicated" in the paper. I would definitely suggest to use the
>conventional terms "eutrophic, oligotrophic, etc."

We changed the term "eutrophicated" to "eutrophic".

>Referring to total alkalinity as Talk in the paper is very confusing. Better to type in its

>conventional abbreviation “TA”.

Talk is a traditional expression of total alkalinity in the field of ocean carbon monitoring, and current major programs such as GO-SHIP still use this expression. However, I agree that TA is more used in the field of ocean acidification study, and I changed abbreviation of this parameter to TA in the revised manuscript.

>Sections 3.3. and 3.4.: You have to mention the r^2 and the significance of each
>equation/relationship in each station. It would be good to add them in a separate table of
>the plots (Figure 9).

The r^2 value and RMSE were added in each equation listed in Figure 9.

>Figure 7: You are showing the years in a decimal way which might be confusing for readers.
>I would suggest to add the months and the year abbreviation, e.g., Jan-20, Jan-21, etc.

>

All x-axis in Figures 7, 8, 10, 11, and 12 were changed to show date/month/year instead of years in a decimal.

>In general, the discussion needs more references, particularly for the statements used to
>explain the results, the methodology of using SDs and plotting those against each other. Also,
>this section still needs more comparison with other studies from the area and beyond.

We added several references to compare our results with former studies; such as

*annual pH ranges (Lines 318-320)

*low buffer effect in low-salinity water (Lines 403-404)

*DIC vs DO relationship (Lines 649-650)

*effect of reduction of nutrient loadings to diminish short-term drawdown of Ω_{ar} (Lines 730-732)

>P1, L20: The forcings mentioned here are already changing due to climate change, and not
> “can change”. Please edit accordingly.

We changed the words to "...forcings are changing..."

>P1, L25,27: What do you mean by “anthropogenic loadings”? Could be replaced by
>“anthropogenic pressures” that encompass all types of atmospheric and terrestrial
>discharges and emissions generated by anthropogenic activities and infrastructures.

Here we meant "anthropogenic nutrient loadings," but "anthropogenic pressures" are better. In the revised text we changed these words accordingly.

>P2, L38: The sentence is not correct, as the ocean is not lowering its pH per say. Please edit
>this sentence “The ocean is lowering its pH because of anthropogenic CO2 input...” to “The
>ocean is witnessing a reduction in its pH due to anthropogenic CO2 sequestration...”
Sentence corrected.

>P2, L40: Please remove “also” after pH and edit as follows: the pH shows...
corrected.

>P2, L42: It’s not only coastal upwelling but also the downwelling that is taking atmospheric
>CO2 from surface layers to the deep ocean. I would suggest to replace it with “coastal
>dynamics”.
Here we keep the word "upwelling," because we are discussing about coastal shallow waters.

>P2, L43: Please replace “input of river water” to “rivers’ inputs”.
corrected.

>P2, L41,43,44: Please add “s” as follows: water mass changes, terrestrial nutrients’ inputs.
added.

>P2, L49-51: Please edit as follows: These short-term pH variations in coastal waters are
>important for local ecosystems as they are mostly caused by natural forcings that have been
>acting before the industrial period, and hence, the local ecosystem is expected to adapt to
>such short-term pH variations as long as they are natural in terms of timing and amplitude.
corrected. Thank you for this nice edition.

>P2, L53: Please edit “changes in land use”.
corrected.

>P2, L560: Please remove “Japan” and add “the country”, to avoid repetitions.
corrected.

>P2, L563: Please replace “among the stations” by “throughout the stations”.
corrected.

>P3, L72: Please replace “in this study” by “Here,”.

corrected.

>P3, L76: Please add “in the following stations”, after “around the coast of Japan”.
added.

>P3, L94: Please add “:” after “three rivers” and remove the word “Rivers” before the
>parentheses.

corrected.

>P3, L95: Please add “s” to nutrient.

corrected.

>P3, L96: Please add a unit after the population numbers here and throughout the manuscript.

>E.g. “residents” could work.

added residents.

>P4, L111: to add always pH units after adding a pH value, e.g., <0.006 pH units. Was this
>difference significant?

The words "pH units" were added to all pH values appeared in the text.

0.006 of pH difference between water intake and settling tank is not significant as this number is smaller than the estimated uncertainty of monitored pH values after drift corrections (± 0.010 for Mitako, Kashiwazaki, and Ohno and ± 0.010 for Shizugawa and Hinase).

>P5, L134: Please remove “was”.

removed.

>P6, L179: Please remove the “s” from “numbers”.

removed.

>P6, L180: Please edit: “..to one third of what they had before..”

corrected.

>P6, L181: Please replace “diminish” by “reduce”.

corrected.

>P6, L186: Please replace “marked” by “remarkable”.

>P7, L219: Please replace the “,” to “;” before the reference here and throughout the text
>(e.g., L221, 222, etc.).
corrected throughout the text.

>P8, L255: To replace “were hung” with “were installed/fixed”.
corrected.

>P9, L274: Do you mean “extremely high”? Please correct.
Sorry for mistyping. It's corrected.

>P9, L274-275: Do you mean that SS variation has a timescale of < 10 days? Please edit.
Here we meant that surface salinity temporally reach to the level of less than 10 salinity unit
in these two stations. We slightly modified sentence in the revised text.

>P11, L288: Please add “values” after “raw pH”.
Here, we changed the words "raw pH" to "pH after drift correction" following to the comment
of another reviewer. So, we don't add "values" in the corresponding point.

>P11, L296: Please add “pH units” after “0.8”.
added.

> P11, L296: Please add the “p” in “pCO₂” in *Italic* throughout the text.
Typeface corrected.

>P11, L299: “Phenomena” is not the correct term here. Maybe better to use “patterns”.
word changed.

>P11, L296-303: Better to use references in this part.
We added one reference that show artificial diurnal variation of ocean parameters caused by
biofouling.

>P11, L318: I suggest to add “concentrations” instead of “values for total alkalinity in the
>entire text (e.g., L329, etc.). And these concentrations “oscillate in a narrow range between
>2222 and 2236...”. Please edit.
In the revised manuscript, we stopped to use "Talk values" and just use TA as "alkalinity" itself
involves the meaning of "concentration" in its definition.

We revised expression of L318 (now L328-329) in the revised text.

>P13, L325: Which phenomenon are you referring to? If you mean the similarity in S, then
>you can write: This similarity in salinity indicates...

Here, we aimed to indicate about similarity of TA range between northwestern Pacific surface waters and the "oceanic" end members in Miyako, Kashiwazaki, and Shizugawa. We revised the sentence to clarify our aim (Line 332).

>P13, L327: What do you mean by "modulated"? maybe use "are not significantly different
>from...".

Sorry for incomprehensible expression here. We changed the words to "different".

>P13, L332-333: Please remove "the fact" and "the" before "the Kuroshio".
corrected.

>P13, L338: Please replace "have already received several modulations by" by "have already
>witnessed several modifications through".
corrected.

>Section 3.3. and Table 1: You are not referring to the methodology used to derive the total
>alkalinity from TA-S relationship. Please note that Jiang et al. (2014) demonstrated that the
>river end-member can only be reliably estimated from this relationship when river input
>dominates as the only significant mechanism controlling the AT-S relationship (e.g., in
>estuaries and river plumes), since physical and biological processes can decouple TA from
>the river water concentration. More information in Hassoun et al. (2019).

>References:

>Jiang, Z.-P., Tyrrell, T., Hydes, D.J., Dai, M., Hartman, S.E., 2014. Variability of alkalinity
>and the alkalinity-salinity relationship in the tropical and subtropical surface ocean. *Glob.*
>*Biogeochem. Cycles* 28, 729–742. <https://doi.org/10.1002/2013GB004678>.

>Hassoun, A.E.R., Fakhri, M., Abboud-Abi Saab, M., Gemayel, E. and De Carlo, E.H., 2019.
>The carbonate system of the Eastern-most Mediterranean Sea, Levantine Sub- basin:
>Variations and drivers. *Deep Sea Research Part II: Topical Studies in Oceanography*, 164,
>pp.54-73. <https://doi.org/10.1016/j.dsr2.2019.03.008> .

We added brief discussion on the validity of simple two end-member mixing model in these study areas, referring Jiang et al. (2014) (Lines 351-353). We concluded that non-SDC changes of TA is small in our study areas, as the calculated freshwater endmember in each

area roughly agreed with the observed river TA that flows into each study area (Table 1).

>Section 3.4: Please use the conventional abbreviations of omega calcite (Ω_{ca}) and aragonite (Ω_{ar}) in the entire paper.
abbreviations corrected.

>P14, L382-383: Please write it as follows: “Since Talk was calculated as a linear equation of >salinity, it was expected to have a resemblance between the Talk and salinity >patterns”.
corrected.

>P14, L387: What do you mean by “were totally oversaturated with CO₂”? Compared to >what?
We meant to say that observed annual averages of $p\text{CO}_2$ were in equilibrium with, or even higher than current atmospheric concentration of CO₂. The sentence was revised (Lines 407-408).

>P16, L400: Please write “Our results indicate...”.
corrected.

>P16, L403: Please start the sentence with “The”.
corrected.

>P16, L412: Please replace one of the “noted” by “highlighted”.
corrected.

>In all the section 4.1, you are referring to your result(s) as phenomenon(a). This is wrong.
>Please edit, and refer to a specific result as a “result”, while the phenomenon is usually a >process that is happening/happened in your study areas, such as OA, eutrophication, etc.
Corrected. Thank you for pointing out this.

>P20, L513-523: Did you check if these annual maximum SDs of the three parameters are >happening significantly at the same time? You can do ANOVA/ANCOVA tests to check this.
We cannot directly check the simultaneity of the occurrence of maximum SDs of these three parameters by ANOVA/ANCOVA, because distribution of SD10 against month was not normal. However, we have checked that not only the month of the occurrence of maximum

SD, seasonal distribution of monthly-averaged SD10 has positive correlation with statistical significance ($r^2 > 0.5$) among these three parameters. In the revised manuscript, we mentioned about this correlation as additional information that support same cause of seasonal variation of SD10 among these parameters (Lines 536-537).

>P24, L606-607: What do you mean by “approximately positive correlation”? It should be >either positive or negative. Is it significant?

Sorry for incomprehensible sentence. The observed positive correlations were statistically significant, only but data of Shizugawa divided into two groups that follows regression line of Hinase and Ohno. In the revised text we changed this sentence to simply indicate significant positive correlation (Line 640).

>P24, L613: This equation is used according to whom? Please mention the reference (the >same for the caption of Fig.13b, the purple dashed line).

Here we used classical Redfield ratio of $O_2 : C = 138 : 106$. I added Redfield et al. [1963] as a reference in this sentence.

>P24, L622: What do you mean by “lateral affection”? I’m sure the word “affection” is irrelevant here. We changed the word to "lateral propagation".

>P26, L626: which analysis are you referring to?

Here we meant to point out our analyses made by Sections 3.2 and 3.4. We changed the sentence in the revised manuscript. (Line 662)

>P26, L629: Please replace “issue” with “aspect”.
corrected.

>P27, L651: Do you mean “we determined the nutrients’ concentrations...”? Please clarify/edit. Your understanding is right. We changed the sentence in the revised manuscript. (Lines 688-689)

>P27, L660-667: References?

This speculation comes from our recent observation, and will be published elsewhere. We modified the sentence in the revised text.

>P28, L680: Please replace “derived” by “controlled”.
corrected.

>P28, L687: Please replace “acidified” with “acidification”.
corrected.

P28, L691-692: How do you know? You didn’t discuss this in the paper! I’d suggest to remove this statement.

This is our observation result and are reported in other paper (Fujii et al., 2023). We added this reference (Line 725) .

>P28, L695-696: Any projections studies confirming this?

This projection is also discussed in Fujii et al. (2023). We add this reference in the text (Line 729).

>P28, L696-698: You also didn’t tackle this in the paper. Such proposed “solution” needs to be
>analyzed taking into consideration various scenarios. Please remove.

We had analyzed in Section 4.3 that the amplitude of the short-term drawdown of pH related to low-salinity events is linear function of liver nutrient concentration, and hence we can say that the short-term drawdown of pH related to low-salinity events will decrease if we can reduce the nutrient concentration of rivers. Similar result has already been reported by Kessouri et al. (2021), so we added this reference in the text (Line 730-732).

On the other hand, I agree that further consideration will be needed to conclude whether such treatment function as the "solution" of future coastal acidification. We hence deleted the latter half of this sentence.

>Table 1: Please make the last column on the right wider, so units can fit next to the concentrations.
Width corrected.

>What is the source of the atmospheric CO₂ number (400 μ atm)? Why are you μ atm using instead of
>ppm?

We corrected this value to 416 ppm, globally averaged atmospheric CO₂ concentration at 2021 (WDCGG, 2023).

>I suggest to change the color of one of the stations in this plot as many readers wouldn’t tell the
>difference.
color changed.

>Please reorder the references from the old to the newest ones (e.g., P2, etc.)

references reordered.

Responses to comments from Reviewer 2

Thank you for thorough and constructive comments. Based on the comments from you and another reviewer, we have revised the manuscript as follows:

>1. In the abstract, the author highlighted that this study also revealed the short-term acidification
>occurs more frequently during low-salinity events yet it is not well explained and emphasized in the
>whole manuscript

>2. Figure 7,8, 10 and etc.. The notations on the x-axis are not immediately recognizable to readers.
>Please change to the Year/Month directly. Also, in Figure 7, the notation of 2021.2 = 36.5 days after
>Jan is incorrect.

We have changed notations of X-axis in Figures 7, 8, 10, 11 and 12 to year/mon/date at each graduation.
Explanation of X-axis in each figure is changed accordingly.

>3. Should report the pH scale of your pH measurement

Although we have already described in Section 2 that we have used total-scale buffers for calibration of pH sensors, we additionally specified in lines 295-296 that all pH values used in this paper is in total scale.

>4. In Figure 7 and line 271, the salinity change of Miyako Site is covered by the other stations and
>cannot be distinguished.

Y-axis and dot size of Figure 7b are adjusted so that two low peaks of salinity in Miyako Site can barely be distinguished.

>5. Line 271, citations are needed when linking the salinity to the precipitation events in different
>seasons.

A reference for coastal salinity variation and seasonal rainfall in Iwate Prefecture is added in Line 275.

>6. Line 278, it is crucial to provide evidence or citations to support the claim that the low dissolved
>oxygen (DO) observed during the summer is attributed to temperature rather than biological
>processes. Because other factors such as the late stage of algal blooms or other events may also lead
>to reduced DO levels in the water

Dominant contribution of water temperature to DO seasonal variation is proved by using DO(eq) in

Section 4.2. In the revised manuscript we add a notation in Line 281 to refer section 4.2 for more detailed discussion about this statement.

>7. Line 279 – 280 The manuscript should consider the potential influence of salinity changes on oxygen solubility, particularly in regions like the Ohno Strait with significant salinity variations.

In these lines we refrain discussion about the causes of short-term DO variation and just described the existence of short-term variation. The origin of short-term DO variation is then discussed in In Section 4.2, where we mentioned that, as you pointed out, shot-term variation of salinity played major contribution to the observed DO variation in Hinase and Ohno.

>8. Line 280- 282, in addition, instead of discussing DO, AOU can serve as an appropriate indicator. In section 4.2 we calculated DO(eq) and DO(diseq), and the later is substantially same variable with AOU. In this paper we use name of DO(diseq) instead of AOU, however, so that we can treat this parameter as a one piece of the biological component package " $C_i(\text{diseq})$," such as $\text{pH}(\text{diseq})$ and $\Omega_{\text{ara}}(\text{diseq})$.

>9. Line 288, why the raw pH data were used not the pH data after drift correction?

Sorry for imprecise expression. All the numbers plotted in Figure 8 were pH values after drift correction. In the revised manuscript, we corrected expression in line 288 (now line 295) and the legend of Figure 8.

>10. Figure 9, all the regressions lack of R squared values, RMSE and other statistical analysis details. RMSE and r^2 value are added in each regression equation in Figure 9.

>11. Table 1, it is unclear how the uncertainty terms of the fresh endmember Talk were calculated.

>Needs descriptions either in the caption or main text.

Uncertainty terms of freshwater endmember was calculated as standard error of $\text{sal} = 0$ intercept in least squares fitting. In the revised manuscript we add this information in the caption of Table1.

>12. Line 380m the pCO_2 , the partial pressure should be italic...

Typeface of partial pressure is corrected to italic.

>13. Major comments in Section 4.1 from line 435

>There are numerous questions concerning the comparison between short-term and long-term variations. Firstly, it is unclear how the standard deviation (SD) was calculated at different time scales.

>For the annual SD, it is not specified whether all data points were used in the calculation. Similarly,
>for the monthly average of 10- days SD, it is unclear whether it was obtained through a moving
>average or only considered the first 10 days, 10-20 days, and 20-30 days of each month. The lack of
>clarity in these definitions and calculations significantly hinders the interpretation of variations, as
>the results heavily depend on how the standard deviations were computed.

Annual average and annual SD were calculated based on the calculated based on the last 1-year data to avoid biases that comes from different time length. We add this information in the caption of Table 2.

\overline{SD}^m_{10} was calculated as the moving average of 10-days SD in each month. We modified the sentence for the explanation of \overline{SD}^m_{10} in lines 455-456.

>14. I do not fully agree that the contribution of the monthly SD to the annual SD can be assessed as
>a ratio or percentage without taking sample size into consideration in the Section 4.1. These need ref
>support.

>Also, if the average monthly SD is smaller than the annual SD, it could be interpreted as the between-
>group variation, implying that monthly mean variations contribute more to the annual variation.
>However, if the average monthly SD is almost the same or even larger than the annual SD, it becomes
>challenging to determine conclusively whether the contribution to the annual variation comes from
>within-group SD (monthly SD) or between-group SD (monthly mean). Again, these discussions
>should have support from statistical literature or equation support.

Following to your comment, we introduced a new variable, annual SD of the monthly averages ($SD_{a<m>}$) in Table 2. contribution of short-term variation with time scale of less than 1 month is then discussed based on the difference between annual SD and $SD_{a<m>}$.

Also, we modified the text of section 4.1 to refrain from discussing about mechanism of temporal variation (e.g., temperature-driven vs biological-driven) at this stage, limiting our discussion to the relative contribution of variations with different time scales.

>15. Line 593. This statement requires further clarification. The thermodynamic process, which
>involves changes in temperature and other constants, also encompasses the air-sea gas exchange
>process. The equilibrium term mentioned in the manuscript is the combined outcome of these two
>processes. It is essential to note that the equilibrium term is often considered negligible because the
>effects of these two processes tend to counterbalance each other.

Please note that we defined $C_i(\text{eq})$ as the estimated concentration of parameter i in equilibrium with the current atmosphere under the observed water temperature and salinity. Variation of $C_i(\text{eq})$ hence only represents purely thermodynamic changes, and non-equilibrium term caused by insufficient gas exchange is incorporated in $C_i(\text{diseq})$ if exist.

In this sentence, we already know that variation $C_i(\text{eq})$ is less than 10 % of that of $C_i(\text{diseq})$. As variation of non-equilibrium term caused by insufficient gas exchange is less than that of $C_i(\text{eq})$ by definition, we can conclude that variation of $C_i(\text{diseq})$ caused by insufficient gas exchange is also negligible without any discussion.

>16. Lines 601-605. The contribution of the SD to the variation of pH should be carefully interpreted.
>Merely relying on the correlation of the SD may not conclusively indicate that pH variations are
>solely contributed by other factors. For example, a situation might arise where high pH variation
>(SDm 10) occurs in the first 5 days of a month, while high salinity variation occurs in the last five
>days of a ten-day period. In such cases, the correlation of the SD might not fully capture the
>relationship between the two variables. To ensure robust and meaningful conclusions, a statistical
>study and appropriate justification should accompany the use of the correlation of the SD.

In lines 601-605 in the original paper, we evaluated relative contribution of non-thermodynamic component to the observed annual variation based not on the correlation between SDs but on the seasonal amplitude of non-thermodynamic component that can be detected both from Table 4 and Figures 11 - 12.