

Referee #1

Thanks for the authors for addressing my previous comments. The ms. has definitely improved a lot. I still have a couple of minor comments and I still think that the discussion could be better enriched with other references and deeper comparisons.

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. 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).

Please replace “this result suggests that the Japanese coastal area can function as the “sample shelf” of the coastal acidification in the world.” to “this result suggests that the Japanese coastal area can be considered as a “sample shelf” for coastal acidification studies.”.

>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 was not selected from the MOE stations but newly launched by two recent pH monitoring programs (Study of Biological Effects of Ocean Acidification and Hypoxia (BEACH) and Ocean Acidification Adaptation Project (OAP)). 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 OAP, three stations were selected from major farming areas of Pacific oyster. We add this information in the revised text (lines 79-82).

>I 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 O₂ monitoring stations operated by different founders / programs.

Please mention that this is “the 1st synthesis effort of OA monitoring stations operated by different founers/programs” clearly in the text.

>P4, L111: You've mentioned that “ O₂ ll 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/CDI O₂ C-105a. Carbon Dioxide

>Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy,

>Oak Ridge, Tennessee. doi: 10.3334/CDI O₂ C/otg.CO2SYS_XLS_CDI O₂ C105a ; Index of >/ftp/co2sys/CO2SYS_calc_XLS_v2.1 (lbl.gov).

Thank you for your comment. I modified the reference.

It is (Pierrot and Wallace, 2006) not (Pierrot et al, 2006).

>P4: O₂ Iso, 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 J O₂ MSTECC was added in Line 129. Uncertainty of carbon-derived parameters (pCO₂ and α_{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 (Lines391-400).

>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 “T_{alk}”.

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 T_{alk} is more used in the field of ocean acidification study, and I changed abbreviation of this parameter to T_{alk} in the revised manuscript.

Please edit in Figure 10 and line 401.

>Sections 3.3. and 3.4.: You have to mention the r² 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 r² 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.

The new format is “year.month”, and is different in Figure 8(b). I still find this confusing.

I would suggest to add the month than the abbreviated year, e.g. Jan-20 or Dec.21, etc.

>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) / Please add “for” before the Baltic Sea.

*low buffer effect in low-salinity water (Lines 403-404) / Please re-arrange the references from old to new.

*DIC vs DO relationship (Lines 649-650) / Please edit: “in the coastal area of East China Sea..”

*effect of reduction of nutrient loadings to diminish short-term drawdown of \square 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.

Then please clarify that you mean anthropogenic nutrients' loadings in L680, 683, 710, 711, 734.

>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.

Please edit: "The country's Ministry of..."

>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 T_{alk} 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 T-S 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 T-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 T-S relationship (e.g., in
>estuaries and river plumes), since physical and biological processes can decouple T-S 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, E.R., Fakhri, M., bboud-bi 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 T-S is small in our study areas, as the calculated freshwater endmember in each

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

Please edit: “In most study areas, calculated freshwater endmember of TA-S regressions agreed with the observed TA of one of freshwater sources (Table 1), indicating that non-dilution/concentration changes of TA (Jiang et al. 2014) was relatively small in these areas, and...”

>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).

Please edit: [WDCGG, 2023](#)

>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 O δ , eutrophication, etc. Corrected. Thank you for pointing out this.

Please edit: “Such a result can be attributed to the perturbation of salinity that...”

>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 δ NOV δ / δ NCOV δ tests to check this. We cannot directly check the simultaneity of the occurrence of maximum SDs of these three parameters by δ NOV δ / δ NCOV δ , 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).

Please edit: "variations with shorter time scale than one month contribute to some parts of the annual variation in the case of salinity."

>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 O₂ : 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.

Please edit: (unpublished data).

>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.

In the caption of Table 2, please edit: "...biases that come from different time length."

P27, L673: Please edit "..., in Hinase and Ohno,"

The discussion is still weak. Results can be better compared with regional and global studies. In addition, many of the discussed statements in the conclusion could be used in the discussion section, while keeping the conclusion section more focused on results and concise.

Referee #2

Overall, this revised version demonstrates significant improvements in clarifying key results and discussions. However, there remain several notable issues that require attention:

The figures and captions appear unchanged despite the author's response indicating revisions were made.

We acknowledge and appreciate the authors' efforts in introducing a new term, which adds clarity. However, it's essential to ensure that this term is adequately described and utilized in the discussion within Section 4.2 for comprehensive understanding.

The use of units, specifically ' $\mu\text{mol kg}^{-1}$ ' or ' $\mu\text{mol/kg}$,' lacks consistency throughout the manuscript.

While there have been improvements, the overall writing quality of the manuscript still requires further refinement. Continued efforts in this regard would contribute to a more polished final product.

Referee #3

This paper presents the findings from continuous measurements conducted at five monitoring sites established along the coast of Japan. The short-period variations in pH are categorized into physical and biochemical factors.

The paper has already undergone initial peer review, and I have read the reports of the two reviewers, the author's responses to them, and the revised manuscript after the first peer review. The data presented are rare in terms of continuous observations conducted annually, and this paper, which provides data from as many as five stations, holds significant value. Therefore, I recommend the publication of this paper in Biogeosciences. While no major revisions are necessary since the paper has undergone peer review once, I suggest some modifications to enhance the comprehensibility of the content for readers.

Firstly, I found the first reviewer's comments regarding the method for calculating standard deviations across the three time scales (10-days, month and year) and their comparative discussion somewhat challenging to grasp at first glance. Although I eventually comprehended the concept after multiple readings, to enhance reader comprehension without requiring additional effort, I suggest including an illustrative example figure. Attached in the PDF is an example displaying the average temperature in Tokyo for the year 2022. The upper panel exhibits the annual variation, while the lower panel displays the January variation. These are supplemented by visual representations of each standard deviation. Providing similar figures using water temperature data from one of the stations as an example would greatly facilitate understanding of the discussion. Please note that the example figure I created is for illustrative purposes and need not match the exact figure used in the paper.

(Line 549) property > parameter?

(Line 556) It is easier to understand if you describe that DO_{diseq} is AOU multiplied by -1.

(Line 559) mol > mole?

(Line 569) The notation "DIC_{min}" is used, but this may be misleading since the DIC calculated from pH_{min} is not smaller, but rather larger. The term "pH_{min}" is introduced near Line 313. If it is explicitly stated after this sentence that "pH_{min}" is to be used for subsequent pH calculations, "pH_{min}" and "DIC" can be referred to as "pH" and "DIC," respectively.

(Line 652) You mention that the respiration quotient in the sediment is possible cause for a larger DIC/O₂ ratio larger the Redfield ratio. However, I believe there is another consideration to be made. The Redfield ratio of 0.77 assumes that both DIC and O₂ are no longer being exchanged with the atmosphere. Given that oxygen is gradually released into the atmosphere over time, it is reasonable to expect that the DIC/O₂ ratio should be significantly higher. The author should know that air-sea difference in pCO₂ can deviate significantly from zero while AOU approaches zero at the surface of the open ocean. If biological activity is occurring at a distance from the station, rather than in its immediate vicinity, it is plausible that the DIC/O₂ ratio could be inflated due to the release or absorption of oxygen into the atmosphere during the advection period.

(Line 675 and Figure 14) I do not agree with the theory that the slope changes with a difference in salinity greater than 1.0. This is because 1.0 salinity has no scientific significance. How is the slope of each area if not divided by salinity 1.0? According to Fig. 14, it may be obvious that the greater the anthropogenic load, the greater the short-term variation of pH in response to the short-term variation of salinity.

(Line 681) the dose > those?

(Figure 7, 8, 10, 11 and 12) The horizontal axis should be corrected to a month-year notation, such as Jan-21, Jan-22. This is an item that was also pointed out by two reviewers in the first peer review.

