Responses to reviewer #2

The authors aim to analyze the influence of riverine input on the CO_2 system in the northwestern Pacific, and its implications for future climate change and coastal acidification. Their analysis focuses on four key parameters of the CO_2 system: total alkalinity (TA), dissolved inorganic carbon (DIC), calcite saturation state (Ωcal), and fugacity of CO_2 (fCO_2).

The manuscript presents an interesting and relevant assessment of the effects of freshwater discharge from the eastern coast of Japan into the northwestern Pacific Ocean. The statistical approach is sound, and the manuscript is well-organized. It should be considered for publication, provided the discussion of acidification processes is further improved.

Thank you for your very helpful comments and references. These were extremely valuable for brushing up our paper. Below are our responses to each comment. We would appreciate it if you could confirm them.

In the Introduction, the authors state that riverine input is the major carbon source for the oceans. However, this claim is not supported by current estimates. Rivers discharge approximately 0.9 to 1.3 Pg of carbon per year into the ocean, in both dissolved and particulate forms. A recent global assessment by Liu et al. (2024) estimates riverine carbon export at 1.02 ± 0.22 (2σ) Pg C yr^{-1}, partitioned into 0.52 ± 0.17 Pg C yr^{-1} of dissolved inorganic carbon, 0.30 ± 0.14 Pg C yr^{-1} of dissolved organic carbon, 0.18 ± 0.04 Pg C yr^{-1} of particulate organic carbon, and 0.03 ± 0.02 Pg C yr^{-1} of particulate inorganic carbon. In contrast, the ocean's uptake of atmospheric CO_2 is estimated to be significantly higher—around 2.9 ± 0.4 Pg C yr^{-1} (Friedlingstein et al., 2025). Therefore, on a global scale, atmospheric carbon uptake is a more substantial source of carbon to the ocean than riverine input. It should be noted, however, that earlier estimates, such as those by Bauer et al. (2013), suggested that in coastal regions specifically, riverine carbon inputs could exceed atmospheric contributions. This distinction between global ocean and coastal zone carbon budgets should be clarified in the manuscript.

We agree with the numerical comparison above. In this introduction, we intended to show that the flux from riverine water is of the same order of magnitude and importance as the flux from the atmosphere. We have rewritten the text to make the quantitative comparison clearer (Line 33-39).

The abstract addresses the issue of coastal acidification; however, this aspect is only indirectly considered in the manuscript, as the selected parameters do not include pH.

Instead, the authors use the calcite saturation state (Ωcal) as a proxy for acidification. While Ω is commonly used in ocean acidification (OA) studies, it is important to note that in the global surface ocean, pH and Ω often exhibit out-of-phase behavior in terms of both spatial distribution and seasonal variability (e.g., Xue et al., 2021; Kwiatkowski & Orr, 2018), which may seem counterintuitive. Although both pH and Ω are widely used as indicators of OA, their asynchronous variability complicates the choice of which parameter better represents the impact of carbonate chemistry on marine organisms (Jokiel, 2013; Waldbusser et al., 2014). This complexity arises from the interplay between long-term trends and short-term natural variability, which can obscure biological responses (Kwiatkowski & Orr, 2018; Landschützer et al., 2018).

Many of the biological effects of ocean acidification depend not only on carbonate ion concentration [CO32−] but also on hydrogen ion concentration [H+], i.e., pH, as well as on bicarbonate [HCO3−] (Cyronak et al., 2015; Jokiel, 2013). If the authors intend to explore the impacts of acidification in depth, it is essential that they include and discuss pH data alongside the other CO_2 system parameters. Without pH, the discussion of acidification remains incomplete and indirect. Alternatively, if the focus is primarily on the carbonate saturation state, the manuscript should clearly state this and align the discussion accordingly.

Furthermore, the authors report Ω (omega) for calcite but do not include Ω for aragonite, which is more commonly used in ocean acidification studies. Aragonite is more soluble than calcite, and organisms that produce aragonitic shells or skeletons are generally more vulnerable to decreasing saturation states under acidified conditions. Therefore, aragonite saturation is typically the preferred indicator when assessing biological sensitivity to ocean acidification. The manuscript would benefit from a clearer justification for the choice of Ω_calcite over Ω_aragonite, or, from the inclusion of both parameters to allow for a more comprehensive assessment.

We agree that omitting a description of pH is very problematic for analyzing the discussion of acidification. The distinct spatiotemporal behavior of pH and Ω, as indicated in the references in this comment, is highly intriguing. Therefore, we have added a description of the analysis of pH together with Ω, which has been recalculated as an aragonite-based one according to the comment. Interestingly, a clear acidification trend was not confirmed by the pH results (see Figure 5). This was due to riverine water causing low SST in Area A, particularly in winter, which counteracted the effects of nDIC inflow. The revised manuscript therefore states that, while riverine water inflow has little effect on hydrogen ion concentration, it does influence biological activities such as calcification (Line 399-410, 434-437). Furthermore, while the effects of hydrogen ion concentration and various carbonate ion concentrations on biological activity are interesting, we found it challenging to provide an individual assessment of these parameters to our manuscript because riverine water inflow affect multiple parameters including SST, SSS, nTA and nDIC, and thus hydrogen ion and carbonate ions simultaneously.

Minor comments

L.21 The sentence should be revised: the affirmation that the riverine wate is not the dominant cause of Dissolved Inorganic Carbon should be better formulated.

We have rewritten as "The analysis showed that riverine water was the main cause of the higher total Alkalinity compared to the surrounding area, whereas its contribution to the increase in Dissolved Inorganic Carbon was relatively minor." (Line 22-24).

L. 23 The supply of by riverine water ": correct.

It was a mistake. We have corrected it.

L371-374 and 380 and 398. The sentences referring to acidification should be revised for clarity and accuracy. As the authors do not include pH data in their analysis, they are not directly addressing the acidification process, which is fundamentally defined by changes in hydrogen ion concentration [H+]. Instead, their focus is on the carbonate saturation state (Ω), which is indeed affected by acidification but also depends on the buffering capacity of the system.

Given this, I suggest that the authors frame their discussion primarily in terms of the saturation state of the studied system, rather than acidification per se, unless they choose to explicitly include calculated or measured pH values. Including pH—either measured or derived from the available CO_2 system parameters—would allow for a more comprehensive and direct discussion of the acidification process.

As responded to above major comment, the descriptions of pH have been added. As a result, differing behavior was observed: while pH showed little change, omega exhibited a clear acidification trend. As noted in the comment, including pH enabled a comprehensive analysis of acidification (Figure 5, Line 399-4104, 34-437, and numerous other lines).

L. 386-387 Regarding the statement that the DIC flow were more complex than the TA flow it is not very clear if the authors refer to the advection to the biologically related process, they should explain better how the flow is affected by a residual term -- which are the processes behind it?

We have rewritten the sentence as “The DIC flows were more complex than the TA flows because they were influenced not only by horizontal advection but also by vertical advection and biological activities such as photosynthesis and remineralization.” (Line 421-423).

L174-176. "Please add units to the colour bars in panels (b) and (c) of Figure 1 to ensure clarity and consistency. Additionally, the significance level (e.g., p-value) for the linear regression should be provided to assess the statistical robustness of the trend. The term 'approximate line' is ambiguous—please clarify its meaning. If it refers to a fitted trend line, specify the method used (e.g., least squares regression).

We have corrected the figure and caption accordingly (Figure 1, Line 185-188). The approximate line is the linear regression line using the least squares method.

L. 237-240 in figure 4 b and e I suppose that the units should be μmol kg-1 month-1 not year-1

The unit have changed to be per month accordingly (Figure 4).

L280-281 However, no regional difference in calcification rate of up to 10 μmol kg-1 year-1 has not been reported… The are two negations which means an affirmation The sentence should be corrected.

It was a mistake. We have corrected it (Line298-299).

L. 331 In figure 6a the units should be μmol kg-1 month-1 not year-1. As the data represented are monthly averages.

We have corrected it like Figure 4 (Figure 6).

L.345-347 Due to the wide offshore oceanic area with respect to the shallow coastal waters of I wonder if the role of submerged aquatic vegetation can be considered relevant. Could you provide more support to this affirmation.

The words “submerged aquatic vegetation” described on the image of coastal observation, but there is no data guaranteeing it has a significant impact on the study area. Therefore, the words have been removed (Line 373).

L.369 Correct “air-sea CO2 flues”.

A mis typo. We have corrected it (Line 396).

Text S1 Vertical profile an advection

“They ranged frm” : correct

A mis typo. We have corrected it.

Text S2 Riverine supply from mainland of Japan

It is not clear to me if the authors assumed the same flow rate for all the rivers or if the compute the total riverine flow considering a proportionality with the flow rate of each river. Could you explain better this part.

We calculated the flow rate considering the proportion. We agree that Text S2 was generally difficult to understand, so we have completely rewritten it.

Could the authors provide a table similar to S1 comparing the characteristic of area A and B in order to show the different oceanographic characteristics

We agree that such table is useful. We added a Table (Table S2, former Table S2 was renamed S3).

"The English throughout the manuscript should be revised by a native speaker or a professional language editor to improve clarity, and overall readability.

Thank you for your comment regarding the English of the manuscript. The manuscript had already been proofread by a professional language editing service. In response to the reviewer’s suggestion, we have carefully re-checked the text and made additional minor revisions to further improve clarity and readability.

References

Bauer, J. E., Cai, W. J., Raymond, P. A., Bianchi, T. S., Hopkinson, C. S., and Regnier, P. A. G.: The changing carbon cycle of the coastal ocean, Nature, 504, 61–70. https://doi.org/10.1038/nature12857, 2013.

Cyronak, T., Schulz,K.G., Jokiel,P.L. 2015.TheOmega myth: what really drives lower calcification rates in an acidifying ocean. ICES Journal of Marine Science, doi: 10.1093/icesjms/fsv075.

Friedlingstein, P. et al. 2025. Global Carbon Budget 2024 Earth Syst. Sci. Data, 17, 965–1039.

Jokiel, P. L. 2013. Coral reef calcification: Carbonate, bicarbonate and proton flux under conditions of increasing ocean acidification. Proceedings of the Royal Society B: Biological Sciences, 280(1764), 529–549. https://doi.org/10.1098/rspb.2013.0031

Kwiatkowski, L., & Orr, J. C. (2018). Diverging seasonal extremes for ocean acidification during the twenty-first century. Nature Climate Change, 8(2), 141–145.

https://doi.org/10.1038/s41558-017-0054-0
Landschützer, P., Gruber, N., Bakker, D. C. E., Stemmler, I., & Six, K. D. (2018). Strengthening seasonal marine CO2 variations due to increasing atmospheric CO2. Nature Climate Change, 8(2), 146–150. https://doi.org/10.1038/s41558-017-0057-x
Liu, M., Raymond, P.A., Lauerwald, R. et al. Global riverine land-to-ocean carbon export constrained by observations and multi-model assessment. Nat. Geosci. ***17****, 896–904 (2024). https://doi.org/10.1038/s41561-024-01524-z)*
Xue, L., Cai, W.-J., Jiang, L.-Q., Wei, Q. 2021. Why are surface ocean pH and CaCO3 saturation state often out of phase in spatial patterns and seasonal cycles? Global BiogeochemicalCycles, 35, e2021GB006949. https://doi.org/10.1029/2021GB00694
Waldbusser, G. G., Hales, B., Langdon, C. J., Haley, B. A., Schrader, P., Brunner, E. L., et al. 2014. Saturation-state sensitivity of marine bivalve larvae to ocean acidification. Nature Climate Change, 5, 273–280. https://doi.org/10.1038/NCLIMATE2479

Thank you very much for adding the reference list. This was very helpful for the revision. In the revised manuscript, we have cited Friedlingstein et al., 2025, Kwiatkowski and Orr, 2018, Lie et al., 2024, and Xue et al, 2021 in response to the major comments above.

Assessing the impact of riverine water on the Northwest Pacific using normalized Total Alkalinity

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[2]Administration Department, Japan Meteorological Agency, Tokyo, Japan.
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Abstract. The impact of riverine water was assessed using salinity-normalized Total Alkalinity observations. The observational data included surface carbonate parameters from decades of surveys conducted by volunteer cargo ships and research vessels in ~~the Northwest Pacific~~. Statistical processes, such as re-gridding and Fourier regression, used in a previous study were also applied in this study to improve the spatiotemporal resolution. First, the seawater area affected by riverine water was identified using an Empirical Orthogonal Function analysis of normalized Total Alkalinity. The differences in normalized Total Alkalinity and Dissolved Inorganic Carbon from the surrounding area were then analysed to evaluate the factors such as riverine water supply, advection effects, and biological activities. In addition, the impact of riverine water on oceanic CO_2 uptake and acidification in the study area was assessed. The analysis showed that riverine water was the main source of total coastal Alkalinity but was not the dominant source of Dissolved Inorganic Carbon. The supply of riverine water had little effect on oceanic CO_2 uptake throughout the year. The supply of ~~by~~ riverine water was a factor in coastal acidification; however, the supplied Total Alkalinity reduced the overall acidification trend by 65%. The results of this study are expected to be further improved by enhancing observations, such as the vertical profiles of carbonate parameters, and are expected to expand to other sea areas and be applied to global budgets.

注釈の一覧 :Responses to Reviewer #2 and Editor.pdf

ページ :7

番号 : 1　作成者 : Tatsuki Tokoro　タイトル : ノート注釈　日付 : 2025/10/10 15:28:43

Thank you for your valuable comments. They have greatly contributed to improving the quality of the revised manuscript. Responses to the comments are provided as replies within each comment box. For simple grammatical comments, the suggested corrections have been reflected accordingly, and no individual replies are provided.

番号 : 2　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:26:17

Please rephrase, the reader should know where the study is conducted in the first sentence.

The northwestern pacific, including the coastal areas of Japan (please specify the coordinates of the geographical extent)

> 作成者 : Tatsuki Tokoro　タイトル : ノート注釈　日付 : 2025/10/10 15:50:02
>
> We have added the information of the study area and the coordinates (Line 14-15).

番号 : 3　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:29:48

obtained

番号 : 4　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:30:00

this area

番号 : 5　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:30:36

Sentence very unclear. What you mean is that you did the same analysis that was done in a previous study but by including more data, and a broader spatiotemporal covergae, right?

> 作成者 : Tatsuki Tokoro　タイトル : ノート注釈　日付 : 2025/10/10 15:50:18
>
> No, whereas the previous study focused on atmospheric-oceanic CO2 flux, this study focuses on nTA diffusion. This sentence was intended to convey that the data used for the analysis itself is largely the same as in the previous study. We have rewritten the sentence to make it easier to understand (Line 16-18).

番号 : 6　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:30:17

番号 : 7　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:31:52

the causes of what?

> 作成者 : Tatsuki Tokoro　タイトル : ノート注釈　日付 : 2025/10/10 15:50:34
>
> We meant "the causes of the difference. We have rewritten as "the potential drivers such as ~" to make it easy to understand (Line 20).

番号 : 8　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:32:19

As noted by R#2, sentence very unclear, rephrase

> 作成者 : Tatsuki Tokoro　タイトル : ノート注釈　日付 : 2025/10/10 15:50:46
>
> We have rewritten to make it easier to understand.
> "The analysis showed that riverine water was the main cause of the higher total Alkalinity compared to the surrounding area, but it did not dominate the increase in Dissolved Inorganic Carbon." (Line 21-23)

番号 : 9　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:33:25

番号 : 10　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:34:00

of

番号 : 11　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:33:42

1. Introduction

The flow of riverine water to the ocean is one of the most important flows in the Earth's system. In the carbon cycle, riverine water is the major carbon source for the oceans (Aufdenkampe et al., 2011; Bauer et al., 2013; Borges et al., 2005; Cai, 2011; Chen and Borges, 2009; Chen et al., 2013), which is responsible for atmospheric CO_2 emissions and acidification in coastal areas (Carstensen and Durate, 2019; Duarte et al., 2013; Tranvik et al., 2009). However, strong carbon flows, such as biological pumps, impact carbonate distribution in coastal areas (Passow and Carlson, 2012; Regnier et al., 2013, 2022). ~~For example, increased biological pump in coastal areas will increase CO_2 absorption from atmosphere, but it will also progress the anoxia on the sediment.~~ In coastal areas, the ~~dec[illegible]sition~~ of organic matter supplied by riverine water is expected to cause more severe acidification, which may [illegible] harmful algal growth and adversely affect marine products such as bivalves (Fitzer et al., 2018; Kessouri et al., 2021; Wallace et al., 2014). Quantifying these complex flows is important for [illegible]~~ing~~ future climate change and ocean acidification as well as for [illegible]~~ing~~ [illegible]~~ical~~ changes in coastal areas.

The Northwest Pacific Ocean, including the coastal areas of Japan, plays a crucial role in global carbon cycle due to [illegible] ~~strong sinks~~ of atmospheric CO_2 (Takahashi et al., 2002, 2009). While several studies on carbonate systems have been conducted in the Northwest Pacific Ocean (e.g., Ishii et al., 2001; Murata et al., 1998; Takamura et al., 2010; Tokoro et al., 2023; Yoshikawa-Inoue et al., 1995, 2014), spatiotemporal variations in riverine water contribution in this region have not yet been quantified. Riverine water tracers (e.g., salinity, stable isotope, geogenic solutes like silica) are one of the most effective methods for evaluating the influence of riverine water. Although salinity is the most frequently assessed factor in riverine water transportation, other factors such as precipitation and evaporation also impact salinity. This can be a source of major error in the assessment of water transportation in the northwestern Pacific region, where high precipitation and evaporation by low-pressure systems and heat waves, respectively, have been observed (Kitamura et al., 2016; Miyama et al., 2021; Sugimoto et al., 2013).

The total Alkalinity (TA) normalized by salinity is a potential indicator ~~to~~ assess the influence of riverine water on ocean. TA [illegible] ~~a carbonate parameter in seawater and~~ is defined broadly [illegible] he charge difference between ~~the~~ proton [illegible] and a[illegible]or (see Zeebe and Wolf-Gladrow, 2001 f[illegible]re detailed definition). TA [illegible]~~s~~ by several factors, such as advection from different water masses and biological [illegible]~~ism~~, including ~~the~~ calcification and dissolution of calcium carbonate. Because TA is also highly correlated with salinity, TA normalized to [illegible]eference salinity (nTA) has been used to quantify the abov[illegible]tors (e.g., Broecker and Peng, 1982; Lee et al., 2006; Millero et al., 1998). nTA is calculated as follows:

$$\mathrm{nTA} = \mathrm{TA} \cdot \frac{S_{ref}}{S} \quad (1)$$

where, S and S_{ref} are the measured and reference salinities (traditionally 35), respectively. Similarly, Dissolved Inorganic Carbon concentration (DI[illegible]s also normalized (nDIC). Equation (1) is formulated based on the assumption that a water mass with zero salinity has zero TA, otherwise the right-hand side would go to infinity. However, this assumption is not true for riverine water because its TA is greater than zero, even when salinity is zero, owing to the weathering of carbonate and silicate rocks (e.g., Friis et al., 2003; Lehmann et al., 2023; Rassmann et al., 2016; Taguchi et al., 2009). Therefore, when

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番号 : 1　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:14:56

番号 : 2　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:14:25
For example, an increased biological pump in coastal areas will enhance CO_2 absorption from the atmosphere, but it will also promote anoxia in the sediments.

番号 : 3　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:15:35
mineralization

番号 : 4　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:15:21

番号 : 5　作成者 : Frédéric Gazeau　日付 : 2025/09/30 14:25:06
stimulate

番号 : 6　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:16:28

番号 : 7　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:19:06
biogeochemical

番号 : 8　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:18:26
projecting

番号 : 9　作成者 : Frédéric Gazeau　日付 : 2025/09/30 14:24:37
anticipating

番号 : 10　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:17:12

番号 : 11　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:18:41

番号 : 12　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:18:50

番号 : 13　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:19:44
its strong sink

番号 : 14　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:19:33

番号 : 15　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:21:28
as

番号 : 16　作成者 : Frédéric Gazeau　日付 : 2025/10/08 14:38:53
donors

番号 : 17　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:21:17

番号 : 18　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:21:22

番号 : 19　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:21:41

番号 : 20　作成者 : Frédéric Gazeau　日付 : 2025/10/08 14:40:08
a

番号 : 21　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:22:03
acceptors

番号 : 22　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:22:41
is influenced

番号 : 23　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:22:32

番号 : 24　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:23:07
activity

番号 : 25　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:23:00

8 ページの注釈は次へ続きます

1. Introduction

The flow of riverine water to the ocean is one of the most important flows in the Earth's system. In the carbon cycle, riverine water is the major carbon source for the oceans (Aufdenkampe et al., 2011; Bauer et al., 2013; Borges et al., 2005; Cai, 2011; Chen and Borges, 2009; Chen et al., 2013), which is responsible for atmospheric CO_2 emissions and acidification in coastal areas (Carstensen and Durate, 2019; Duarte et al., 2013; Tranvik et al., 2009). However, strong carbon flows, such as biological pumps, impact carbonate distribution in coastal areas (Passow and Carlson, 2012; Regnier et al., 2013, 2022). ~~For example, increased biological pump in coastal areas will increase CO_2 absorption from atmosphere, but it will also progress the anoxia on the sediment.~~ In coastal areas, the ~~dec sition~~ of organic matter supplied by riverine water is expected to cause more severe acidification, which may harmful algal growth and adversely affect marine products such as bivalves (Fitzer et al., 2018; Kessouri et al., 2021; Wallace et al., 2014). Quantifying these complex flows is important for ~~pr ing~~ future climate change and ocean acidification as well as for ~~p ing bio ical~~ changes in coastal areas.

The Northwest Pacific Ocean, including the coastal areas of Japan, plays a crucial role in global carbon cycle due to ~~strong sinks~~ of atmospheric CO_2 (Takahashi et al., 2002, 2009). While several studies on carbonate systems have been conducted in the Northwest Pacific Ocean (e.g., Ishii et al., 2001; Murata et al., 1998; Takamura et al., 2010; Tokoro et al., 2023; Yoshikawa-Inoue et al., 1995, 2014), spatiotemporal variations in riverine water contribution in this region have not yet been quantified. Riverine water tracers (e.g., salinity, stable isotope, geogenic solutes like silica) are one of the most effective methods for evaluating the influence of riverine water. Although salinity is the most frequently assessed factor in riverine water transportation, other factors such as precipitation and evaporation also impact salinity. This can be a source of major error in the assessment of water transportation in the northwestern Pacific region, where high precipitation and evaporation by low-pressure systems and heat waves, respectively, have been observed (Kitamura et al., 2016; Miyama et al., 2021; Sugimoto et al., 2013).

The total Alkalinity (TA) normalized by salinity is a potential indicator to assess the influence of riverine water on ocean. TA ~~is a carbonate parameter in seawater and~~ is defined broadly he charge difference between ~~the~~ proton and a or (see Zeebe and Wolf-Gladrow, 2001 f re detailed definition). TA ~~c s~~ by several factors, such as advection from different water masses and biological ~~me ism~~, including calcification and dissolution of calcium carbonate. Because TA is also highly correlated with salinity, TA normalized to ference salinity (nTA) has been used to quantify the abov tors (e.g., Broecker and Peng, 1982; Lee et al., 2006; Millero et al., 1998). nTA is calculated as follows:

$$\mathrm{nTA} = \mathrm{TA} \cdot \frac{S_{ref}}{S} \quad (1)$$

where, S and S_{ref} are the measured and reference salinities (traditionally 35), respectively. Similarly, Dissolved Inorganic Carbon concentration (DIC s also normalized (nDIC). Equation (1) is formulated based on the assumption that a water mass with zero salinity has zero TA, otherwise the right-hand side would go to infinity. However, this assumption is not true for riverine water because its TA is greater than zero, even when salinity is zero, owing to the weathering of carbonate and silicate rocks (e.g., Friis et al., 2003; Lehmann et al., 2023; Rassmann et al., 2016; Taguchi et al., 2009). Therefore, when

番号 : 26 作成者 : Frédéric Gazeau 日付 : 2025/09/22 17:23:18

番号 : 27 作成者 : Frédéric Gazeau 日付 : 2025/09/22 17:23:42

a

番号 : 28 作成者 : Frédéric Gazeau 日付 : 2025/09/22 17:23:27

番号 : 29 作成者 : Frédéric Gazeau 日付 : 2025/09/22 17:23:53

mentionned

番号 : 30 作成者 : Frédéric Gazeau 日付 : 2025/09/30 14:41:12

, which is the sum of carbon species,

Equation (1) is applied to areas affected by riverine water, the nTA value is higher than that of the surrounding seawater. Conversely, the influence of riverine water, along with factors such as water mass advection and biological ~~[illegible]sm~~, can be quantified by assessing the distribution of nTA. Unlike other methods that use salinity as a tracer for riverine water, nTA defined in Equation (1) excludes the effect of precipitation and evaporation, which is advantageous because contamination by water masses with almost zero salinity and TA can be excluded. [illegible] also easier to quantify the influence of seawater from different local areas because the TA has different values in different local areas, even if the salinity is the same (Lee et al., 2006; Takatani et al., 2014).

In this study, we aimed to analyse the influence of riverine water on [illegible]~~nate systems in the ocean~~ using nTA and other carbonate parameters measured by voluntary cargo ships and research vessels ~~in the Northwest Pacific~~. First, spatiotemporal variations in the area in which riverine water significantly affected surface seawate[illegible]re identified using [illegible]pirical Orthogonal Function (EOF) analysis of the nTA distribution. Second, we focused on the differences in TA and DIC between the riverine water-affected area and surroundin[illegible]as. We quantified the riverine water supply and other contributing factors that affect TA and DIC. The final step involved the evaluation of the effects of riverine water on the environment in riverine water-affected area. Seawater CO_2 fugacity (fCO_2) and the calcite saturation state of seawater (Ω_{cal}) were the two carbonate parameters that were used as the index of environmental changes caused by riverine water input. ~~The former parameter affects the air-sea CO_2 flux.~~ The TA and DIC supplied by riverine water ~~[illegible]hange~~ seawater fCO_2 and oceanic CO_2 uptake. Meanwhile, [illegible]s the ratio of the concentration product of [Ca^{2+}] and [CO_3^{2-}] to the solubility product of calcite and is an index of ocean acidification. Calcite is a mineral composed of $CaCO_3$ and constitutes foraminifera, coccolithophorids, and ~~the ou[illegible]yer~~ of bivalves. Therefore, the analysis of changing Ω_{cal} is expected to lead to a more detailed assessment of coastal acidification in the study area. This study also aimed to evaluate the effects of riverine water on future climate change and coastal acidification and to predict the effects of future environmental changes.

2. Methods

2.1 Data for analysis

The observational data in this study were collected by the National Institute for Environmental Studies (NIES), Meteorological Research Institute (MRI) of the Japan Meteorological Agency (JMA), and Japan Fisheries Research and Education Agency (FREA). The NIES data were produced as part of the Voluntary Observing Ship (VOS) programs for cargo ships (namely, M/S Alligator Hope, M/S Pyxis, M/S New Century 2, and M/S Trans Future 5). MRI, JMA, and FREA collected data from research vessels (R/V Mirai and R/V Hakuho-maru for MRI; R/V Keifu-maru and R/V Ryofu-maru for JMA; and R/V Wakataka-maru and R/V Soyo-maru for FREA). These data were uploaded to the Surface Ocean CO_2 Atlas (SOCAT; Pfeil et al., 2013; Bakker et al., 2016, https://socat.info/index.php/data-access/) and Global Ocean Data Analysis Project (GLODAP, Key et al., 2015; Olsen et al., 2016; Olsen et al., 2020, https://glodap.info/). These observations were

番号 : 1　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:57:11
activity

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番号 : 3　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:57:45
I do not understand this sentence, please rephrase

作成者 : Tatsuki Tokoro　タイトル : ノート注釈　日付 : 2025/10/10 15:51:17
We have rewritten as "Furthermore, nTA can be used to quantify the influence of seawater inflow from different local areas because distinct regional differences in nTA have been reported (Kakehi et al., 2017; Lee et al., 2006; Takatani et al., 2014).".(Line 72-74)

番号 : 4　作成者 : Frédéric Gazeau　日付 : 2025/09/22 17:58:36
the crbonate chemistry of the Northwest Pacific

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an

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carbonate chemistry

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unaffected

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番号 : 11　作成者 : Frédéric Gazeau　日付 : 2025/10/08 14:55:18
influence

番号 : 12　作成者 : Frédéric Gazeau　日付 : 2025/09/22 18:01:08

番号 : 13　作成者 : Frédéric Gazeau　日付 : 2025/09/22 18:01:52

番号 : 14　作成者 : Frédéric Gazeau　日付 : 2025/09/22 18:03:00
Please check comment from R#2, I concur, you should restrict here the use of omega not from an ocean acidification perspective but changing saturation conditions impacting calcifiers.

作成者 : Tatsuki Tokoro　タイトル : ノート注釈　日付 : 2025/10/10 15:51:34
We agree that omitting pH in the acidification discussion is problematic. However, unlike omega, the calculated pH did not confirm a trend of acidification. This is considered due to the SST dependency of pH. Because the difference between the two acidification index is interesting, we have decided to describe both pH and omega in the revised manuscript. Furthermore, according to the comment, omega has been changed to the aragonite based one.(Figure 5, Line 399-4104, 34-437, and numerous other lines).

番号 : 15　作成者 : Frédéric Gazeau　日付 : 2025/10/08 14:58:38
shells

番号 : 16　作成者 : Frédéric Gazeau　日付 : 2025/09/22 18:04:56

statistically processed by re-gridding, a second-order approximation of TA, carbonate equilibrium calculations, and Fourier regression into datasets with a spatial resolution of 1° × 1° and a temporal resolution of 0.1 year, as suggested in a previous study (Tokoro et al., 2023). Similar to the previous study, the data was excluded from the analysis if there was insufficient temporal data (n > = 60, [illegible]spond to 6 years data) in the 1°×1° spatial grid. Although the time interval (from January 1, 2000, to December 31, 2019), covered area (latitudes of 20–50°N and longitudes of 120–160°E), and original observations in the SOCAT by NIES were the same as those in the previous study, the original observations by MRI and JMA increased slightly (287750 to 291138) with the upgrade of the product (SOCAT ver. 2019 to 2023). The FREA dataset is a new addition to those used in previous studies. As the FREA data (n = 566175) were collected every 1 minute while the other data were collected every 10 min, the FREA data were weighted 1/10 with respect to the other data and weighted averaged as the gridded data. The GLODAP data for surface (<10 m depth) TA and DIC from the MRI and JMA were updated using ver. 2.2020 to 2.2023. The number of data points for JMA data increased from 2080 to 2163. However, the effect of increase in the data was small, which did not statistically affect the result (Table S1). Additionally, observations up to a depth of 150 m within the GLODAP were used to evaluate the effects of vertical advection (Text S1).

2.2 EOF analysis, Cause Analysis, and Environmental Impact Assessment

In this study, areas significantly affected by riverine water were identified by applying EOF analysis to the spatiotemporal variability of nTA. The EOF analysis breaks down spatiotemporal variations into multiple orthogonal modes with multiple spatial patterns (principal EOF patterns) and time series (principal component time series) (e.g., Denbo and Allen, 1984). In addition to the effect of riverine water, the target area in this study is assumed to be subject to a mixture of multiple fluctuations, such as seasonal fluctuations of the Kuroshio and Oyashio flows. Compared to the direct use of carbonate parameters, the EOF analysis is expected to identify the influence of riverine water because the analysis is appropriate to quantify and separate multiple fluctuations. Using the principal EOF pattern and principal component time series with respect to the spatiotemporal variation of nTA, we identified the area significantly influenced by riverine water and labelled [illegible]s “Area A”. We then labelled the surrounding [illegible]vith almost the same area as “Area B.” The influence of riverine water was quantified as the value of Area A minus that of Area B (*dAB*) for all related parameters such as SST (Sea Surface Temperature), SSS (Sea Surface Salinity), nTA, and nDIC. Causal analysis of *dAB* of TA and DIC was performed using the following equation:

$$\frac{\partial dAB}{\partial t} = Sup_{rw} + C_{flux} + C_{res} \qquad (2)$$

The left-hand side of the equation represents the time derivative of *dAB* of nTA and nDIC. Sup_{rw} is the TA or DIC supply by riverine water; C_{flux} is the term of difference in air-sea CO_2 flux between Areas A and B divided b[illegible]ed layer depth (*MLD*). The MLD with the same spatiotemporal resolutions as the processed data (1° × 1° and 0.1 year) was calculated from the reanalysis ~~data~~ of seawater temperature profiles by Japan Agency for Marine-Earth Science and Technology (JCOPE2M;

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corresponding to 6 years of data

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region, with a rather similar surface area, as...

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it

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番号 : 5　作成者 : Frédéric Gazeau　　日付 : 2025/09/22 18:41:05

Miyazawa et al., 2017, 2019, https://www.jamstec.go.jp/jcope/htdocs/distribution/). We determined an isothermal depth at $\Delta T = 0.2$ °C with linear interpolation (de Boyer Montégut et al., 2004; Holte and Talley, 2009). C_{flux} was applied only to the DIC input. C_{res} is the TA or DIC input due to other residual factors related to *dAB* (e.g., horizontal and vertical advection and biological metabolism).

Sup_{rw} was estimated from river discharge and TA or DIC of river water as follows:

$$Sup_{rw} = \frac{Flow_r \cdot C_r}{MLD \cdot A} \tag{3}$$

where, $Flow_r$ is the monthly total flow rate of the river, Cr represents the TA or DIC of the associated rivers, A is the area of Area A. The flow rate was estimated using the Water Information System database of the Ministry of Land, Infrastructure, Transport, and Tourism of Japan (http://www1.river.go.jp/). As TA and DIC data were not available for all rivers, the referential values for zero-salinity endmembers in the three most river-influenced inner bays (Tokyo Bay, Ise Bay, and Osaka Bay) in the relevant coastal areas were used (518–1006 and 475–1371 μmol kg^{-1} for TA and DIC, respectively; Taguchi et al., 2009; Tokoro et al., 2021). The DIC range includes the value predicted as the range of seasonal variation (± 200 μmol kg^{-1}, Tokoro et al., 2021). Although TA range did not consider seasonal variations, the range is estimated to be an order of magnitude smaller than the range among the three bays [illegible] the literature value (Romero-Mujalli et al., 2018). All data were regridded to the resolution of the processed SOCAT and GLODAP data (spatial and temporal resolution of 1° × 1° and 0.1 year, respectively).

Air-sea CO_2 flux (F) was determined as follows:

$$F = k \cdot K_0 \cdot (fCO_{2water} - fCO_{2air}) \tag{4}$$

where, k is transfer velocity defined by the wind speed above the sea surface (Wanninkhof, 2014). K_0 is the solubility of CO_2 in seawater and was calculated using an empirical equation based on SST and SSS (Weiss, 1974). fCO_{2water} and fCO_{2air} are the fCO_2 in surface seawater and air, respectively. C_{flux} was determined by the difference in air-sea CO_2 flux between Areas A and B, which was calculated from the processed fCO_2 data and wind velocity from the database of the Cross-Calibrated Multi-Platform (CCMP, Atlas et al., 2011; Mears et al., 2019, https://data.remss.com/ccmp/v02.0/, version 2.0). The details of the calculations are the same as those used in a previous study (Tokoro et al., 2023).

The effect of riverine water on air-sea CO_2 flux and acidification were quantified by multivariate analysis using *dAB* of seawater fCO_2 and Ω_{cal} as the objective variables and *dAB* of SST, SSS, nTA, and nDIC as the explanatory variables. Ω_{cal} was determined using the equation of a calcite solubility product (Mucci, 1983) and the concentration of Ca^{2+} and CO_3^{2-} estimated using the CO2SYS program (Lewis and Wallace, 1998). Both seawater fCO_2 and Ω_{cal} can be unambiguously determined from equilibrium calculation using SST, SSS, nTA, and nDIC (Zeebe and Wolf-Gladrow, 2001). However, it is difficult to intuitively understand the contribution of the explanatory variables because these have a non-linear relationship. Therefore, we considered that a multivariate linear model should be useful in this study. Partial least squares regression (PLS regression; Wold et al., 2001) was used in the multivariate analysis to prevent multicollinearity, especially considering the strong correlation between SSS and nTA and nDIC.

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concentrations

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concentration in

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Not clear, could you rephrase?

作成者 : Tatsuki Tokoro　タイトル : ノート注釈　日付 : 2025/10/10 15:51:47
We have rewritten as "The DIC range was determined based on the maximum (1171 μmol kg_{-1} in Tokyo Bay) and minimum (675 μmol kg_{-1} in Ise Bay) freshwater endmember values among the three bays, with an additional ±200 μmol kg_{-1} to account for seasonal variation estimated in the previous study (Tokoro et al., 2021)." for clarification. (Line 143-145)

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it

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番号 : 7　作成者 : Frédéric Gazeau　日付 : 2025/09/22 18:43:56
what does that mean?

do you mean “based on values from the literature”?

作成者 : Tatsuki Tokoro　タイトル : ノート注釈　日付 : 2025/10/10 15:52:16
The value from a literature means the global range of riverine TA variation due to water temperature change. We have rewritten the sentences for clarification. (Line 145-148)

番号 : 8　作成者 : Frédéric Gazeau　日付 : 2025/09/30 17:04:04
at 10 m

番号 : 9　作成者 : Frédéric Gazeau　日付 : 2025/09/22 18:48:29
Comment from R#2

作成者 : Tatsuki Tokoro　タイトル : ノート注釈　日付 : 2025/10/10 14:22:37
As mentioned above, we have decided to use both pH and omega for the index of acidification.

3. Results

Figure 1 represents the processed spatial distributions of SSS, TA, and nTA. The SSS showed a north-south gradient, which was attributed to the high-salinity Kuroshio current and the relatively low salinity Oyashio current. There was also an area of reduced SSS along the Pacific coast of mainland Japan (32–34°N, 132–140°E; Figure 1a). These trends were similar for TA, which exhibited high correlation with SSS (R^2 = 0.94) (Figure 1b, d). The nTA was high in the northern part of the study area, and slightly high values were also observed along the Pacific coast of Japan (Figure 1c). The intercept of the regression line between SSS and TA was 528.04 ± 2.00 μmol kg^{-1} (Ave ± SE), which was consistent with the TA value of large river on the continental side like the Amur River (589 μmol kg^{-1}; Andreev and Pavlova, 2009) and Japanese river water (518–1006 μmol kg^{-1}) (Figure 1d). Because the intercept value was above zero, nTA seemed to be inversely proportional to SSS (R^2 =0.57), with a lower SSS tending to have a higher nTA (Figure 1e). These results indicate that the study area was affected by freshwater with TA above zero, especially in the northern area and on the Pacific coast of Japan, and that nTA could be used as a tracer for ~~the~~ freshwater [illegible].

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inputs

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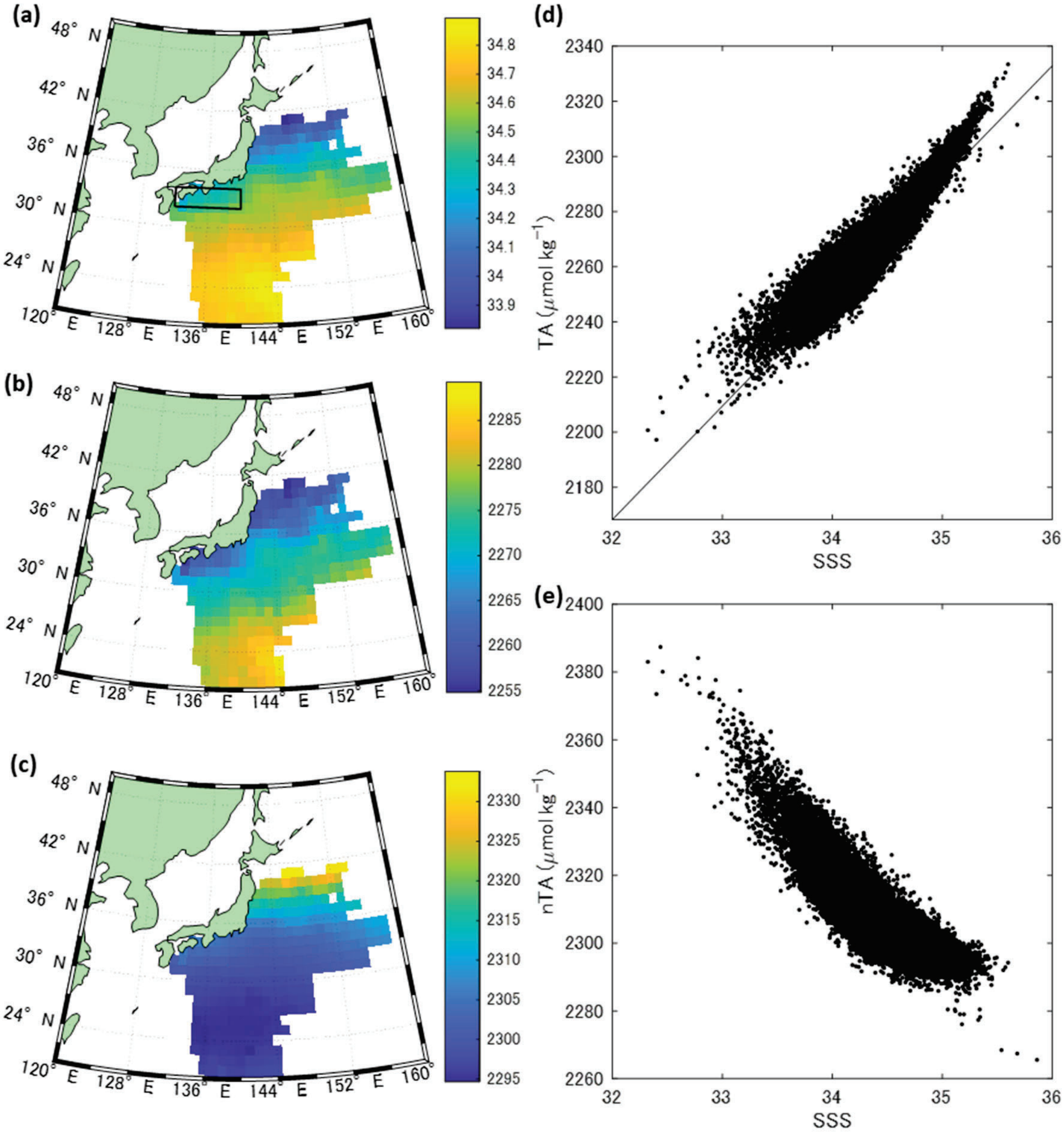

re 1 (a–c): Spatial distribution of (a) mean SSS (b) TA and (c) n he rectangular in (a) show the reduced SSS area along the Pacific coast of mainland Japan (32–34°N, 132–140°E) (d and e): Scatterplot of (d) SSS-TA and (e) SSS-nTA. The black line is the approximate line (R^2 = 0.94, TA = (50.46 ± 0.06) × SSS + (528.04 ± 2.00) (Ave. ± SE)).

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Figure captions should not include undefined acronyms, please detail all (e.g. Sea Surface Salinity (SSS), Total alkalinity (TA) etc..

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Missing units on the left plots (or add them to the caption)

The spatial distribution of nTA in the three most dominant modes of the EOF analysis is shown in Figure 2(a–c). The principal EOF patterns were indicated by a parameter defined as "EOF anomaly," denoted by red or blue in Figure 2. The spatiotemporal variation of the nTA anomaly in each mode was expressed as the product of the EOF anomaly and the principal component time series. In the most dominant mode (Mode 1), the principal patterns exhibited a clear north-south difference at approximately 37°N (Figure 2a). The principal component of the time series in Mode 1 (Figure 2d) indicated that the annual cycle was predominant. Annual maximum of nTA was observed in summer in the area south of 37°N, while it was in winter in the area north of 37°N (data not shown). The seasonal variation south of 37°N can be explained by the fact that riverine water supply is proportional to the precipitation over land, thereby reaching its peak in summer on the Pacific side of Japan (Database of Japan Meteorological Agency, https://www.data.jma.go.jp/stats/etrn/index.php). For the area north of 37°N, this may be due to the southward transportation of high-nTA surface seawater in the Pacific subarctic region by north winds in winter or winter vertical mixing that supplied subsurface high-nTA seawater to the surface. In addition, the EOF anomaly south of 37°N was significantly correlated with distance from the Japanese mainland (Figure 2e). Because EOF anomaly is a parameter indicating the strength of annual cycle fluctuation in nTA, as shown in Figure 2d, Figure 2e indicates that the degree of temporal variation in nTA increased significantly closer to mainland Japan in this mode. The other major modes (Modes 2 and 3; Figure 2b, c) had an east-west distribution north of 37°N. A distribution such as this could represent seasonal or multi-year variations in the flow path of the Kuroshio Extension. After mode 4, the variation north of 37°N was also explained, and thus mode 1 was dominant for the variation south of 37°N.

Based on the above considerations, we determined that it was appropriate to use the Mode 1 EOF anomaly south of 37°N as an indicator of the influence of riverine water from mainland Japan. Although the area north of 37°N contains Japanese rivers with large flow rates (e.g., the Kitakami River), the effect of riverine water was excluded from the analysis in this study because the influence of high nTA in the subarctic gyre was too dominant to extract the influence of riverine water from Japan. The influence of subarctic Pacific seawater can be expected even around 37°N; however, this is not expected to have a significant effect on the statistics in Area A, such as the spatial mean values. Area A is located south of 37°N and within 250 km of land (Figure 2f). This distance is defined using change-point detection (Killick et al., 2012), meaning that the mean EOF anomaly south of 37°N changes abruptly before and after this distance. The outer edge of Area A roughly aligned with the Kuroshio axis, as indicated by the JCOPE2 SSH data (SSH = 0.2 m). Although the EOF anomaly outside Area A also shows a relatively weak correlation with distance, this correlation is thought to be due to the influence of riverine water from areas other than Japan, such as East Asia, which was difficult to quantify using the measurement data in this study. To minimize the influence of riverine water outside Japan and differences in latitude and longitude, Area B was determined as adjacent to Area A, similar in size to the area for comparison (Figure 2f).

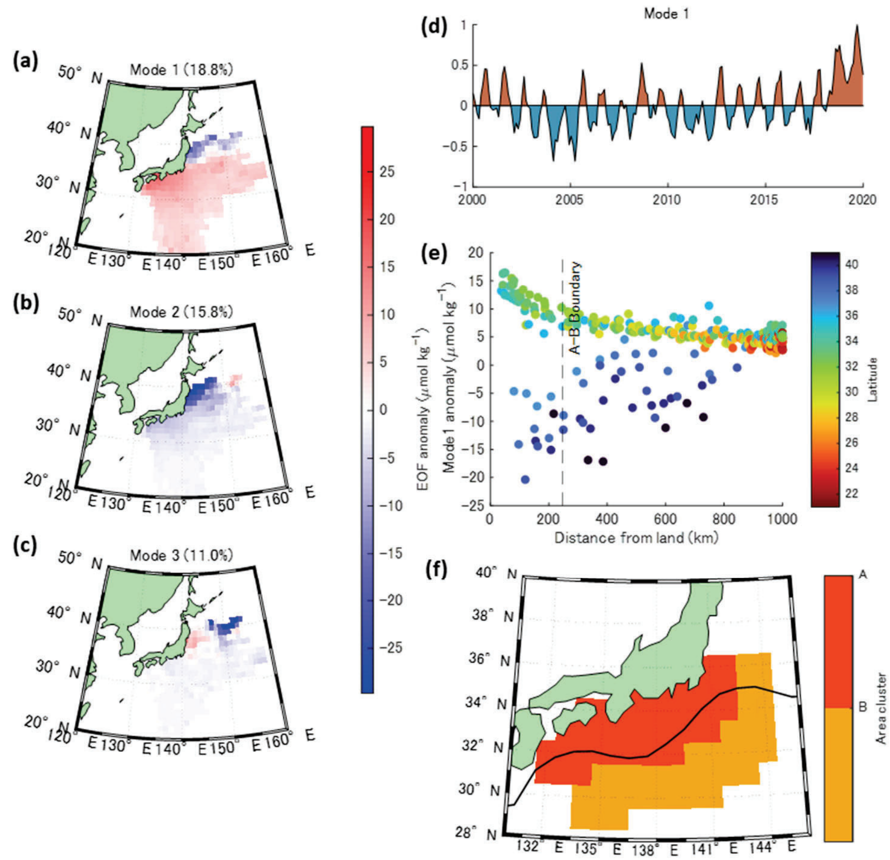

Figure 2. (a–c): EOF anomaly of (a) Mode 1, (b) Mode 2, and (c) Mode 3. The percentage in the bracket (18.8%) indicates the contribution of this mode to the original variation. (d): Principal component of time series of Mode 1. (e): Scatterplot of EOF anomaly of Mode 1 versus distance from Japan mainland. The plot colours indicate the latitude. The dotted line represents the boundary of Areas A and B estimated using the changepoint analysis. (f): Distribution of Area A and B. The black curve indicates mean Kuroshio axis during the whole period in this study (2000-2019).

The monthly and annual average values of the differences in the relevant parameters (SST, SSS, nTA, and nDIC) between Areas A and B (*dAB*) are shown in Figure 3. All *dAB* parameters showed seasonal variation, and the absolute values of *dAB* were largest in winter for SST and nDIC and in summer for SSS and nTA. On a decadal scale, the absolute values of all *dAB* tended to increase significantly. This indicates an increase in the supply of riverine water, which is consistent with the Japan Meteorological Agency's report (https://www.data.jma.go.jp/cpd/cgi-bin/view/index.php) that precipitation in relevant areas of Japan increased during the period analysed in this study.

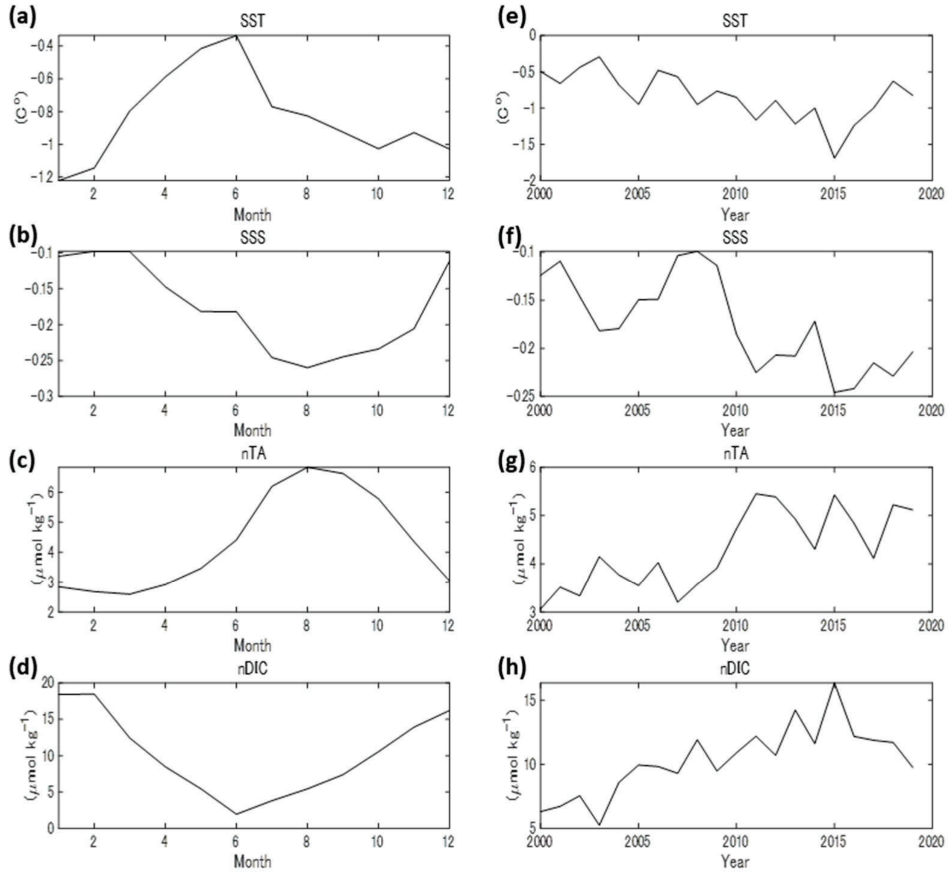

Figure 3. The differences in SST, SSS, nTA, and nDIC between Areas A and B (*dAB*). (a)~(d): Seasonal variations. (e)~(h): Annual variations. Although the time step in this study was 0.1 year, the monthly values were calculated by spline interpolation of values at 1/12-year intervals. The same applies to the other figures.

Figure 4 represents the time-series variations of each term in Equation (2) used for the causal analysis of the *dAB* of nTA and nDIC. The river discharge for Sup_{rw} was calculated as the sum of the 37 rivers bordering Area A (see Text S2). The maximum nTA and nDIC Sup_{rw} were observed during the summer months, when precipitation on the Pacific coast of Japan was the highest. Though the air-sea CO_2 flux term (C_{flux}) also showed a clear seasonal variation, the annual average did not. The residual term (C_{res}) tended to be negative for both TA and DIC; however, the seasonal patterns and ranges of variation differed considerably.

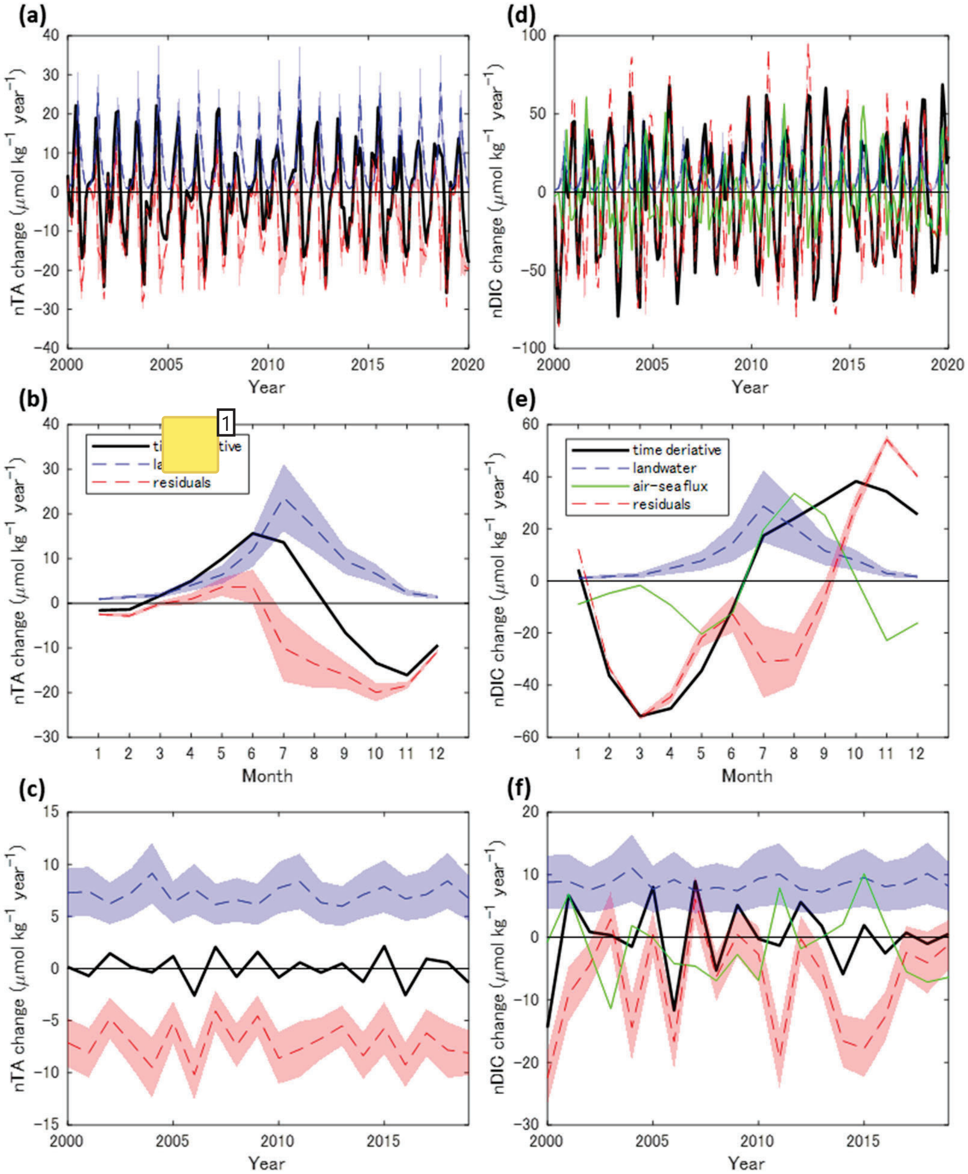

Figure 4. (a): Temporal variations of nTA time derivative and related terms in Equation (2). (b): The monthly averages. (c): The yearly averages. (d)~(f): Similarly, but for nDIC. The shaded areas are error ranges calculated from the upper and lower limits of the TA (518-1006 µmol kg^{-1}) for and DIC (475-1371 µmol kg^{-1}) in riverine water. The ranges are not random errors thus have remained the same error range for monthly or yearly averaging. The dotted lines in shaded areas represent the average values.

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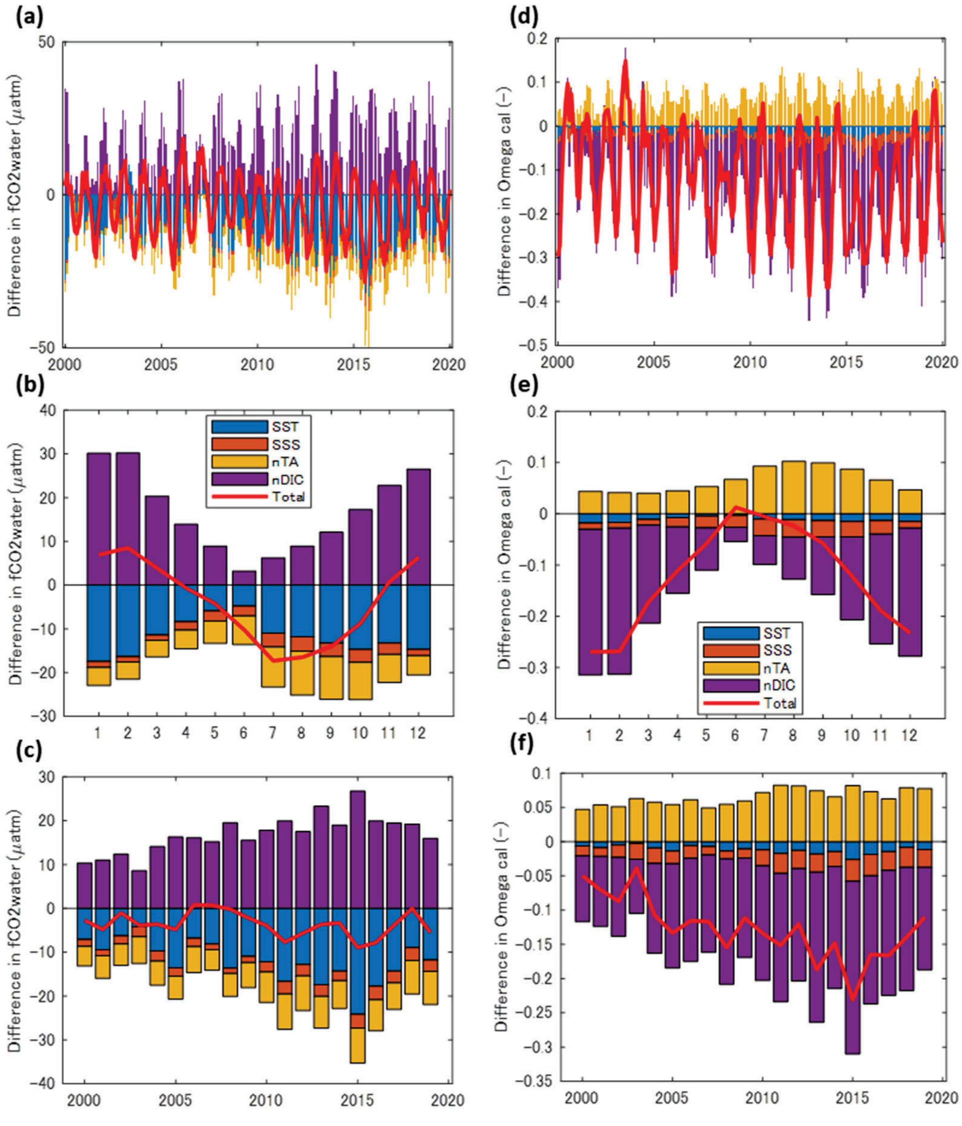
derivative

The PLS regression showed that the explanatory variables SST, SSS, nTA, and nDIC explained well the objective variables seawater fCO_2 ($r^2 = 0.996$) and Ω_{cal} ($r^2 = 0.997$) (Figure 5). The average contributions due to each explanatory variable in the PLS analysis were consistent with those in the equilibrium calculation for seawater fCO_2 and Ω_{cal} using the averages in Area B and the respective *dAB* values (Table S2). Area A had lower seawater fCO_2 and Ω_{cal} than Area B and both seawater fCO_2 and Ω_{cal} showed clear seasonal variations.



Figures 5. (a): Temporal contributions in the PLS analysis by SST, SSS, nTA, and nDIC to the difference in seawater fCO_2 between Area A and B. (b): The corresponding monthly averages. (c): The corresponding yearly averages. (d): Temporal contributions in the PLS analysis by SST, SSS, nTA, and nDIC to the difference in Ω_{cal} between Area A and B. (e): The monthly averages. (f): The yearly averages. . The red lines represent the sum of each contribution and is almost equal to temporal variation of *dAB* of seawater fCO_2 and Ω_{cal}.

4. Discussion

The extent of Area A defined using nTA (Figure 2f) was consistent with the low-salinity area of the Pacific coast of Japan (Figure 1a). The SSS south of 37°N exhibited an abrupt change in value 205.74 km from mainland Japan (Figure S4), which is consistent with the distance of the boundary between Areas A and B (250 km). SSS is affected by precipitation and evaporation; therefore, nTA was a more accurate indicator of riverine water than SSS. However, the consistency between the two results provides strong evidence that the nTA in Area A was derived from riverine water. These distances were calculated statistically using 20 years of observational data on SSS and nTA. It would be challenging to obtain a similarly significant result from single observations. Therefore, the findings of this study, which quantify the extent of the influence of riverine water, are novel and offer new insights.

To evaluate the effect of the Kuroshio Current on riverine water distribution, another EOF analysis was performed using data from the Kuroshio Large Meander period (2017–2020 in this study) in which the Kuroshio path followed an alternative meandering path south of its usual course (Kawabe, 1985). Area A was within the range of 350 km from land, utilizing the Mode 1 EOF anomaly during the meander period, whereas it was 250 km during the entire period. This result was consistent with the observed Kuroshio meandering path and supports the assumption that the Kuroshio Current effects Area A. However, Area A extended further out of the open ocean than the Kuroshio axis during the entire study period (Figure 2f). Therefore, the Kuroshio Current limited the spread of riverine water to some extent but did not completely inhibit it.

The supply of TA by riverine water was higher than the time deviation of *dAB* for almost the entire period, suggesting that the supply was one of the main factors contributing to the increase in *dAB* (Figure 4). On the other hand, the annual rate of increase in the *dAB* of nTA was almost zero (-0.02 ± 0.05 μmol kg^{-1} $year^{-1}$). Therefore, the residual term would be the negative value of the same scale with Sup_{rw} term for TA. The primary cause of the negative residual term is assumed to be horizontal advection. The advection effect can be estimated from the product of the current velocity and *dAB* of nTA. The period of increasing *dAB* of nTA (Figure 3c) is con[illegible] 1 with the period of increasing absolute value of the residual term in Figure 4b. In addition, vertical advection could n[illegible] the main cause of the negative residual term, because the vertical gradient for calculating vertical advection was small and insignificant (see Text S1). Other 2 causes of the residual term are differences in biological metabolism, such as calcification and nitrate consumption; 3 ~~[illegible]ication by coccolithophores is the most promising process for changing oceanic nTA~~. However, no regional difference in calcification rate of up to 10 μmol kg-

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番号 : 1　作成者 : Frédéric Gazeau　日付 : 2025/10/08 17:44:23

?

作成者 : Tatsuki Tokoro　タイトル : ノート注釈　日付 : 2025/10/10 15:49:30

We have rewritten as "On the other hand, the effect of vertical advection estimated from the vertical gradient of nTA and mixed layer depth was not significant (see Text S1)."(Line 292-293)

番号 : 2　作成者 : Frédéric Gazeau　日付 : 2025/09/22 19:01:32

calcification by coccolithophores is one of the most important process driving changes in oceanic nTA

番号 : 3　作成者 : Frédéric Gazeau　日付 : 2025/09/22 19:01:24

[1] year^{-1} (Figure 4b) has ~~not~~ been reported between the Japanese coastal Area A (Area A) and the surrounding sea area (Area B) (Hopkins and Balch, 2018; Krumhardt et al., 2019). Another component of TA include nitrate, which is consumed during photosynthesis to raise nTA (Brewer and Goldman, 1976). The data on total nitrate (TN) concentration is available in GLODAP, and the mean values were 1.19 ± 0.11 μmol kg^{-1} and 0.34 ± 0.04 μmol kg^{-1} in Area A and B, respectively. As the monthly difference were largest in the winter (2.11 μmol kg^{-1}) and almost reach zero in the summer, the main source of nitrate would be vertical advection rather than input from land. Nonetheless, the effect of nitrate on TA was probably one order of magnitude smaller than that of the riverine water supply and is negligible as the main source of nTA.

The DIC variation is more complicated than the TA variation because the spatiotemporal variation in biological metabolism and vertical advection is larger and more significant than that of TA, in addition to the effect of CO_2 exchange with the atmosphere. Unlike the TA, the seasonal variations in the riverine water supply were not consistent with those in the time derivative (Figure 4e). This trend indicated that riverine water supply was not the primary driver of DIC variation in Area A. The effect of oceanic CO_2 uptake was highly variable and unclear on a decadal scale. However, on a seasonal scale, it was more distinct, resulting in negative shifts in the summer residual terms and positive shifts in the winter residual terms. This seasonal variation indicated that the oceanic CO_2 uptake in Area A was larger than that in Area B in summer, and vice versa in winter. This was caused by the lower seawater fCO_2 in Area A during the summer season and higher wind velocity in Area B during the winter season. Consequently, the residual term of nDIC indicated a strong and complex seasonal variation, which included a maximum in the winter and two minimums in spring and summer with a difference of 100 μmol kg^{-1} year^{-1}. Contrary to TA, DIC was affected by nutrient and organic matter loading from terrestrial sources, and was strongly affected by differences in biological metabolism between Areas A and B. For DIC, in contrast to TA, a significant vertical profile trend was observed (Text S1); consequently, some of the residual terms might have been influenced by vertical advection.

To quantify the effect of biological metabolism, the effects of horizontal and vertical advection were estimated based on the following assumptions: 1) the residual term for nTA (C_{res_TA}) was assumed to be equal to the horizontal advection term, based on the considerations that the effects of vertical advection and biological metabolism on nTA are negligible. 2) The horizontal advection term was proportional to dAB of nTA (dAB_{TA}) and nDIC (dAB_{DIC}). Several previous studies have used normalized values for the calculation of TA and DIC advection (Broecker and Peng, 1992; Keeling and Peng, 1995) and have also proven their validity as an approximation (Robbins, 2001). Because the advection term is calculated as the product of concentration gradient and flow field, and the associated distance and flow velocity are the same at dAB_{TA} and dAB_{DIC}, it can be assumed that the horizontal advection effect on DIC (C_{hadv_DIC}) can be estimated using the following equation:

$$C_{hadv\_DIC} = K\frac{dAB_{DIC}}{D_{AB}}$$

$$C_{res\_TA} = K\frac{dAB_{TA}}{D_{AB}}$$

$$C_{hadv\_DIC} = C_{res\_TA} \cdot \frac{dAB_{DIC}}{dAB_{TA}} \quad (5)$$

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番号 : 1　作成者 : Frédéric Gazeau　　日付 : 2025/09/22 19:02:30

番号 : 2　作成者 : Frédéric Gazeau　　日付 : 2025/09/22 19:02:55

What about organic alkalinity?????

作成者 : Tatsuki Tokoro　　タイトル : ノート注釈　　日付 : 2025/10/10 15:49:11

Thank you for your valuable comment. As you rightly pointed out, it was an oversight not to address organic Alkalinity in the analysis of land-derived TA. The sentences about organic Alkalinity has been added at the end of this paragraph.
The added sentences state that, based on referential values for organic Alkalinity from mainland China (no Japanese data could be found), it was considered negligible under the conditions in this study. It also adds that it could have a significant impact on TA budget analysis in environment with lower salinity or abundant wetland such as Hokkaido -which were not included in this study. (Line 302-312)

番号 : 3　作成者 : Frédéric Gazeau　　日付 : 2025/10/09 11:19:26

Nonetheless

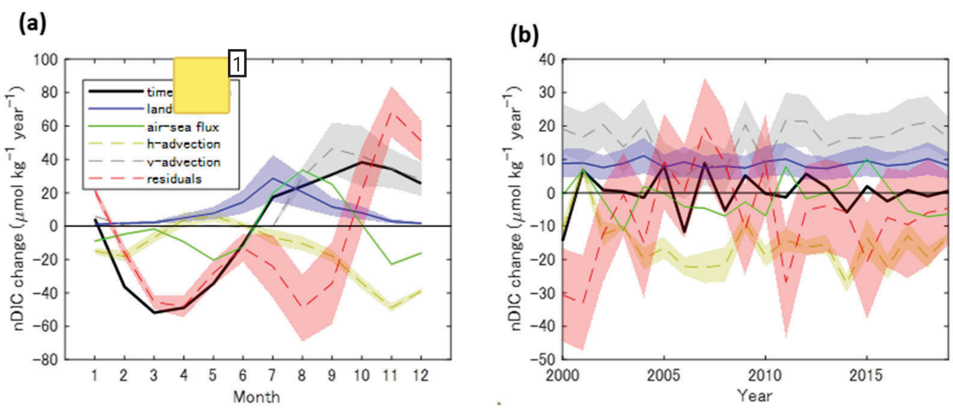
where, K and D_{AB} are the index values associated with the current field and horizontal distance between Areas A and B, respectively. These parameters were assumed to be the same for TA and DIC calculations. 3) Vertical advection for nDIC (C_{vhadv_DIC}) was estimated from mixing with subsurface water when the mixed layer was deepened. Specifically, we used a simplified equation of the method of a previous study (Ishii et al., 2001).

$$\int C_{vadv_{DIC}}(t)dt = \frac{\{\Delta nDIC(t+1)+\Delta nDIC(t)\}}{2} \times \frac{\{MLD(t+1)-MLD(t)\}}{\{MLD(t+1)\cdot\rho_{MLD}(t+1)\}}$$

$$\Delta nDIC(t) = \{\mathrm{nDIC}(July, MLD(t)) - \mathrm{nDIC}(t, MLD(t))\}\cdot\rho_{MLD}(t) \quad (6)$$

where, $\Delta nDIC(t)$ is the difference between the average nDIC in July ($nDIC(July)$) and the nDIC at time t at mixed layer depth ($MLD(t)$) multiplied by the seawater density at $MLD(t)$ ($\rho_{MLD(t)}$). The nDIC profile at a depth below the mixed layer was assumed to be maintained at its average value in July; as the mixed layer deepened, the difference from the July profile was added to the nDIC in the mixed layer. The effect of vertical advection on nDIC was negligible when the mixed-layer depth did not change or became shallower. The details on the calculation of the nDIC profiles are provided in Text S1.

The effects of horizontal and vertical advection on nDIC are shown in Figure 6. However, due to the large error range in the vertical advection term for each time step (the median was 137%), only the monthly and annual averages are shown in the figure. The horizontal and vertical advection effects were identified mainly from autumn to winter with an annual mean value of -15.55 ± 1.35 μmol kg^{-1} $year^{-1}$ and 15.55 ± 2.25 μmol kg^{-1} $year^{-1}$, respectively (Ave ± SE). Seasonally, the residual term had a maximum in the winter and two minimums in spring and summer, with an overall mean of -7.30 ± 3.24 μmol kg^{-1} $year^{-1}$.



Figures 6. Temporal variations of nDIC time derivative and related terms in Equation (2) (a): The monthly averages. (b): The yearly averages. The shaded areas of horizontal advection (h-advection) and vertical advection (v-advection) are the random error calculated using the error propagation (see Text S1). Therefore, unlike the error ranges of Sup_{rw}, these were reduced by $10^{-0.5}$ or $20^{-0.5}$ by monthly or yearly averaging, respectively.

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derivative

Although the above results were not definitive because of many assumptions and large error ranges, the fact that the final residual term on the decadal scale was almost negative supports the idea that Area A is likely under heterotrophic conditions within the mixed layer. The positive shift in the residual term before and after 2007 was due to the low vertical advection term caused by the small MLD during winter. The 2007 minimum SST in Area A (19.41 °C) was the highest of the overall annual minimum SST (17.71 ± 0.66 °C, Ave ± SD), and thus the surface seawater mixing would be weaker than that in other years. Therefore, the residual term for this period does not necessarily indicate changes in biological metabolism. However, the high SST during winter suggests that the decomposition of biofixed carbon may be accelerated. Seasonally, the March-April minimum in the residual term was assumed to be the result of phytoplankton blooms in the open ocean area in Area A. Another minimum in July-August was assumed to be due to the primary production of phytoplankton and submerged aquatic vegetation under eutrophic conditions in the inner bays and near-shore areas. The winter maximum can be explained by the upwelling of organic matter and the decomposition of bio-fixed carbons within the mixed layer.

It should be noted that the residual term above includes the effects of–the water CO_2 flux in near-shore areas. However, unlike the air-sea CO_2 flux in the ocean (C_{flux} in Equation (2)), the air-water CO_2 flux in near-shore areas is very difficult to quantify because observations of water fCO_2 and the physical regulating factor defined as "transfer velocity" (e.g., Wanninkhof, 2014) are limited. The estimation of this CO_2 flux varies widely among the existing studies. For example, global average models (Aufdenkamp et al., 2011; Tranvik et al., 2009) have estimated that approximately half of the carbon supply from land is released into the atmosphere via the air-water CO_2 flux in near-shore areas. However, the inner bays affected by riverine water in this study showed a trend in atmospheric CO_2 absorption (Tokoro et al., 2021), that is unlikely to follow the global average trend. Therefore, enhanced observation and analysis of atmospheric CO_2 exchange in nearshore areas is essential for a more accurate assessment of the residual term as an indicator of biological metabolism.

Despite Area A having a higher nDIC than Area B (Figure 3), seawater fCO_2 in Area A tended to be lower than in Area B (-3.61 ± 0.70 μatm). This was mainly because of the lower SST and higher nTA in Area A than those in Area B (Figure 5). The low SST can likely be attributed to riverine water, which is mainly supplied by snowmelt and rainfall from the mountains and highlands and therefore tends to have lower original water temperatures. In particular, the short flow paths of Japanese rivers can likely limit the effects of heating due to solar radiation and other factors. The three inner bays with strong river influence had lower average water temperature (18.66–19.23 °C, Tokoro et al., 2021) than areas A (22.02 °C) and B (22.86 °C), which support our assumption. The decrease in seawater fCO_2 peaked in the summer (-15.90 ± 4.72 μatm from July to September), when the decrease in nDIC due to biological metabolism and the increase in nTA due to river supply coincided. However, this decreasing in seawater fCO_2 had little effect on annual oceanic CO_2 uptake in Area A. Compared to the hypothetical case where riverine water did not affect seawater fCO_2 (seawater fCO_2 in Area A were the same as that in Area B), the change in air-sea CO_2 flux would be similar (0.00 ± 0.33 mol m^{-2} $year^{-1}$). This was because the decrease in air-sea CO_2 flux in summer almost offsets the increase in winter. Although the increase in seawater fCO_2 in winter was smaller than the decrease in summer, the air-sea CO_2 flues in summer and winter were coincidentally balanced, owing to higher wind speeds in winter.

For Ω_{cal}, seawater in Area A was notably more acidified than in the surrounding sea area, based on a 20-year average (-0.13 ± 0.01). The impact on coastal acidification was particularly significant in winter, with an average decrease of -0.25 (5.33 to 5.08). nTA was only mitigation factor for coastal acidification among the explanatory variables, and reduced acidification due to other factors to 65% (-0.20 to -0.13). This coastal acidification was found to be progressing, with *dAB* of Ω_{cal} decreasing by -0.52 ± 0.14 per decade. This can be attributed to an increasing trend in *dAB* of nDIC (0.32 ± 0.08 μmol kg^{-1} $year^{-1}$) due to the trend of increasing precipitation in Japan.

5. Conclusions

In this study, we identified areas affected by riverine water in the Northwest Pacific Ocean by using statistically processed observational data. We also evaluated the contribution of riverine water to oceanic CO_2 uptake and coastal acidification by comparing the TA and DIC values in the sea area affected by riverine water and surrounding sea areas.

The Area A affected by riverine water (Area A) was within 250 km of mainland Japan and, to some extent, along the Kuroshio axis. This area was consistent with the low-SSS area on the Pacific coast of Japan. In addition, the range increased to 350 km during the period when the Kuroshio Current meandered south, indicating that the Kuroshio Current path influences the spread of riverine water.

Both nTA and nDIC were higher in surrounding sea of Area A. The main source of TA was riverine water in summer, which was balanced by a decrease due to horizontal advection in autumn and winter. The DIC flows were more complex than the TA flows and were more strongly affected by a residual term than by the riverine water supply. The contribution of biological metabolism is expected to be more influential on the residual term after excluding the horizontal and vertical advection effects. Biological metabolism showed a maximum in winter and two minima in spring and summer. The annual mean suggests heterotrophic conditions within the mixed layer in Area A. In any case, because the air-water CO_2 flux in near-shore areas is still difficult to estimate, a more thorough quantification of DIC flow in relation to riverine water needs to be considered.

Seawater fCO_2 in Area A decreased mainly in summer because of the supplied riverine water, which has low-temperature water with high nTA. However, riverine water supply was found to have virtually no effect on oceanic CO_2 uptake at an annual scale. This is because CO_2 emission was enhanced by strong wind speeds in winter, in addition to the relatively small increase in seawater fCO_2 in winter. The change in the air-sea CO_2 flux in winter was coincidentally balanced by the change in summer, and the annual average was almost zero. Although acidification progressed more in Area A than in the surrounding sea area, the supply of TA by riverine water mitigated the acidification to 65%. However, an increase in precipitation may have led to increases in nDIC and acidification in Area A.

This study makes a significant contribution to the analysis of carbon flows in the boundary seawater between terrestrial and oceanic areas because the quantification of carbon flows in these areas is often uncertain in space and time. Enhancing the observational data allows the results to be spatially extended to larger regional or global scales. The analysis results are expected to have a smaller error range owing to the high spatiotemporal resolution of the vertical profiles of the carbonate data and air-water CO_2 flux data in near-shore areas.

Data Availability

The SSS, SST, and fCO_2 in air and seawater datasets are available in the SOCAT database (Pfeil et al., 2013; Bakker et al., 2016, https://socat.info/index.php/data-access/). The TA and DIC data were available from the GLODAP database version 2.2020 (Key et al., 2015; Olsen et al., 2016; Olsen, 2020, https://glodap/info/). Wind speed data were obtained from the CCMP database (version 2.0) (Atlas et al., 2011; Mears et al., 2019; https://data.remss.com/ccmp/v02.0/). The river flow datasets were obtained from the Water Information System of the Ministry of Land, Infrastructure, Transport, and Tourism of Japan (http://www1.river.go.jp/). Vertical water temperature datasets for calculating MLD were obtained from the JCOPE2M dataset (Miyazawa et al., 2017, 2019, . https://www.jamstec.go.jp/jcope/htdocs/distribution/).

Author Contribution

TT designed the study and wrote the first draft on the manuscript. SN, ST, SS, SD, KE, MI, NK, TO, KT, and YN supplied and managed data for SOCAT and GLODAP. SN supported data processing. All authors contributed to manuscript writing and proofing.

Competing interests

The contact author has declared that none of the authors have any competing interests.

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