Response to reviewers' comments on "Variability and drivers of carbonate chemistry at shellfish aquaculture sites in the Salish Sea, British Columbia"

We thank the reviewers for their thoughtful and thorough comments, which have improved the overall quality of this manuscript. Here we repeat reviewer's comments in bold and provide our response below in normal font.

We have grouped together reviewer's comments regarding the results section, which overlapped and which was the largest revision piece, as have presented this first. We provide examples of changes made following the reviewers' comments, but refer the reader to the tracked changes manuscript for the full results revision.

Reviewer 1: "The ms presents interesting and relevant data and elaborations addressing the issue of the variability of the CO2 system at shellfish aquaculture sites and the relevance of the main drivers. The authors base their study on 14 campaigns over a period of 4 years. They address both seasonal (two season) and diel variability considering two depth (surface and midlayer) at 4 study sites. The data presentation is clear and the ms is well structured.

Reviewer 1 Comment 1: The ms has a long descriptive part which could be more appropriate for the technical report than for ta scientific paper. It could be summarized, in particular, when presenting the saturation state o aragonite and calcite which have quite similar variations as shown in figures 2 and 3."

Reviewer 2 Comment 1: The article is well-written, even if it is slightly lengthy. I recommend it for publication with the following minor modifications: The detailed presentation of numerous results could be condensed to focus primarily on the origins of variations."

Reviewer 2 Comment 8: L 260: It is quite difficult to compare surface and mid-layer water differences. It would be nice to gather Fig. 2 and Fig. 3 to easily distinguish the major difference. Thus, the description of these data could be summarized.

We agree that the results section could be refined. We have edited the results section to be more succinct, taking the reviewers' suggestion to combine the calcite and aragonite results. We have reduced text by focusing on the key findings and patterns of variability, as well as by reducing detail in the description of minor drivers. We have also combined the presentation of surface and mid-layer results as suggested by Dr. Sebastien Petton. By doing so, having both mid-layer and surface results and discussion presented together will also allow for better and easier comparison between the two layers, which we hope will address Dr Petton's point 8 regarding the combination of figures.

While we appreciate that compiling both figures 2 and 3 together may make some comparison of different depth layers easier, we feel that both figures are already busy with 10 panels each, now with the addition of dissolved oxygen requested by reviewer 1. We feel that combining the two would make the plots difficult to read. We considered breaking the figures into four so that each row would become a separate figure, which would allow space for both the surface layer and mid-layer to be side by side. However, this change would remove the ability to easily identify similar or different patterns of variability in physical, chemical and biological drivers of carbonate chemistry, which we feel is the main focus of this paper. We would therefore like to keep the division of sub-panels in Figures 2 and 3 as they are. We will however revise Figures 2 and 3 to ensure that corresponding subplots have the same scale on the y-axis (i.e., DIC range in the surface will be the same as the DIC range in the mid-layer), for easier comparison of the results. We will revise the caption to point out that these scales are the same.

We have revised all sections of the results accordingly. For example we have revised section 3.1.1 (line 300) so that it now reads:

"Surface temperatures (T) experience modest variability across all locations in winter, ranging from ~6 to 11 °C, with similar median T ~9 °C across sites (Fig. 2b). The lowest surface T variability of the nearshore locations is found in Sansum Narrows where tidal mixing is strong. Winter mid-layer T are warmer than the surface (~1°C) and exhibit lower variability (Fig. 3b). Summer surface T are higher (median ~15 to 19 °C) reaching up to 22 °C in Baynes Sound and Evening Cove beach. Summer T are also more variable than in winter, with particularly large ranges in surface T at Evening Cove beach and Sansum Narrows (Fig. 2b). In our observations, Baynes Sound surface T are the highest and we observe cooler summer T in Okeover Inlet, with one exception - the unusual conditions that occurred during the 2016 coccolithophore bloom (NASA, 2016), when summer T were unusually high (up to 22 °C). Although summer surface T exhibit a similar range in the SOG and nearshore sites, median summer surface T in the SOG are cooler than in the nearshore in our observations. Summer mid-layer variability is lower than surface variability, ranging from 8 to 20 °C; and mid-layer T are ~4 °C cooler than surface T."

In Section 3.1.6 where we describe the aragonite and calcite results separately we have condensed these results into two paragraphs instead of four, to read as following:

"Surface Ωa and Ωc follow a similar variability pattern although their absolute values differ (i.e., Ωc is greater; Fig. 2i,j). Winter surface and mid-layer Ω are lower than summer surface Ω. Almost all locations, nearshore and the SOG, are undersaturated throughout the water column with respect to Ωa in winter (Fig. 2i), with only a few outlying samples that are supersaturated. While median winter surface Ωc are slightly greater than the saturation threshold (Ωc = 1), there is some Ωc undersaturation in our winter data at nearshore sites, particularly at Baynes Sound and Okeover Inlet (Fig. 2j), with the beach sites being the only nearshore locations to not experience any Ωc undersaturation in our data. Median surface winter Ωa is similar across all sites (~0.8) and absolute variability is comparatively low relative to summer at all nearshore locations.

Summer Ωa is high at our nearshore locations, which are mostly all supersaturated and reach values as high as 3.2 (Okeover and Baynes Sound). However, there is some summer surface undersaturation (Ωa) in Sansum Narrows and in the SOG, where Ω are typically lower (Fig. 2i,j). Ωc is supersaturated at all locations, and values and variability are much higher than in winter (Fig. 2j), with maximum values reaching ~ 4.5 to 5. High Ωa in Okeover Inlet stands out from other nearshore locations, where the highest median Ωa are found at Okeover beach and Okeover Inlet (>=2.8, Fig. 2i). Following patterns in pH and oxygen, summer mid-layer Ωa are typically lower than in the surface, when the system is stratified and productive (and mid-layer summer saturation states in Sansum Narrows are similar to surface values). Variability in the mid-layer is similar to the surface, with variability being higher in Okeover Inlet and Baynes Sound, and lower in Sansum Narrows and the SOG."

Here we combine reviewer comments regarding the conclusions and implications sections and shellfish mortality as there was some overlap:

Reviewer 1 Comment 5: "The conclusions are not coherent with the main objectives, they seem to be more implications deriving from conclusions. The chapter named "implications" instead contains conclusions both should be revised in order to provide more clearly the conclusions related to the objectives of the ms."

Reviewer 2 Comment 2: Additionally, the "Implications" section contains assertions that may be too strong in relation to the manuscript's presentation and should be rephrased.

Considering the data presented, it is understandable to want to relate them to shellfish farming conditions. According to the study, I agree that shellfish farming in deeper zones indeed appears to offer an opportunity to locally mitigate the effects of acidification and carbonate depletion. However, introducing the issue of mortality seems somewhat ambitious given the provided data. Even if this is not my area of expertise, diseases for these species can manifest with threshold effects, where the mean values may not be the sole determining factor. Moreover, there could be other complex physiological impacts associated with changing environments.

Reviewer 1 Comment 3: "The authors outline that there has been shell fish mortalities attributed to temperature and diseases in the study area however the info is very generic,

some more detail on the organisms which caused the mortalities (of both natural and cultivated clams and oysters?) in the area would be useful to understand the potential relationship with temperature increase."

Thank you for pointing out this opportunity to provide greater detail and description of what is known regarding the organisms (i.e., bacteria and viruses) recorded in BC that could be contributing to mortality in the introduction. We will add the following additional information in line 78:

"Shellfish mortality has become a global issue and a recurring challenge during summer in the Salish Sea, which has been attributed to temperature stress, disease, and harmful algal blooms (King et al., 2019; King et al., 2021; Cowan, 2020; Morin, 2020). In the SOG, large scale die-off events of cultivated C. gigas have been reported (e.g., Drope et al., 2023; Morin, 2020, Cowan, 2020). The cause of these mortalities is not well understood, but mortalities have been linked to elevated water temperatures and the presence of the marine bacteria *Vibrio aestuarianus* (Cowan, 2020); as well as OA (Drope et al., 2023), although the role that changing carbonate chemistry conditions contributes to these mortalities in the SOG remains unknown."

We agree that the final sections needed restructuring. We have combined and renamed this section "Conclusions and Implications" and ensured that the main objectives and questions are addressed at the beginning, with implications of these conclusions at the end of the section. To address Reviewer 2's comment no. 2, we have also focused on the findings of the paper, taking care that our discussion of implications does not extend outside of the scope of our study (please see below).

We agree that some assertions in the Implications section were too strong. In particular, the first submission placed too much emphasis on mortality of shellfish and disease as these issues were not explored within this study. They played a role in motivating this study and we have edited the text accordingly to reflect these facts. Specifically, the cause of oyster mortality in BC is not well understood but is currently an issue in the Salish Sea. Our determination of carbonate chemistry conditions in the major grow areas (and estimation of key drivers) suggests that acidification is not likely the key culprit, even though it may contribute as a multi-stressor. We did not extrapolate beyond providing references to the literature which discusses disease as a possible cause in the revision.

For example, in response to reviewer 1's comment no. 3, we have added detail as to the current state of understanding of the organisms affected (Pacific Oysters). We will also include the link between the presence of *Vibrio aestuarianus*, harmful algal blooms and elevated temperatures in the introduction to provide greater context.

Other key revisions include the abstract. Specifically, we have removed reference to disease and mortality because we do not directly study these issues. We will revise the final sentences:

From: "Shellfish mortality events coincide with highly favourable pH and Ω conditions during summer and are most likely linked to high surface temperatures and disease rather than ocean acidification. To reduce shellfish mortality, shellfish could be hung lower in the water column (5–20 m) to avoid high temperatures and disease, while still experiencing favourable pH and Ω conditions for shellfish."

To: " We find that during summer at mid-depth (5–20 m), where it is cooler, pH, Ω, and oxygen conditions are still favourable for shellfish. These results suggest that if shellfish are hung lower in the water column, they may avoid high sea surface temperatures, without inducing OA and oxygen stress."

In the Implications section, we have removed the sentence in line 879 (original document), where we suggest that chronic exposure to low saturation states could make shellfish more susceptible to disease, as this statement is largely conjecture. The two paragraphs beginning on lines 784, and 794 (new document) which discuss mortality will be edited so that they would read:

"Although OA may cause stress by increasing energy expenditure in shellfish (e.g., Pousse et al., 2020), OA does not appear to be directly responsible for mortality events in our region. Most shellfish mortality events recorded in the Salish Sea have occurred in summer (Cowan et al., 2020; Morin, 2020; King et al. 2021) when pH and Ωa are relatively high, and not in winter when chronic undersaturation of Ωa and some Ωc undersaturation occurs (Fig. 3b,c). Higher temperatures linked to disease appear to be a more immediate concern to shellfish growers in the Salish Sea (e.g., Morin, 2020). It is possible that wild shellfish have adapted to, or that commercial shellfish species are already tolerant of, this chronic exposure to lower Ωa conditions in winter (e.g., Waldbusser et al., 2016). Additionally, values of Ωc (which are mostly supersaturated) rather than Ωa are likely more relevant to shellfish during winter because juveniles are typically out-planted in summer and have reached maturity and transitioned to calcite structures by winter (e.g., Stenzel 1964).

Growers may wish to consider placing shellfish, especially juveniles, deeper than the surface layer in summer where temperatures are lower, and oxygen and carbonate chemistry conditions are still favourable for shellfish growth. Temperatures in the mid-layer are cooler, and although pH tends to be slightly lower, the mid-layer mostly remains supersaturated with respect to both Ωa and Ωc in summer (Fig. 2, 3). In addition, beaches do not appear to have a clear advantage over tray hang sites in terms of carbonate chemistry. However, beach sites experience the highest temperatures of all locations and may become less favourable locations in the future as temperature rises (e.g., Hesketh and Harley, 2023). Indeed, extreme heat events have already caused mass mortalities of invertebrates in the inter-tidal areas of the Salish Sea (White et al., 2021)."

Here we continue to list and address remaining reviewer comments chronologically.

Reviewer 1 Comment 2: "The authors discuss the biological role assessing that "DIC drawdown by primary production is the dominant driver of seasonal and diel pH and carbonates saturation state changes at nearshore locations but they do not present dissolved oxygen data among the Biologically significant parameters (figures 3 and 4) but only in the figure A10 in the Appendix (not very easy to read) instead a better representation of the seasonal variation in the different site would be very useful for the discussion where may variation are explained on the basis of the change in primary productivity.

Regarding the oxygen saturation presented in the Fig A10 It would be interesting to explain the existence in Summer at Okeover inlet of surface waters where there are both strong oversaturation and at least there cases of anoxia but no minima of pH."

R1 makes a great point and we agree. These (discrete) O2 data are valuable and were collected with great care. We have added a dissolved oxygen panel (% saturation) to each of Figures 2, 3 and 4. With the addition of the dissolved oxygen to these figures, we have also add brief descriptions of oxygen variability (where it was/is missing) to the results and discussion, keeping in mind the need for the results section to be made less dense.

We have added oxygen sections to the seasonal (3.1.4) and diel results sections (3.3.4) as follows:

Winter surface dissolved oxygen (DO; expressed as % saturation) are mostly undersaturated (i.e., <100%, with medians ~ 75-85%) at our nearshore locations; and variability is low across sites (Fig. 2a). Sansum Narrows has relatively low DO compared to the other nearshore locations. Mid-layer winter DO is lower than in the surface (< 80%) (Fig.3a). In summer, surface DO is mostly supersaturated, with high DO and large variability, especially in Okeover Inlet. However, the well mixed Sansum Narrows location has the lowest DO, which is mostly undersaturated (Fig. 2a, 3a). Mid-layer DO in summer is also higher than it is in winter. At times this depth zone includes a strong oxycline and ranges from supersaturation to undersaturation (although still oxygen replete, lowest DO usually > 70% saturation, Fig. A1, 3a). Ranges of DO in the SOG are similar to those in the nearshore in winter, but are lower in summer (Fig. 2a, 3a).

Winter DO was undersaturated on our sampled days in Baynes Sound and Sansum Narrows, with little variability over the day at both locations (Fig, 4a). The beach location in Evening Cove however, has higher DO on the sampled day, with some oversaturation occurring in the afternoon. Dissolved oxygen tends to increase throughout the day at the beach site in winter, and all of our nearshore locations in summer, when there is widespread DO supersaturation. Sansum Narrows has the lowest summer saturation state of the four locations, with a smaller increase over the day, resulting in lower variability.

We thank R1 very much for their care. The cases of anoxia (shown in Okeover) highlighted by the reviewer in Figure A10, are erroneous. These errors resulted from Niskin casts where dissolved oxygen data were missing, and should have been removed at the QA/QC stage. We apologize for this oversight and have removed these data and double checked our QA/QC code, to ensure that there are no other ‘missed samples’ included in these plots.

Reviewer 1 Comment 4: “The authors should specify in the ms at least on which scale of pH they have chosen, the reader has not to go to another paper to know this.”

We used the total pH scale and have added this detail to the methods in line 180, thanks for pointing this omission out.

Reviewer 1 Comment 6: “Tables 1 and 2. it is not clear what the number between parenthesis represent.”

The value between parenthesis in tables 1 and 2 is the estimated uncertainty associated with the entry, we have added this clarification to the table captions.

Reviewer 1 Comment 7: “Lines 684-685. It is not clear what is the meaning of “is highly sensitive to carbonate space”.”

In lines 684-685 we agree that some greater clarification is needed to explain what is meant by sensitive carbonate space. We are referring to the point in the carbonate system where small changes in DIC result in large changes in pH. We have added text to clarify this so that this line now reads (line 611 in new document):

“TA is also relatively high, and DIC:TA ratios are close to 1, which places the Salish Sea in highly sensitive carbonate space, where small changes in DIC result in large changes in pH and Ω (e.g., Egleston et al., 2010; Hu and Cai, 2013)”

Reviewer 1 Comment 8: “Line 896. Only omega calcite undersaturation occur in winter or also omega aragonite?”

The text (line 896) would benefit from clarification. We added that both aragonite and calcite undersaturation occurs, while highlighting that the occurrence of calcite undersaturation is considered unusual. We have also added a citation to support our assertion about the future (which is not a direct result of our research). We will edit this line from:

“Some Ω_c undersaturation already occurs in winter, and these conditions will become more common and widespread, increasing stress for adult shellfish in the winter season.”

To (line 804 in new document): “Chronic Ωa, and even some Ωc undersaturation already occurs in winter. Undersaturated Ωc conditions will likely become more common and widespread in the future (e.g., Hauri et al., 2013), increasing stress for adult shellfish in the winter season.”

Reviewer 1 Comment 9: “Table A2. Caption Suggest to specify that end members are related to freshwater and seawater.”

Agreed. We have added this detail to the table caption, stating that endmembers are from fresh and salty sources, as well as another column to the table which indicates whether the endmember is fresh water or salt water.

Reviewer 1 Comment 10: “Table A4. PSU is adimensional therefore remove the “unit” specify in the methods section that how salinity is expressed.”

Agreed, we have removed the word unit from this table; and mention in the methods that we report salinity on the practical salinity scale, which is unitless in line 182 of the revised document.

Reviewer 1 Comment 11: "**Table A5** a**lign the numbers in the column with the title of the column."**

Thank you for identifying this format issue, we have aligned numbers in the table to centre.

Reviewer 1 Comment 12: "Table A6 I wonder the reason for expressing Temperature uncertainty on the basis of the instrumental uncertainty whereas salinity uncertainty on the basis of geometric mean pooled deviations of replicate pairs of all the campaigns."

Salinity was measured in discrete bottle samples, whereas our temperature values are from the CTD instrument. It was critical for this study to have accurate salinity values for each discrete DIC and TA bottle sample for our normalisation - in our region it is challenging to be S-accurate with CTD-S given the strong vertical stratification. The uncertainty of the salinometer is captured in the uncertainties from the pooled standard deviation of replicate pairs. We could not apply a similar method to temperature as we only have CTD profile data, for which we cannot pool replicates. We wanted to be thorough and include an uncertainty estimate for temperature as well as other drivers, and so included the instrument uncertainty. We have added clarification to the table caption to be clear that salinity data are from discrete bottle samples.

Reviewer 1 Comment 13: "Tables A9 and A10. Explain in the caption the significance of the numbers between parenthesis and those in bold."

The numbers in parenthesis are uncertainty values, the numbers that have been bolded are changes larger than uncertainty. We have added this clarification to the table captions.

Reviewer 1 Comment 14: "References**: Check and correct the subscript for "CO2""**

Thank you. We have carefully checked and corrected all references with 'CO2' so that '2' is subscript.

Reviewer 2 Dr Sebastien Petton

"The paper by Simpson et al. presents a highly interesting study aimed at characterizing and explaining the variability of carbonate cycle parameters, specifically pH and Ω, in coastal areas. Based on data collected during campaigns spanning from 2015 to 2018, the authors define the origins of daily and seasonal variations across shellfish production sites. They compare conditions observed in surface waters (next to shellfish farmings) with those in a transitional zone (mid-layer). The skillful application of Taylor series expansion for signal decomposition allows for a brilliant discernment of the contributions from environmental factors, highlighting the pivotal role of biological activity as the primary driver of variability.

Comment 1 is addressed above within the results section revisions.

Comment 2 is addressed above within the conclusions and implications section revisions.

Comment 3: L 108: Remove this sentence "Shell midden have been suggested…" as it is not a key element for the manuscript.

We will remove this sentence if the reviewers believe that the discussion of shell middens is not relevant. We have included this sentence as shell middens are locally important and common along the coast of British Columbia and as such are being considered for their potential to mitigate the effects of OA (Doyle and Bendell, 2022; Kelly et al., 2011). The findings of this paper show no TA increase (and therefore no TA driven changes in pH or saturation states) at the shell midden site. We believe this result is interesting and useful; and we would argue that this point could be retained.

To strengthen this point, we have added in line 112 that "Shell middens are prominent features along the coast of British Columbia, and are being evaluated for their potential for mitigating the effects of OA in the region, as it is thought that dissolution of the shell hash will add TA back into the water and elevate pH (e.g., Doyle and Bendell, 2022; Kelly et al., 2011)."

And in line 701 that "There were also no clear indications of pH elevation related to TA increase at the shell midden beach location in Okeover over other beaches or other nearshore sites."

Comment 4: L 159: It might be good to cite which type of CTD as uncertainties estimation depend on it (even if Castaway & RBR CTDs are in given in supplementary of Simpson et al. 2022).

Thank you for pointing this detail out, we used a Castaway CTD and have added this detail in line 162.

Comment 5: L 174: Specify here once for all the used pH scale

This omission was also raised by reviewer 1 – we used the total scale and have added this detail in line 180.

Comment 6: L 176: Precise which hydrogen fluoride dissociation did you use?

The Orr et al. (2018) implementation of CO2Sys we used in this study uses the KF constants reported in Dickson and Riley (1979). We have added this detail in line 180 of the revision.

Comment 7: L 221: Correct $\Delta\Omega$ to $\Delta\Omega_c$

Thank you for pointing out this oversight, we have made this correction.

Comment 8: L 260: is addressed above within the results section revisions.

Comment 9: You could also skip the Ω_c graph as it is similar to Ω_a.

We have combined both the aragonite and calcite results to reduce the length of the results section (as above). As both reviewers have noted, the patterns of variability are similar and so important details have not been lost. We would however like to keep both aragonite and calcite panels in the figures, as although patterns in these saturation states are similar, the timing and extent of undersaturation are different. Calcite undersaturation is not often considered when addressing the impacts of OA on calcifying organisms, and it is interesting that calcite is undersaturated at times in our region. We refer to these figures and compare aragonite and calcite in the discussion and conclusions and would argue they are an important piece to keep.

Comment 10: However, another subplot presented normalized TA vs DIC may be interesting.

Each sub-region in our study has a different mean annual salinity (Table A3), which dictates the location of the normalised TA bars (by region) in the subpanels (Figures 2i, 3i, 4i). If the data from each region were normalised to the same salinity then they would converge to about the same value, within our uncertainty. Normalised DIC shows large variation, mainly because of the strong biological drivers (shown in our Taylor expansion analyses). In short, a normalised TA vs normalised DIC (to a common S) would become a 1-dimensional plot (a horizontal line) in our data. Thus, we do not feel that this plot would add within this study.

On the other hand, we agree with Dr Petton that TA-s vs DIC-s plots could provide an interesting view of sub-regional variation with our study region. We would need to collect significantly more samples and endeavour to reduce our TA uncertainty to be able to tease out potential (relatively small) sub-regional TA differences and identify potential (also relatively small, but not necessarily unimportant) biologically mediated TA-fluxes. Amongst other things we believe that we would need to over-predict the carbonate system and/or directly measure the organic TA component, especially in the more brackish samples (e.g., see work by Brian Hunt, UNH) to increase the accuracy of our carbonate-TA estimation. Our current dataset is dominated by samples in the high (for this region) salinity range (S>~26) because those salinities are typical at the grow sites. We would need significantly more samples at lower salinities, where freshwater end-members appear to diverge. DIC on the other hand does vary by sub-region in our data. (Simpson et al 2022 show TA-S plots (and DIC-S) in Figs 2 and 3 for two of the sub-regions in this study.).

Similarly, sub-regional variation in DIC vs S (but not TA vs S) is seen in the larger, more open waters, of the Salish Sea; specifically, the Juan de Fuca Strait and Strait of Georgia (Ianson et al. 2016, Figure 2).

Comment 11: L 502: Reformulate the sentence "The magnitude of the seasonal …ranging from 0.04 to 0.53". Thank you for pointing out the repetition in this sentence. We have removed the "second ranging from 0.04 to 0.53"

Comment 12: L 838 – 844: This paragraph gives obvious and unnecessary assumptions about numeric model expectations. I would remove it.

We agree that this paragraph is high level and does generalise and have removed it as suggested.

In addition to the edits made to address the above comments, we have also made minor changes to the text to improve clarity and flow.