Combined authors response and changes-tracked revised manuscript of OS_2016_09.

In the following we provide a detailed point-by-point response to all referee comments and specify all changes in the revised manuscript. The response to the Referees includes comments from Referees (in black) and author's response and author's changes in manuscript (in blue). In addition, we provide a marked-up manuscript version showing the changes.

Reviewer 1 (Dr. M. Ribas-Ribas).

The present paper under review for Ocean Science describes seasonal cycles and controlling factors on Aragonite saturation states and pH and proposes a nice way to monitore Ocean Acidification from continuous underway pCO2 measurements. I appreciate the effort to maintain the VOS line for so long and the extra sampling cruise. Together, makes a valuable dataset! It is also really easy to read as it flows. I will recommend the publication after moderate corrections/clarifications.

Response: We thank the reviewer for encouraging and constructive comments as well as for suggested corrections and improvements.

However, I have two major concerns about the current version of the manuscript: 1. Statistical analysis need to be better explained and improved to have more robust conclusion. For example, fig. 2 B (8-249, 9-264) explain that they are UW and RF are really similar but I would like to know the slope and if this slope is statistically different than 1 (same apply for B and D). I would add other stats comments and the list of general comments.

Response: We agree with the reviewer on this point.

In the revised manuscript, the statistics of all comparisons are presented. An additional table (Table 3) summarizing the statistics of the comparisons on Fig. 2 is included. Further, in order to introduce how these statistics is used the following paragraph has been added to the beginning of section 3.1 "In this section we present the regression equations identified in this study in addition to validating the various estimation procedures used by comparing the estimated values with those measured/computed. The results of these comparisons are summarized in Table 3. For each comparison, the coefficient of determination (R^2) and the significance level (p-value) are used as metrics for the goodness of the correlation while the associated root-mean-square error (rmse) is benchmarked against the maximum target uncertainties developed by the Global Ocean Acidification Network (GOA-ON) and the California Current Acidification Network (C-CAN) of ± 0.2 for Ω_{Ar} (McLaughlin et al., 2015), which corresponds to maximum uncertainties of ± 0.02 , ± 1.25 or ± 1.8 in pH, SST, or SSS, respectively."

2. I would like to know more about interannual variability. This is also somehow what I expect after reading introduction (3-91). But still we only have some sentences at the end of section 3.3. Related to this, I don't understand why you choose VOS data since 2009 but then you have CS since 2010, a cruise in 2015 and a sensor in 2012. I understand it's not easy to treat data from VOS as quick as they are generated but at least some words of cautions will be nice.

Response: we agree with the reviewer to write more about the interannual variations (IAV), although the focus of the study is on the seasonal variations.

In the revised manuscript, IAV have been computed as the standard deviations of monthly means. The results are summarized in additional table (Table 4) and results commented in the end of section 3.3.

Regarding the temporal coverage of the datasets, we used all underway data that has been available from the VOS line which was in operation in the period September 2005 to September 2009. Cruise data with dates beyond the operating period of the VOS line has been used in order to maximize the data (in terms of number of data points as well as in spatial and temporal coverage) available from the study area to identify the regional SSS/SST – Alkalinity relationship (Eq. 2 in the manuscript). In order to clarify the above points, the first paragraph of section 2.1 in the revised ms, includes the sentence "*The VOS line was in operation in the period September 2005 to September 2009*."

My comments will be with format page-line (so 5-3 means page 5, line 3); page-line:line (so 7-7:9 means page 7 from line 7 until line 9).

1-17 What coherent means in this contest?

Response: with "Spatially Coherent" we mean that seasonal changes with similar magnitudes occur everywhere at the same time.

1-21 (and elsewhere in the ms) Please choose as many decimal place you want to report according to your method accuracy and error propagation and stick to them. Here you start with 2 and then past to 1 but further in the ms, suddenly there is 4-5 digits (9-272, 10-284). Response: in the revised ms, pH values are presented with three decimal points while two decimals are used for the saturation state.

2-36 What is Peters reference referring to? It looks strange.

Response: this has been replaced with more relevant reference (Le Quéré et al., 2015) for updated assessment of global emissions.

2-51-54 Long paragraph

2-61 "Only few studies" I expect at the end of the sentence some citation

Response: in the revised ms (page 3), this sentence has been extended to "*However*, only a few studies on the carbon cycle of Norwegian fjords exist in the literature, and these are from the high Arctic at Svalbard (Fransson et al., 2014; Omar et al., 2005)."

3-75:80 I think that should flow better with a bit of rearranging to avoid repetition together with 1-61:2-68

I really like this part where it is explain the importance of the study, to "fill the gap". It is indeed extremely important to fill these coastal gaps. I think it will also be nice to mention that this study set the baseline for future studies to evaluate OA and how this change could affect.

Response: we thank the reviewer for a good suggestion.

In the revised ms (page 3), we have rearranged the first sentence as suggested. We have also extended the paragraph with "Observations of the carbon cycle dynamics in the fjord system will not only further our understanding and ability for prediction, but they will also serve as benchmarks against which future changes are compared."

5-130 "coastal open ocean" is somehow misleading, although I understand might be good to rephrase.

Response: in the revised ms, the phrase "coastal open ocean" has been replaced with "adjacent coastal North Sea".

5-143:146 Economic important could be also tourist and other ecosystem functions? Response: we tried to stick to the natural ecosystems only. However, we agree that we needed to elaborate the economic significance. Thus, in the revised ms (end of section 1.1), we extended the paragraph with "*The fjord system also contributes to the important aquaculture production that, with its annual fish production of* >700 *tonnes, ranks Norway within the tenth place worldwide. About one fifth of this is produced in the Hordaland County where the fjord system studied here is situated (http://www.diercke.com).*"

5-149 "of" is double CO₂ is without subscript (also in equation 3 and 4 and 12-360 and some references)

Response: corrected.

5-151 webpage without hyperlink, like for example 6-166 (some happens in 7-197) Response: corrected.

6-166 knowing the authors, I suppose there are plan to submit the remaining data to SOCAT, right?

Response: the reviewer is right. Actually, the data is submitted as mentioned in revised ms (last sentence of section 2.1).

6-176 (and elsewhere in the ms, for example table 1 or 7-195), unify dates format Response: done.

7-197 It will be nice to state here a few details from sensor, like accuracy or/and precision, as it is important to propagate the other uncertainties in the calculations. Response: done.

7-217 Here an important point, TA-SSS algorithm comes only from summer values, right? Please clarify and add some sentence of caveat about it.

Response: we agree with the reviewer.

In the revised ms, we have discussed this issue in connection with the comparison between wintertime pH estimates and in situ pH measurements from January 2012. In the end of section 3.1 we conclude "It must be noted that the above total error was derived from all available observational data including the in situ sensor data (shown in Fig. 2c and in described section 2.3), which are the only wintertime measurements used in this study. This is important because the lack of wintertime data in the CS dataset which was used for the identification of AT-SSS/SST relationship (Eq. 2) means that wintertime AT(sst) might be overestimated so that corresponding pH(sst) values would be overestimated. In fact, during the aforementioned comparison between pH(sst) and measured pH we noted that for this particular dataset pH(sst) overestimated the measurements. However, the estimates were consistent with the observations to within the total error of ± 0.01 pH units. Thus, by utilizing the above total errors, we also accounted for the effect of this possible caveat of Eq. 2 arising from the lack of wintertime TA measurements."

8-222 Which other ancillary parameters? Where they come from?

Response: in the revised ms (page 8), this sentence has been modified to "*The UW fCO2* together with TA (sst), UW SST, and SSS(sst) were then used to characterize the full seawater CO2 chemistry using CO2SYS (Lewis and Wallace, 1998; van Heuven et al., 2011), with K1 and K2 constants from Lueker et al. (2000)."

8-244 You haven't describe RMS before. Salinity is undimension. It will be nice to have some idea here of what this 0.81 could means in terms of pH or OmegaAr, with initial error propagation

Response: in the revised ms, statistics of equations and comparisons clarified, and rmse is defined. Additionally, the errors in pH, SST, and SSS that can produce the maximum target uncertainty of ± 0.2 for Ω_{Ar} developed by the C-CAN and GOA-ON have been estimated (see 1st paragraph of section 3.1).

10-281 Fig. 2 should read Fig. 3. Also normally better to start the sentence in more direct way. I would like to know more details how is this data collapsed: what we see is means? What about std? This comment is together with this interannual variation maybe.

Response: what we show on Fig. 3 is gridded data based on weighted averages. In the revised ms (section 3.2, and tables), a table showing the interannual variations has been added (Table 3). Additionally, the first paragraph of section 3.2 is modified to "In order to present the mean distributions across the different fjords throughout the annual cycle, we collapsed the data into one virtual year by projecting it onto non-equidistant rectangular grids using the "weighted-average gridding" method of the Ocean Data View software (Schlitzer, 2015). As evident from Fig. 3, there is a clear seasonality in both pH(sst) and Ωar (sst)."

10-284 text said maximum is 8.2 but figure goes until 8.35

Response: winter pH values are typically about 8.25, but extreme pH values of >8.3 occurred during the winter of 2008.

In the revised ms (section 3.2, and tables), this is clarified by stating that pH "...varies between minimum values (8.05) around New Year to typical maximum values of around 8.25, which occur during the late winter and/or spring (March-April)." Additionally, a table showing the interannual variations has been added (Table 3).

I like this comparison with C-CAN \blacksquare

10-311 Could you add a sentence in methodology or somewhere explaining how DIC is normalize, which salinity has been used, which reference? Also note OmegaAr (sst) has something between t and). Also extra space before brakets ini 11-313 Response: corrected.

11-314 State the table or fig that is showing that (I think Fig. 4) Response: done as suggested by the reviewer.

11-315 Maybe nice to state the basics about Lauvset methods. Also note extra brakets before Lauvset and missing before the year.

Response: done as suggested by the reviewer.

In the revised ms (page 12), we added the sentence "*This method estimates the monthly pH changes expected from corresponding changes observed in SST, SSS, DIC, and TA as well as their sum.*"

11-316 Left panels maybe better for state the letters (a, c...) Response: done as suggested by the reviewer. 11-319 Fig. 4 c, d, e (mismatch of figure number, spaces missing after comma and letters in the figure are capital and in the test are not, please unify). Also amend for 11-321,11-333, 12-369 and 11-322.

Response: corrected as suggested by the reviewer.

11-324:325 This sentence need more explanation. Although I agree, somehow contradict the idea of if we measure pCO_2 and temperature we can estimate OA parameters.

Response: The sentence is not about our ability to estimate OA parameters. It is about our ability to analyze the controls of OA parameters for which measured SSS and AT are best. If estimated AT(sst) is used, on the other hand, the magnitude of the alkalinity control will be more uncertain while its form will resemble that of SST.

In the revised ms (page 13), the sentence is extended to "*This emphasizes the need for* measured SSS and TA values when the objective is to analyze the controls of pH and $\Omega_{Ar}(sst)$ variations."

11-334 I think that should read Fig. 4F

After reading this twice, an idea can be to do some "Multiple linear regression" to support your analysis in a statistical way.

Response: here we disagree with the reviewer. First, "Fig. 4" is correct i.e. that is where the seasonal amplitude of $\Omega_{Ar}(sst)$ is shown. Secondly, we have already verified that multiple linear regressions are not a better alternative for our analyses. Such regression would only give us indication on how well the $\Omega_{Ar}(sss)$ values can be modelled from a given set of parameters, but the values of the regression coefficients would NOT necessarily reflect sensitivity of $\Omega_{Ar}(sss)$ to the associated parameters.

11-337 impact. (extra space before point) Also in 11-342 before sst).

Response: corrected as suggested by the reviewer.

12-359 equations 5 and 6 or equation 3 and 4 (somehow not easy to understand) Response: we agree with the reviewer. The ambiguity arised as result of a typo. In the revised ms (page 14), the sentence is corrected to "*This, together with the fact that equations 3 and 4 can...*"

12-366 another stats comment, how significantly is this significantly?

12-371 there are two commas before R2

13-372 there is extra point after (6)

Response: Thank you! Both the above typos have been corrected in the revised text.

13-373 or somewhere else, just though that another importance to use VOS pCO₂ is that they are more frequent that normal oceanography cruises in winter, especially in these regions. Response: Good idea! This is implemented in the revised manuscript (page 15).

A note of caution about the slopes, we need some +- because if we took the slope of red square will be really different. But still I like the comparison with RCP scenarios. Response: we agree with the reviewer.

In the revised ms (end of section 3.4), an uncertainty of ± 0.2 in the Ω_{Ar} estimates is included and the text has been changed accordingly.

13-387 cannot

Response: we are not sure what this comment means, but the word "cannot" was used intentionally in the original manuscript.

13-401 extra : at the end of captions Response: corrected.

14-420:423 I miss some words of what this low omegaAr 1.3 could mean for calcifying organism. Some studies point that there's no need to be below 1 to have some affect in organism. Some "bio"-words could expand the readers-users of these nice results. Response:

14-424:425 Maybe I miss something but this sentence is not really new or surprising, right? Response: in the revised text (page 17), this sentence has been modified to "Strong correlations of pH and Ω_{Ar} with fCO_2 and $fCO_{2@meanSST}$ (fCO_2 adjusted to the mean temperature), respectively provide an approach to interpolate pH and Ω_{Ar} over large areas in the fjords of western Norway where underway measurements of fCO_2 , SST, and SSS are available."

Table 1 What's the different between 1 and 5 m. Are they both underway water? Response: The RF dataset are averages over measurements five depths 1-5 m. This is clarified in the revised manuscript (page 7, and Table 2 and its caption) 22-607 Should be table 2 Response: corrected.

Figure 1 may benefit from a more global map, for no European readers. Also if possible add the meaning of the arrows (NCC) in the figure, so we don't have to read legend. Response: a more global map is used in the revised manuscript.

Fig. 2A, is colour scale and x-axis the same SST Response: No, the color scale shows months. This is clarified in the revised manuscript (Fig. 2A).

Fig. 3 is missing A-D and parameter and unit on the colour scale. Also unify decimal units Response: The missing legends "A-D" has been added to Fig. 3 of the revised manuscript.

Fig. 4 You explain good that SST and TA are not independent. So question is, could you still use this method with dependent variables? Also careful because labels are cutting x-axis. Response: Yes, we can use this procedure even if TA is based on SST data. This is because the decomposition procedure computes the effect of changes in each driver (i.e. changes in SST, SSS, TA, and DIC) separately. Moreover, the seasonal changes in TA and SST are opposite i.e. TA decreases when SST increases and vice versa, and the magnitude of the former is about ten times bigger. Thus, even though the similarity of Fig. C and E (in from) is as expected, the magnitudes of the resulting effects from the two drivers can be different (these are currently nearly equal by coincidence) if, for instance, we use a TA Revelle Factor slightly different form the global value of -10.

Dr. Reviewer 2 (E.M. Jones)

Comment:

"This study presents seasonal cycling and controlling factors of the ocean acidification parameters, aragonite saturation and pH, in fjords of western Norway. The data and discussions are an important contribution to CO2 research in a relatively undersampled region and the methods used to determine the whole carbonate system from underway pCO2 data form a novel approach and are complimentary to future studies in this field. The choice of the journal fits very well and I recommend publication of this article. In the version reviewed here there are a number of ameliorations that can be made in terms of clarity, coherence and rigour before proceeding with publication. The authors may find comments, questions and linguistic corrections analytically below."

Response: We thank the reviewer for these encouraging and constructive comments as well as for the suggested corrections and clarifications.

C: 1. A very minor comment regarding language; as it is a European journal I would suggest replacing the use of "fall" with "autumn". Response: done as requested by the reviewer.

2. Make sure it is clear how TA is derived from SSS(sst) and SST, i.e, how strong is a relationship between SSS and SST to make such a derivation and secondly what steps are carried out to determine TA?

Response: this has been clarified as suggested by the reviewer.

In the revised manuscript (page 10), it is stated "To estimate a corresponding TA value for each UW fCO2 observation obtained from MS Trans Carrier, we used salinity values estimated from UW SST data by using Eq.1. The results (denoted as SSS(sst)) were then inputted into Eq.2 to obtain TA(sst) (Table 2). The fact that TA(sst) are based on SSS(sst) rather than measured SSS values introduced an additional error in the estimated pH(sst) and $\Omega Ar(sst)$. In order to assess this error...."

3. Include a clear statement of the precision and accuracy of the measured parameters. This would fit well before the detailed description of the propagation of errors and further place them in context.

Response: in the revised ms, the measurements accuracy of DIC, TA, and pH is stated (section 2.2 and 2.3).

4. The influence of sea ice and glacial ice dynamics has not been explicitly considered. Is this area free from ice influences and if so perhaps state this or does sea ice formation in winter and melt during spring-summer contribute to the correlation in SST and SSS?

Response: the study area is free from ice influence.

In the revised ms (section 1.1), this is clarified by stating "...snow melt and run-off are the main local sources for freshwater since the fjord system is generally ice free."

5. For salinity normalised DIC, the terms DICS and nDIC are both used. Perhaps try and use one term for consistency and clarity throughout the paper and include a statement of how the normalisation was carried out, i.e., what salinity reference is used, if the standard correction was made or non-zero endmembers were included, as described in Friis et al. (2003).

Response: "DICS" was found in Eqs. 3 and 4 in the original the manuscript. However, this is referring to the product "DIC" times the parameter "S". The latter has been defined in Egleston et al. (2010) as referenced in the text.

In the revised manuscript, all abbreviations used DIC, nDIC are properly defined. Additionally, an overview of the symbols used for quantities estimated and/or derived from the measurement-based variables is given on Table 2. 6. Figure 2: in relation to the various regression analyses made, a statement on the interpretation of the regression and the significance of the relationships is required. This would make the comparisons of the parameters more relevant and strengthen the choice of those used in other calculations.

Response: we thank the reviewer for the suggested improvement.

In the revised manuscript, the statistics of all equations and comparisons are presented, and an additional table (Table 3) summarizing the statistics of the comparisons on Fig. 2 is included.

7. Check Table numbering; Table 2 is absent.

Response: corrected.

Specific comments:

Abstract

Page 1

Line 9 write "carbon dioxide (CO2)" in full (first time use in main text). Line 10 insert "the" between "lowers" and "aragonite saturation state". Line 19 change to "carbonrich". Line 24-25 replace "brings up" with "enables" and add "reach" between "to" and "the". Line 32 define "SSS" for first time use.

1. Introduction

Page 2

Line 38 replace "incur" with "cause". Line 39 replace "ocean" with "oceanic". Line 43 insert "," after the reference. Line 51 insert "," after "regions". Line 63 insert "a" between "is" and "prerequisite".

Page 3

Line 64 remove the "s" on "latitudes". Line 65 insert "CO2" between "be" and "sources". Line 76 delete "important" to refrain from using the same word twice in one sentence. Line 81 delete "the" between "in" and "surface". Line 89 insert "the" between "present" and "mean".

1.1 The Study Area

Page 3

Line 93 instead of capital letters use lower case "n" and "s" for "north" and "south", respectively. Line 95 replace "at" with "along".

Page 4

Line 103 delete "the" at the end of the line. Line 106 replace "it" with "Korsfjord" to reduce the use of generic "it" and be specific to the subject under description. Line 108 remove full-stop and replace with and to join sentences for better flow. Line 109 correct "witch" to "which". Line 118 replace "mix" with "mixes". Line 119 insert second bracket ")" after "(NCC". Line 121 change "wind" to "winds" and insert "the water in" after "circulation of". Line 122 replace "that follows" with "following". Line 124 suggestion to add "forcing" after "wind". Line 125 replace "," with "and inputs of". Line 125-126 between "seasonal time scales," and "during spring-summer" replace text with "salinity drives stratification". Line 126 add "the water column is" between "and" and "more". Page 5

Line 129 specify what "the temperature" refers to, i.e., a certain depth or depth range

in the water column or certain location in the fjord. Line 131 delete "the" start line with "Water". Line 132 following "oxygen" insert "to the area". Line 132-133 delete "the fjords enhance their" and insert "is enhanced in the fojrd" between "production" and "which". Line 133 replace "enables them to host" with "supports". Line 137 replace "decisive" with "dominant controls of" and delete "for the". Line 140 write "nutrient-rich" and add "on from" after "follows". Line 142 insert "upper" before "water column", i.e., assuming that the sub-surface/ deeper waters remain nutrient rich? Line 143 insert 2 "," either side of "with its adjacent waters". Line 152 replace "It" with "The ship". Line 153 replace "." after "(Fig. 1)" with "," and delete "It".

Line 6

Line 161 replace "in" with "during". Line 166 add SOCAT reference: Bakker, D. C. E., Response: all the above correction have been implemented in the revised text.

Page 7

Line 192 instead of capital letter use lower case "s" for "station". Line 200 clarify the estimated pH values – estimated from what source? CO2SYS?

Response: this is clarified in the revised ms, by modifying the sentence to "*These sensor data* were used to assess the uncertainty in our pH values estimated as described in the next section."

2.4 Methods

Page 7

Line 206-207 insert "an" before "empirical" and define the parameters used to derive the empirical relationship.

Response: this is clarified in the revised ms, by modifying the end of the sentence to "...sea surface salinity (SSS) was determined from empirical relationship <u>with SST</u>."

Line 217 replace "Second" with "Secondly". Line 217 clarify how TA is derived from SSS(sst), see general comments. Response: see response to the general comments.

3. Results and discussion
3.1 Correlations and validations
Page 8
Line 236 insert "the" before "data".
Line 237-239 Is sea ice present in the region? see general comments.
Response: see response to the general comments.

Line 274 insert "through the calculations" between "propagated" and "to".

Response: in the revised text (page 11), the sentence have been modified to "*These two error* estimates were combined (as the square root of sum of squares) to determine the total error in our estimates, which were found to be ± 0.01 and ± 0.1 for pH and ΩAr , respectively."

Line 239 clarify where the "runoff" originates. Line 240 insert

"degree of" between "high" and "scatter". Line 244 reverse order of "indeed is" to "is indeed". Line 247 delete "a" after "As" and re-write sentence following "verification" as "that the RF SST dataset is spatially representative,". Line 249 replace "in" with "across". Line 256 replace "statics" with "statistics". Line 257 replace "functions" with "acts" and insert "an" between "as" and "indicator". Line 259 add a space to separate "with" and "values". Line 260 add commas to read "data ,i.e., pH(sss)". Line 265 insert "closely" between "

Ar(sst)" and "reproduce" and delete "very well". Line 269 replace "show" with "shown".

3.2 Spatiotemporal variations

Page 10

Line 281 replace "collapsed" with "condensed". Line 284 delete "the" following "after". Line 286 replace "outweighs" with "counteracts". Line 287 replace "begins" with "begin" and also specify what processes are being referred to. Line 288 insert "the" after September replace "SSTs" with "SST". Line 289 re-phrase sentence between "mixing," and "as mentioned" to read "which enables deep, carbon-rich coastal water to penetrate the surface layer,". Line 290 replace "as" between "and" and "reflected" with "is" and delete "the" before "increasing". Perhaps specify which DIC values are increasing by the autumnal mixing, i.e., sea surface, surface layer, upper ocean: : : Line 293 replace "drives up the " with "enriches". Line 294 insert "the" before "concentration", delete "the" between "of" and "carbonate" and add an s to read "ions". Line 296 rephrase to read "due to inputs of run-off" and replace "reinforces" with "enhances". Line 297 replace "over" with "exhausted".

Response: all the above corrections have been implemented in the revised text.

Line 298 replace "The" with "However," and delete

", on the other hand,".

Response: we find this sentence clear in the original text.

Line 300 replace "mismatch" with "decoupling of". Line 307 correct "embody" to "embodied".

3.3 Controls of seasonal variability and trends

Page 11

Line 310 replace "arranged the data into" with "computed". Page 11 Line 315 re-write the reference to "Lauvset et al. (2015)". Line 316-317 replace "are shown on Fig. 4 (left panels) where it can be seen" with "show" – removes the double Fig. reference in the sentence. Line 326 replace "letting one of the drivers" with "varying them independently".

Line 327 delete "to vary" and replace "drivers" with "controls" – removing double word use in the same sentence. Line 329 replace "for" with "of" and correct "driver" to "drivers". Line 331 correct "induce" to "induces". Line 332 insert "other" between "the" and "hand". Line 333 correct "induce" to "induces". Line 334-335 re-write to "We therefore conclude that variations in DIC, followed by TA, are the most important drivers for changes in

Ar(sst)." Line 337 replace "have" with "has" and delete "In

terms of processes" so that the next line starts with "This means". Line 338 correct to "carbon-rich". Line 339 insert "inputs" after "runoff". Line 343 replace "changes" to "change".

Response: all the above corrections have been implemented in the revised text.

The first use of nDIC – how is defined and how are the normalised values determined, perhaps add reference, e.g., Friis et al. (2003).

Response: in the revised text (page 13), this part of the sentence has been modified to "...both for SST and DIC normalized to the mean salinity (nDIC) according to Friis et al. (2003)"

3.4 Inference of OA parameters from VOS underway data
Page 12
Line 353 the use of DICS is not specifically defined in the text and is an additional
term to nDIC – do they both refer to salinity normalised DIC determined in the same
way? Please clarify to be sure.

Response: please see the general comments regarding "DICS".

Line 362 remove ", while the CO2 system is fully determined only occasionally," as I don't think it adds anything it would reduce the length of the sentence.

Response: we chose to keep "while the CO_2 system is fully determined only occasionally" in the text in order to emphasize that occasional determination of the full CO2-system is necessary for the method to work.

Line 355 remove the space after (2010). Line 359 reference to equations 4 and 5 might not be correct, i.e., should it be equations 3 and 4. Please check and amend if necessary.

Line 368 delete the spare "and" after "Fig. 5" and add "to" between "conform" and "tight". Line 371 replace extra "," with ";" after "10.354" in equation 5.

Page 13

Line 372 check for extra spaces and uses of "," and/ or ";" in separation of terms and consistency with equation 5. Line 377 delete "and" before "thus" and replace "min- imise" with "minimizing". Line 380 replace "with" by "to". Line 391 change to "carbonrich". Line 392 rearrange the line to read "surface water also reflects the properties of the deeper water". Line 394 replace "can" with "could" and insert "to occur" after "expected". Line 399 replace "development" with "CO2 concentrations" and replace "follows" with "follow". Line 400 add "driving oceanic CO2 uptake" after "atmosphere". 4. Summary and concluding remarks

Page 14

Line 402 insert "worth" after "four years" and replace "less" with "sporadic". Line 403 delete "frequent". Line 413-414 use the same number of decimal places for quoted

numbers. Line 415 insert "the" before "phytoplankton" and replace ", which" with "that" and delete "levels". Line 417 replace "brings up" with "allows"; correct to "carbonrich"; insert "mix into" between "to" and "the". Line 418-419 replace "and reinforces the decrease in pH" with "reduces" and delete ", which continuous throughout fall". Line 430-431 finish sentence after "carbonate system".

Page 15

Line 435 replace "development" with "concentrations" and remove "f" from "fCO2", i.e., referring to concentrations of CO2 rather than the concentrations of the fugacity of CO2 that wouldn't be quite correct. Line 436 finish the sentence after "atmosphere".

5. Acknowledgements

Page 15

Line 438 replace "supports" with "support" and replace "by" with "from". Line 440 insert "the" after "and help of".

Tables

Table 1

Page 20 Line 598 replace "about" with "of". Line 599 insert "which" after "for".

Page 21 Line 603 replace "(continue)" with "(continued)".

Table 3

Page 22 nDIC – "DIC normalised to the mean salinity"; please include somewhere in the main body of the text what value was determined for the mean salinity and how. Figure texts

Page 23 Line 620 replace "which" with "where" and replace "tick" with "thick". Line 635 check "ln(nfCO2ts)" – is the "n" correct and currently it doesn't match with the X-axis labelling in Figure 5 (B).

Figures

Figure 2 (A) Y-axis can be expanded to open up the visual regression trend, i.e., range in SSS of 25-35. Confusion of the colour bar underneath is too close to the X-axis and on first inspection looks like it is referring to SST values. Add "month" as the unit to clarify the colour bar and move it closer to panel (A) if possible. Figure (3) 4 time series panels are presented but only 2 parameters mentioned in the Figure text and all colour bars are unlabelled. Add parameter labels and units (where relevant) to each of the coloured panels.

Response: all the above corrections have been implemented in the revised text.

1	Aragonite saturation states and pH in western Norway fjords:	Formatted: Font color: Text 1
3	seasonal cycles and controlling factors, 2003-2009	
4	A. M. Omar ^{1,2} , I. Skjelvan ¹ , S.R. Erga ³ , A. Olsen ²	
5	^{1:} Uni Research Climate, Bjerknes Centre for Climate Research, Bergen, Norway.	
6	^{2:} Geophysical Institute, University of Bergen, and Bjerknes Centre for Climate Research, Bergen, Norway.	
7	^{3:} Department of Biology, University of Bergen, Bergen, Norway.	
8	Abstract:	
9	The uptake of anthropogenic Carbon Dioxide (CO ₂) by the ocean leads to a process known as	Formatted: Font color: Text 1
10	ocean acidification (OA) which lowers the aragonite saturation state (Ω_{Ar}) and pH, and this is	Formatted: Font color: Text 1
11	poorly documented in coastal environments including fjords due to lack of appropriate	Formatted: Font color: Text 1
12	observations.	
13	Here we use weekly underway data from Voluntary Observing Ships (VOS) covering the	
14	period 2005-2009 combined with data from research cruises to estimate Ω_{Ar} and pH values in	
15	several adjacent western Norwegian fjords, and to evaluate how seawater CO_2 chemistry	
16	drives their variations in response to physical and biological factors.	
17	The OA parameters in the surface waters of the fjords are characterized by strong seasonal	
18	and spatially coherent variations. These changes are governed by the seasonal changes in	
19	temperature, salinity, formation and decay of organic matter, and vertical mixing with deeper,	
20	carbon-rich coastal water. Annual mean pH and Ω_{Ar} values were 8.13 and 2.21, respectively.	Formatted: Font color: Text 1
21	The former varies from minimum values (\approx 8.05) in late December - early January to	
22	maximum values of around 8.2 during early spring (March-April) as a consequence of the	
23	phytoplankton spring bloom, which reduces Dissolved Inorganic Carbon (DIC). In the	
24	following months, pH decreases in response to warming. This thermodynamic decrease in pH	
25	is reinforced by the deepening of the mixed layer, which brings upenables carbon-rich coastal	Formatted: Font color: Text 1
26	water to reach the surface, and this trend continues until the low winter values are reached	Formatted: Font color: Text 1
27	again. Ω_{Ar} , on the other hand, reaches its seasonal maximum (>2.5) in mid to late summer	Formatted: Font color: Text 1
28	(July -Sept), when the spring bloom is over and pH is decreasing. The lowest Ω_{Ar} values	
29	(\approx 1.3-1.6) occur during winter (Jan-Mar), when both pH and Sea Surface Temperature (SST)	
30	are low and DIC is highest. Consequently, seasonal Ω_{Ar} variations align with those of SST	
31	and salinity normalized DIC _{τ} (nDIC).	Formatted: Font color: Text 1

We demonstrate that underway measurements of fugacity of CO₂ in seawater (fCO₂), SST;)
and <u>SSSSST</u> from VOS lines combined with high frequency observations of the complete
carbonate system at strategically placed fixed stations provide an approach to interpolate OA
parameters over large areas in the fjords of western Norway.

36 **1. Introduction:**

- 37 The continued emissions of carbon dioxide (CO₂) (PetersLe Quéré et al., 20132015) are of 38 global concern, not only because they are the main drivers of anthropogenic global warming, 39 but also because of the changes in the ocean chemistry they incurcause (Ciais et al., 2013). The increase in the atmospheric CO₂ concentration drives a net ocean CO₂ uptake, which 40 41 leads to higher proton (H⁺) concentration i.e. lower pH, lower concentration of carbonate ion $(CO_3^{2-})(CO_3^{2-})$ and, lower saturation state (Ω) for calcium carbonate in seawater. This process 42 is known as ocean acidification (OA) (e.g. Royal Society, 2005), and it has direct and indirect 43 44 effects on biological activity in the ocean (e.g. Gattuso and Hansson, 2011) including reported inhibition of biogenic calcification by marine organisms which precipitate 0.5-2.0 Gt of 45 carbon as calcium carbonate (CaCO₃) in the global ocean every year (Bach, 2015). 46
- For the open ocean, the rate of OA has been relatively well documented and understood during the last decade. Observations from time series stations and volunteer observing ships in different oceanic regions consistently show systematic changes in surface ocean chemistry that result from OA. Specifically, long-term negative trends of pH and saturation state for aragonite (Ω_{Ar}) have been observed (e.g. Lauvset et al., 2015; Bates et al., 2014).
- For coastal regions observed rates of pH change largely differ from those expected from
 oceanic CO₂ uptake alone, as variations in other biogeochemical processes, related for
 example to changes in nutrient loading and eutrophication, are important as well (Clargo et al.
 2015; Provoost et al., 2010; Wootton et al., 2008).
- The Norwegian west coast (Fig. 1) is dominated by fjords, narrow and deep estuaries, carved
 by glacial processes, with a sill in the mouth where they connect to the coastal North Sea.
 Apart from being important recreation areas and marine pathways, these fjords are important
 ecosystems and their physics, marine life, and associated environmental pressures have been
 relatively well studied (e.g. Matthews and Sands, 1973; Erga and Heimdal, 1984; Asplin et
 al., 2013; Brattegard et al., 2011; Stigebrandt, 2012; and references therein).

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62	However, only a few studies on the carbon cycle of Norwegian fjords exist in the literature $\frac{1}{2}$	
63	and these are from the high Arctic at Svalbard (Fransson et al., 2014; Omar et al., 2005), This	Formatted: Font color: Text 1
64	hampers our understanding of the natural variability and controls of seawater carbonate	
65	chemistry, which is <u>a prerequisite for sound estimates of OA in these ecosystems. Generally,</u>	Formatted: Font color: Text 1
66	in the Northern Hemisphere, high latitudeslatitude coastal regions are thought to be sinks for	Formatted: Font color: Text 1
67	atmospheric CO ₂ , while low-latitude regions are thought to be $\underline{CO_2}$ sources (Borges et al.,	Formatted: Font color: Text 1
68	2005; Cai et al., 2006; Chavez et al., 2007; Chen and Borges, 2009). The few existing studies	
69	of Norwegian fjords confirm the above picture; they act as an annual net sink for atmospheric	
70	CO ₂ (Fransson et al., 2014; Omar et al., 2005).	
74		
/1	The carbon cycle of the northern North Sea, to which the western Norway Ijords are	
72	connected, has been well studied (Thomas et al., 2004; 2005; 2007; 2008; Bozec et al., 2005;	
73	2006; Omar et al., 2010). However, observation-based OA estimates are still scarce. Recently,	
74	Clargo et al. (2015) observed a rapid pH decrease in the North Sea, but after accounting for	
75	biological processes, they estimated an ocean acidification rate consistent with concurrent	
76	atmospheric and open ocean CO_2 increases over the period they studied, 2001-2011.	
77	Filling the knowledge gap on OA (and generally the carbon cycle) in fiords in western	Formatted: Foot color: Text 1
	<u>I ming the knowledge gap on or 1</u> (and generally the carbon eyere) in Ijords in western	
70	Norway has not been described previously. Filling this knowledge gap Norwagian fiords is	Formatted: Font color: Text 1
78	Norway has not been described previously. Filling this knowledge gap Norwegian fjords is	Formatted: Font color: Text 1 Formatted: Font color: Text 1
78 79	Norway has not been described previously. Filling this knowledge gap Norwegian fjords is important because the fjordsthese areas are important spawning grounds for different fish	Formatted: Font color: Text 1
78 79 80	Norway has not been described previously. Filling this knowledge gap Norwegian fjords is important because the fjordsthese areas are important spawning grounds for different fish species (Salvanes and Noreide, 1993; Johannessen et al., 2014), productions sites for pelagic	Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1
78 79 80 81	Norway has not been described previously. Filling this knowledge gap Norwegian fjords is important because the fjordsthese areas are important spawning grounds for different fish species (Salvanes and Noreide, 1993; Johannessen et al., 2014), productions sites for pelagic calcifiers (Berge, 1962; Erga and Heimdal, 1984; Frette et al., 2004), the home for some coral	Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1
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78 79 80 81 82 83	Norway has not been described previously. Filling this knowledge gap Norwegian fjords is important because the fjordsthese areas are important spawning grounds for different fish species (Salvanes and Noreide, 1993; Johannessen et al., 2014), productions sites for pelagic calcifiers (Berge, 1962; Erga and Heimdal, 1984; Frette et al., 2004), the home for some coral reefs (e. g. Fosså et al., 2002), and significant food sources due to the aquaculture industry which operates there. Observations of the carbon cycle dynamics in the fjord system will not	Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1
78 79 80 81 82 83 84	Norway has not been described previously. Filling this knowledge gap Norwegian fjords is important because the fjordsthese areas are important spawning grounds for different fish species (Salvanes and Noreide, 1993; Johannessen et al., 2014), productions sites for pelagic calcifiers (Berge, 1962; Erga and Heimdal, 1984; Frette et al., 2004), the home for some coral reefs (e. g. Fosså et al., 2002), and significant food sources due to the aquaculture industry which operates there. Observations of the carbon cycle dynamics in the fjord system will not only further our understanding and ability for prediction, but they will also serve as	Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1
78 79 80 81 82 83 84 85	Norway has not been described previously. Filling this knowledge gap Norwegian fjords is important because the fjordsthese areas are important spawning grounds for different fish species (Salvanes and Noreide, 1993; Johannessen et al., 2014), productions sites for pelagic calcifiers (Berge, 1962; Erga and Heimdal, 1984; Frette et al., 2004), the home for some coral reefs (e. g. Fosså et al., 2002), and significant food sources due to the aquaculture industry which operates there. Observations of the carbon cycle dynamics in the fjord system will not only further our understanding and ability for prediction, but they will also serve as benchmarks against which future changes are compared.	Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1 Formatted: Font color: Text 1
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78 79 80 81 82 83 84 85 86 87 88 89 90 91 91 92 93	Norway has not been described previously. Filling this knowledge gap Norwegian fjords is important because the fjordsthese areas are important spawning grounds for different fish species (Salvanes and Noreide, 1993; Johannessen et al., 2014), productions sites for pelagic calcifiers (Berge, 1962; Erga and Heimdal, 1984; Frette et al., 2004), the home for some coral reefs (e. g. Fosså et al., 2002), and significant food sources due to the aquaculture industry which operates there. Observations of the carbon cycle dynamics in the fjord system will not only further our understanding and ability for prediction, but they will also serve as benchmarks against which future changes are compared. In this study, we present first estimates of OA parameters in the surface waters of several adjacent western Norway fjords (Fig. 1), based mainly on weekly underway data from Voluntary Observing Ships (VOS) covering the period 2005-2009. We combine the underway data with available station data from research cruises to facilitate a complete description of the seawater CO ₂ chemistry in accordance with the recommendations of OA core principles by McLaughlin et al. (2015). We focus on analyses of Ω_{Ar} and pH values and evaluate their variations in response to the physical and biological factors: summer warming and stratification, spring phytoplankton bloom, and deep mixing during fall and winter. First we	Formatted: Font color: Text 1 Formatted: Font color: Text 1

95	Hardangerfjord) to understand the spatiotemporal patterns, then we collapse all data into a			
96	monthly time series to analyze the seasonal controls and resolve any interannual or multiyear			
97	temporal patterns.			
98	1.1 The study area			
99	The study area covers, from Northnorth to Southsouth, the interconnected Raunefjord	F	formatted: Font color: Text 1	
100	(centered around 60.27°N; 5.17°E), the Korsfjord (centered around 60.17°N; 5.21°E),	- F	ormatted: Font color: Text 1	
101	Langenuen, and southern parts of the Hardangerfjord-, which are all situated atalong the	F	ormatted: Font color: Text 1	
102	western coast of mainland Norway (Fig. 1). The area stretches over some 60 km, but the main	- F	ormatted: Font color: Text 1	
103	focus here will be on the area from the Korsfjord to the Hardangerfjord from which the vast			
104	majority of the data has been acquired.			
105	The bathymetry and hydrographic conditions of the fjords have been described elsewhere			
106	(Helle, 1978; Mathew and Sands, 1973; Bakke and Sands, 1977; Erga and Heimdal, 1984;			
107	Asplin et al., 2014). In the following only a brief account, based on the above studies, is			
108	given.			
109	The Korsfjord is 690 m deep in its main basin and situated about 25 km south of the			
110	Norway's second largest city, Bergen. To the west itKorsfjord is relatively well connected to	F	formatted: Font color: Text 1	
111	the open coastal ocean of the northern North Sea through a 250 m deep sill at Marsteinen. To			
112	the north it connects with -the Raunefjord through the 100 m deep strait Lerøysundet -	F	formatted: Font color: Text 1	
113	between the islands Sotra and Lerøy. At the eastern end itthe fjord branches into the smaller	F	formatted: Font color: Text 1	
114	and shallower fjords Lysefjord and Fanafjord. To, and to the southwest it connects with the	F	formatted: Font color: Text 1	
115	open coast through the Selbjørnsfjord, witch which has a sill depth of 180 m at Selbjørn. To	F	formatted: Font color: Text 1	
116	the south it connects to the Hardangerfjord through the 25 km long and 300 m deep strait			
117	Langenuen.			
118	The Hardangerfjord is a 179 km long fjord ranking as the fourth longest fjord in the world. It			
119	stretches from the coastal open ocean in the southwest to the mountainous interior of Norway.			
120	Our study includes the southern parts of the fjord. This is bounded by the larger islands Stord			
121	and Tysnesøya in the north, the Haugaland peninsula in the south, and the smaller islands			
122	Fjellbergøya and Halsnøya on the south/east side. This part of the fjord is over 300 m deep in			
123	its basin (around 59.76N; 5.55E) and connects with the smaller fjords Ålfjord and Bjoafjord			
124	in the south.			
125	In the fjord system run-off from land mixmixes with salty water originating from the	F	formatted: Font color: Text 1	
126	northward flowing Norwegian Coastal Current (NCC, to produce a typically salinity	F	Formatted: Font color: Text 1	

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present the mean distribution across the different fjords (Korsfjord-Langenuen-

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127	stratified water column with a complex circulation, forced both by external and internal			
128	factors. In particular, the coastal windwinds have a profound influence on the water	F	ormatted: Font color: Text 1	
129	circulation of the deep water that	F	ormatted: Font color: Text 1	
130	follows periods of prolonged northerly winds (Svendsen, 1981; Erga and Heimdal, 1984).	F	ormatted: Font color: Text 1	
121	Basidas wind forcing the hydrography of the fiords is also influenced by winter cooling	[F	ormatted: Font color: Text 1	
131	besides wind <u>toreing</u> , the hydrography of the fjords is also influenced by winter cooling,	F	ormatted: Font color: Text 1	_
132	summer warming, show melt and run-off. The fjords also receive freshwater through the			
133	NCC, which carries water originating from the Baltic Sea and rivers in the southern North Sea			
134	(Skagseth et al., 2011 and references therein). However, snow melt and run-off are the main			
135	local sources for freshwater since the fjord system is generally ice free. Thus, on seasonal	F	ormatted: Font color: Text 1	_
136	time scales, the fjords are generally salinity stratified drives stratification during spring-	F	ormatted: Font color: Text 1	
137	summer and the water column is more homogenous during winter. Additionally, Asplin et al.	F	ormatted: Font color: Text 1	
138	(2014) reported regular episodes of water exchange between Hardangerfjord and the NCC	F	ormatted: Font color: Text 1	
139	that homogenized the upper 50 m of the fjord by mixing with coastal water. During these			
140	events the water temperature inside the Hardangerfjord regularly becomes identical with that	F	ormatted: Font color: Text 1	_
141	of the adjacent coastal open oceanNorth Sea (Asplin et al., 2014).	F	ormatted: Font color: Text 1	_
1/12	The water Water exchange with the NCC is important for the fiord ecosystems as it supplies		ormatted: Font color: Text 1	
142	The water water exchange with the Nee is important for the fjord ecosystems as it supplies	(F	ormatted: Font color: Text 1	
143	nutrients and oxygen to the area (Aure and Stigebrandt, 1989). In response, the fjords enhance	{F	ormatted: Font color: Text 1	
144	their primary production is enhanced in the fjords, which enables them to hostsupport rich and	F	ormatted: Font color: Text 1	
145	diverse marine life (Erga and Heimdal, 1984; Erga, 1989; Salvanes and Noreide, 1993).		ormatted: Font color: Text 1	
146	Erga and Heimdal (1984) studied the dynamics of the spring bloom in the Korsfjord and	F	ormatted: Font color: Text T	
147	estimated a total primary production of 74 g C m ⁻² during the period -February – June.	F	ormatted: Font color: Text 1	
148	Further, they reported light regime and water column stability to be decisive fordominant			
149	controls of the onset of the bloom. They also pointed out that changes in the alongshore wind	F	ormatted: Font color: Text 1	_
150	component are important for the bloom dynamics, with persistent northerly winds inducing			
151	upwelling of nutrient-rich coastal water that promotes blooming while the opposite situation	F	ormatted: Font color: Text 1	
152	follows on from persistent southerly winds. During calm periods strong stratification	F	ormatted: Font color: Text 1	
153	develops, which can ultimately lead to nutrient exhaustion in the <u>upper</u> water column.	F	ormatted: Font color: Text 1	_
154	The study area, with its adjacent waters, is ecologically and economically important because	F	ormatted: Font color: Text 1	
155	it covers spawning grounds for a number of different fish species (Lie et al., 1978;	F	ormatted: Font color: Text 1	
156	Johannessen et al., 2014). Additionally, the largest concentration of coral reefs in western			
157	Norway is found in the Langenuen strait (Fosså, 2015). The fjord system also contributes to			
158	the important aquaculture production that, with its annual fish production of >700 tonnes,			
159	ranks Norway within the tenth place worldwide. About one fifth of this is produced in the			
160	Hordaland County where the fjord system studied here is situated (http://www.diercke.com),	F	ormatted: Font color: Text 1	_

161 2. Data and methods

2.1 Weekly underway VOS data 162

163 Weekly underway measurements-of of fugacity of CO₂ in seawater (fCO₂) and SST were obtained aboard the containership MS Trans Carrier (operated by Seatrans AS, Norway, 164 www.seatrans.no). During the study period, the ship sailed from Bergen to ports in 165 166 southwestern Norway on a weekly basis. It passed through several fjords including the 167 Korsfjord and the Hardangerfjord (Fig. 1). It), then crossed the North Sea mostly along a 168 transect roughly at 5°E longitude to Amsterdam, Netherlands, and then back on the same route (Omar et al., 2010). The measurement method used aboard MS Trans Carrier was 169 170 described in Omar et al. (2010). The instrument recorded one fCO₂ and SST measurement about every three minutes and automatically shut off when the ship approached ports in 171 Bergen (20-30 km from port ≈60.2°N) and Amsterdam, in order to protect the inlet filter from 172 potentially polluted seawater. Between February and December 2006 the VOS line was 173 serviced by a sister ship, MS Norcliff, which was equipped with the same measurement 174 system in that period. 175

Underway dataduring that period. The VOS line was in operation in the period September 176

2005 to September 2009. Data acquired between $59.74^{\circ}N - 60.16^{\circ}N$ and $5.17^{\circ}E - 5.58^{\circ}E$ 177 178 (The Korsfjord, Langenuen, and southern parts of the Hardangerfjord) from September 2005 179 to September 2009 are used for the current analyses. This dataset will be referred to as the 180 UW (e.g. UW fCO₂ and UW SST) which stands for underway. The UW data from the years 181 2005, 2006, and 2007 are available from the SOCAT database (http://www.socat.info/). (http://www.socat.info/), the 2008 and 2009 UW data has been submitted for the SOCAT 182 183 Version 4 release.

2.2 Cruise and fixed station data

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188

We augment the VOS data with station data acquired during scientific cruises in the study 185 area in the period 2007-2010 and in 2015, and during regular visits (1-4 times per month) to a 186 fixed station in the Raunefjord in 2007 and 2008. Table 1 summarizes details of these three datasets, which will be referred to as the CS, 2015 and RF datasets, respectively.

189 Five of the cruises were conducted in the Korsfjord and the Raunefjord (Fig. 1, Table 1) onboard RV Hans Brattstrøm as part of the EU FP7 educational project CarboSchools (CS) in 190 2007-2010. The CS dataset covers mainly the spring and summer seasons reflecting the 191

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192	somewhat opportunistic nature of the sampling campaign. The 2015 cruise took place during	
193	fall (September 24) as part of the Ocean Acidification project funded by the Norwegian	
194	Environment Agency, and measurements were taken at three stations in the Korsfjord,	
195	Langenuen and southern Hardangerfjord (Fig.1, red squares).	
196	During each of the above cruises water samples were collected for analyses of parameters	
197	including DIC, total alkalinity (TA), salinity and temperature at 1-2 stations. The DIC	
198	concentrations were determined by the coulometric method (e.g. Johnson et al., 1993) with a	
199	precision of $\pm 1 \ \mu$ mol kg ⁻¹ . TA was measured by potentiometric titration with strong acid	
200	(HCl), and a precision of $\pm 2 \mu$ mol kg ⁻¹ . Accuracy was checked by using Certified Reference	
201	Material supplied by A. Dickson (SIO). Once all samples have been corrected with respect to	
202	offsets determined from the CRM measurements, the DIC and TA measurements were	
203	accurate to within the respective measurement precision (above). Only surface data	Formatted: Font color: Text 1
204	(depth<=4m) from within the geographical rectangle $59.74-60.34^{\circ}N$ and $5.17-5.55^{\circ}E$ were	
205	used in the current study.	
206	The Department of Biology, UoBUniversity of Bergen has acquired CTD (SAIV) data from a	Formatted: Font color: Text 1
207	fixed station in Raunefjord (RF) during 27 days in 2007 and 35 days in 2008 as part of a	
208	monitoring program close to the Marine Biological field Station at Espegrendstation at	
209	Espegrend. These data contained temperature and salinity profiles with one meter resolution.	
210	Averages of the uppermost five meters have been used in this study and will be referred to as	
211	the RF dataset,	Formatted: Font color: Text 1
212	2.3 In situ pH sensor data	
213	In January 2012 we carried out an evaluation of two pH sensors of the type Submersible	
214	Autonomous Moored Instruments (SAMI_pH, second generation) at the Marine Biological	
215	field Station at the eastern shore of the Raunefjord. The sensors were suspended from a	
216	wooden frame attached to the floating docks around a raft-house in the fjord –some hundred	
217	meters from land. The instruments were submersed at about one meter depth in the fjord and	
218	were left for 50 hours starting 24.01.2012 10:00 GMT, recording one measurement each hour.	
219	A full description of the measurement method for these instruments is found at	
220	http://www.sunburstsensors.com/. In addition to pH, these instruments also recorded the	
221	seawater temperature, and they have measurement precision and accuracy of <0.001 and $+/-$	
222	0.003 pH units, respectively, During the test, salinity was also recorded using a Seaguard	Formatted: Font color: Text 1

223	RCM from Aanderaa Data instruments. These sensor data were used to assess the uncertainty	
224	in our <u>pH values</u> estimated pH values as described in section 3.1.	Formatted: Font color: Text 1
225	2.4 Mathada	Formatted: Font color: Text 1
225	2.4 Methods	
226	2.4.1 Complete seawater CO_2 chemistry from SST and fCO_2	
227	A complete description of the seawater CO_2 chemistry from the UW SST and UW f CO_2 data	
228	collected onboard MS Trans Carrier has been obtained through a 3-step procedure. This is	
229	similar to the procedure described in Nondal et al. (2009) with the main modification being	
230	that in the current study, sea surface salinity (SSS) was determined from empirical	
231	relationship with SST,	Formatted: Font color: Text 1
232	First, the RF dataset has been used to determine the regional SSS versus SST relationship.	
233	The RF data was chosen for this purpose because it covered all seasons well, both in 2007 and	
234	2008. The identified regional SSS-SST relationship allowed us to estimate a SSS value for	
235	each UW SST observation from MS Trans Carrier. This step was necessary because the total	
236	number of measured SSS values were less than 150 data points, while the available underway	
237	SST and fCO ₂ data were much more numerous (> 9900 data points), covering most of the	
238	study area during the years 2005-2009. The remaining SST and SSS data (CS, and from	
239	sensors) were used for evaluation to verify that SST-SSS relationship is valid for the whole	
240	study area (section 3.1). Salinity values estimated from SST will be denoted as SSS(sst).	
241	Second, we determined TA from SSS(sst) and SST using an algorithm we identified for the	
242	region using the CS dataset. This allows us to estimate a corresponding alkalinity value for	
243	each UW fCO2 observation obtained from MS Trans Carrier. Alkalinity values estimated from	
244	measured SSS and SST data will be denoted as TA(sss), whereas TA values estimated from	
245	SSS(sst) and SST values will be denoted as TA(sst).	
246	The UW fCO ₂ together with TA (sst), UW SST , and other ancillary parameters <u>SSS(sst)</u> were	Formatted: Font color: Text 1
247	then used to characterize the full seawater CO ₂ chemistry using CO2SYS (Lewis and	Formatted: Font color: Text 1
248	Wallace, 1998; van Heuven et al., 2011), with K1 and K2 constants from Lueker et al. (2000).	
249	The CO2SYS calculation also gives DIC, pH, Ω_{Ar} and all other seawater CO ₂ chemistry	
250	variables. The data estimated using this three stage procedure will be denoted pH(sst) and	
251	$\Omega_{Ar}(sst)$ and are the main focus of this study.	

252	pH and Ω_{Ar} values based on TA(sss) and fCO ₂ will be denoted as pH(sss) and Ω_{Ar} (sss),	
253	whereas values that are either measured or computed from measured TA and DIC will be	
254	denoted as simply pH and Ω_{Ar} . <u>nDIC denotes the DIC values normalized to constant salinity</u>	
255	(the mean value) according to Friis et al. (2003) with freshwater end member DIC	
256	concentration of 1039 µmol kg ⁻¹ inferred from the cruise data. An overview of the symbols	
257	used for estimated and derived quantities used in this study is given in Table 2.	
258	3. Results and discussion	
259	3.1 Correlations and validations	
260	In this section we present the regression equations identified in this study in addition to	
261	validating the various estimation procedures used by comparing the estimated values with	
262	those measured/computed. The results of these comparisons are summarized in Table 3. For	
263	each comparison, the coefficient of determination (R ²) and the significance level (<i>p</i> -value) are	
264	used as metrics for the goodness of the correlation while the associated root-mean-square	
265	error (rmse) is benchmarked against the maximum target uncertainties developed by the	
266	Global Ocean Acidification Network (GOA-ON) and the California Current Acidification	
267	<u>Network (C-CAN) of ±0.2 for Ω_{Ar} (McLaughlin et al., 2015), which corresponds to maximum</u>	
268	uncertainties of ± 0.02 , ± 1.25 or ± 1.8 in pH, SST, or SSS, respectively.	
269	The regional SST-SSS relationship obtained from the RF dataset is given by Eq. 1 and is	
270	depicted in Fig. 2a (filled symbols). Despite a clear covariation between SST and SSS, there	
271	is a lot of scatter in the data and the statistics of the regression equation is not particularly	
272	strong (Eq. 1). The observed correlation most probably arises from the annual cycles; during	
273	summer the study area embodies warm water diluted by runoff, whereas during winter the	
274	surface water is colder and saltier due to little or no runoff. The magnitude of these annual	
275	variations varies with time and space and this is reflected by the high <u>degree of scatter</u> in the Formatted : Font color: Text 1	
276	relationship. Consequently, the identified regression model is able to explain only 27% of the	
277	salinity variations. Nonetheless, the independent station and sensor data (dots, squares, and	
278	stars), which have been acquired from the whole study area, fits well in different seasons, falls,	
279	into a pattern around the relationship described by Eq.1 with an RMSa root-squared-mean	5
280	error, of 0.81 psu. Thus, these data confirm from here on we assume that Eq. 1 indeed is	5
281	representative forable to estimate the seasonal SSS variations across the whole study region.	
282	To verify this we have compared the monthly averages of RE_SSS data with values obtained	
202	Formatted:	\dashv
283	using Eq.1 and monthly KF_SS1. As shown in the last row of Table 3, the estimated values	

284	were significantly correlated with the monthly RF_SSS ($R^2=0.65$ and p=0.002) and the	
285	resulting rmse of 0.3 was lower than the benchmark values of ±1.8.	Formatted: Font color: Text 1
286	$\frac{SSS = -0.142SST + 31.09}{SSS = -0.142SST + 32.09}, \text{ for SSS>29; R^2=0.27; n=61;}$	Formatted: Font color: Text 1
287	rmsrmse=1.2-psu. (1)	Formatted: Font color: Text 1
		Formatted: Font color: Text 1
288	As a further verification of the spatial representativeness of that the RF SST dataset is spatially <	Formatted: Font color: Text 1
289	representative, we compared it with the chronologically co-located UW SST that have been	Formatted: Font color: Text 1
290	acquired onboard Trans Carrier inacross the whole study area. The two datasets arewere	Formatted: Font color: Text 1
250		Formatted: Font color: Text 1
291	found to be almost identical (Fig. 2b <u>; 3^{cd} row Table 3</u>).	Formatted: Font color: Text 1
292 293	The relationship between TA, SSS and SST is given by Eq. 2 according to:	
294	$TA = 30.84SSS - 4.689SST + 3625.4$ $TA = 32.09SSS - 4.39SST + 1210$, $R^2 = 0.9490$; $n = 25$;	Formatted: Font color: Text 1
205	rmc^{22} : $rmco^{-12}$ 0 $umolkc^{-1}$ (2)	Formatted: Font color: Text 1
295	$\frac{1115}{25}, \frac{1115}{111} = 15.0 \mu \text{more} (2)$	Formatted: Font color: Text 1
296		
297	Alkalinity is a semi-conservative parameter and is normally modelled as linear functions of	
298	salinity (e.g. Millero et al., 1998; Bellerby et al., 2005; Nondal et al, 2009). However,	
299	including SST as using a second fit multi-parameter linear regression with SST and SSS as	Formatted: Font color: Text 1
300	independent parameters improved the regression statics statistics considerably. ($R^2=0.90$;	Formatted: Font color: Text 1
301	$n=23$: rmse=13.0 µmolkg ⁻¹) compared to a linear regression with only SSS ($R^2=0.67$: $n=23$:	Formatted: Font color: Text 1
301	$\frac{1-25}{100}, \frac{1}{100}, \frac{1}{1$	Formatted: Font color: Text 1
302	<u>rmse=24.0 µmolkg</u>), This is probably because SST runctions<u>acts</u> as <u>an</u> indicator of the effect	Formatted: Font color: Text 1
303	of nutrient cycling on TA in agreement to what has been reported for the open Atlantic Ocean	Formatted: Font color: Text 1
304	(Lee et al, 2006).	
305	In order to assess the error introduced in pH(sst) and Ω_{Ar} (sst) we compared them withvalues	
306	based on the cruise and sensor data i.e. pH(sss) or pHTo estimate a corresponding TA value	
307	for each UW fCO ₂ observation obtained from MS Trans Carrier, we used salinity values	
308	estimated from the UW SST data by using Eq.1. The results (denoted as SSS(sst)) were then	
309	inputted into Eq.2 to obtain TA(sst) (see Table 2 for nomenclature). The fact that TA(sst) are	
310	based on SSS(sst) rather than measured SSS values introduce an additional error in the	
311	estimated pH(sst) and $\Omega_{Ar}(sst)$. In order to assess this error we compared pH(sst) and $\Omega_{Ar}(sst)$	
312	with values based on the cruise data, i.e. pH(sss) and Ω_{Ar} (sss). First, we computed pH(sss)	Formatted: Font color: Text 1
313	and $\Omega_{Ar}(sss)$ by combining all available measured SSS, estimated TA(sss) from Eq. 2, and co-	
314	located UW SST and UW fCO ₂ . Then we repeated the <u>calculation</u> <u>calculations</u> , but this time	Formatted: Font color: Text 1
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315	we replaced the measured SSS with estimated SSS(sst) from Eq. 1 to compute pH(sst) and		
316	Ω_{Ar} (sst). The very strong linear relationships between the resulting values in Figs. 2c and 2d		
317	(circles) confirms that the estimated pH(sst) and $\Omega_{Ar}(sst)$ reproduce very wellclosely the		Formatted: Font color: Text 1
318	measurement-based values of pH(sss) and $\Omega_{Ar}(sss)$ for the whole study area. This is also		
319	evident from comparison statistics on rows 1 and 2 in Table 3 which show that measured		
320	based values are well correlated ($R^2 \approx 1$, p<0.001) with those estimated with rmse values of		
321	0.003 and 0.04 for pH(sss) and Ω_{Ar} (sst), respectively, which are well within the		
322	aforementioned maximum target uncertainties developed by the C-CAN (last column in Table		
323	3).		Formatted: Font color: Text 1
324	To quantify the total error associated with the pH(sst) and $\Omega_{Ar}(sst)$ estimates, we considered		
325	two main sources for error. First we computed the residuals (estimated – measurement-based)		
326	using the data showshown in Figs. 2c and 2d . (including the sensor data). The mean <	<	Formatted: Font color: Text 1
327	difference for the whole study area was $0.0018002 + - 0.0043004$ and $0.0048005 + - 0.077708$		Formatted: Font color: Text 1
328	for pH and Ω_{Ar} , respectively. Thus, the maximum probable error from this source is	\bigwedge	Formatted: Font color: Text 1
329	0.0061 and 0.08309 for pH and Ω_{Ar} respectively. Additionally, we estimated that the		Formatted: Font color: Text 1
330	computed and/or measured pH values included an error of 0.007 pH units, which under the	K'	Formatted: Font color: Text 1
221	current conditions (mean TA fCO, SST and SSS) would give an error of 0.038 in O_{1}	\backslash	Formatted: Font color: Text 1
227	These two error estimates were propagated to determine the total error in our estimates, which		Formatted: Font color: Text 1
332	These two error estimates were propagated to determine the total error in our estimates, which were found to be ± 0.0003 and ± 0.001 for pH and Ω_{1} _respectively. These error estimates are		
334	well within maximum target uncertainties developed by the Global Ocean Acidification		
334	Network (COA ON) and the California Current Acidification Network (C CAN) of ±0.2 for		
336	$\Omega_{\Lambda_{-}}$ which is indicative for maximum uncertainty of +0.02 in pH (McL aughlin et al.		
337	$\frac{2015}{2015}$ combined (as the square root of sum of squares) to determine the total error in our		
338	estimates, which were found to be ± 0.01 and ± 0.1 for pH and $O_{A_{2}}$ respectively. It must be		
339	noted that the above total error was derived from all available observational data including the		
340	in situ sensor data (shown in Fig. 2c and in described section 2.3), which are the only		
3/1	wintertime measurements used in this study. This is important because the lack of wintertime		
341	data in the CS dataset which was used for the identification of AT-SSS/SST relationship (Eq.		
342	2) means that wintertime AT(set) might be overestimated so that corresponding pH(set) values		
243	would be overestimated. In fact, during the aforementioned comparison between pH(sst) and		
2/5	measured nH we noted that for this particular dataset nH(sst) overestimated the		
240	measurements. However, the estimates were consistent with the observations to within the		
346	measurements. However, the estimates were consistent with the observations to within the		
347	total error of ± 0.01 pH units. Thus, by utilizing the above total errors, we also accounted for		

.

348	the effect of this possible caveat of Eq. 2 arising from the lack of wintertime TA		
349	measurements.		Formatted: Font color: Text 1
350	From the above we conclude that we are able to estimate pH(sst) and $\Omega_{Ar}(sst)$ across the		
351	whole study area and during all seasons with a total errors of ± 0.01 and ± 0.1 for pH and Ω_{Are}		
352	respectively.		
353	3.2 Spatiotemporal variations		Formatted: Font color: Text 1
354	Fig. 2 shows the data collapsed into one virtual year In order to present the mean		Formatted: Font color: Text 1
355	distribution distributions across the different fjords and throughout the annual cycle. There, we		Formatted: Font color: Text 1
356	condensed the data into one virtual year by projecting it onto non-equidistant rectangular grids	-	Formatted: Font color: Text 1
357	using the "weighted-average gridding" method of the Ocean Data View software (Schlitzer,		
358	2015). As evident from Fig. 3, there is a clear seasonality (for the interannual changes see		Formatted: Font color: Text 1
359	section 3.3) in both pH(sst) and Ω_{-1} (sst) (Fig. 3) The former varies between minimum values		Formatted: Font color: Text 1
260	$(\sim (8.05)$ around Naw Year to the tunical maximum values of around 8.225, which accur	\leq	Formatted: Font color: Text 1
500	(-18.05) around New Tear to the typical maximum varies of around 8.22., which occur		Formatted: Font color: Text 1
361	during the late winter and/or spring (March-April). This increase is due to the reduction of	\backslash	Formatted: Font color: Text 1
362	DIC (Fig. 3d), induced by the phytoplankton spring bloom. This clearly <u>counteracts and</u>		Formatted: Font color: Text 1
363	outweighs the negative effect on pH of warming the water column during this period.		Formatted: Font color: Text 1
364	However, during April/May, the latter processes effect of warming begins to dominate and		Formatted: Font color: Text 1
365	pH(sst) starts decreasing. By September SSTsthe SST starts decreasing, while pH continues to		Formatted: Font color: Text 1
366	drop. This is due to the effect of the fall mixing, which brings upenables carbonrich coastal		Formatted: Font color: Text 1
367	water to reach the surface layer, as mentioned in section 1, and asis reflected by the increasing		Formatted: Font color: Text 1
368	DIC during this period (Fig. 3d)	$\overline{\}$	Formatted: Font color: Text 1
200	Die during uns period (1 ig. 5d).	\mathbb{N}	Formatted: Font color: Text 1
369	The mean distribution of $\Omega_{Ar}(sst)$ also shows a significant seasonal variation. There are three		Formatted: Font color: Text 1
370	factors that drive this: (i) reduced concentrations of DIC by the spring bloom drives		
371	upenriches the concentration of the carbonate ionions, (ii) $\Omega_{Ar}(sst)$ increases with rising		Formatted: Font color: Text 1
372	temperature so that warming during the summer actually reinforces the increase of Ω_{Ar}	$\overline{}$	Formatted: Font color: Text 1
373	initiated by biological carbon uptake and (iii) reduced TA due to runoffs also reinforces the		Formatted: Font color: Text 1
374	Ω_{Λ} (sst) increase freshwater input from runoff and mixing of deeper carbon-rich water into		
375	surface layer reduce Ω_{1} (set) during fall. Thus Ω_{2} (set) reaches its maximum (>2.5) in July-	_	Formatted: Font color: Text 1
275	Surface rayer reduce 22 _{AT} (sst) during ran, Thus, 22 _{AT} (sst) reaches its maximum (22.3) in July-		
376	September, when the spring bloom is over and pH has already started decreasing (Fig. 3a, c).		
377	The lowest $\Omega_{Ar}(sst)$ values ($\approx 1.3-1.6$), on the other hand,) occur during winter (January-		Formatted: Font color: Text 1
378	March) when both pH and SST are low, and despite TA is high-due to high SSS values. The	\langle	Formatted: Font color: Text 1
379	mismatchdecoupling in the seasonal cycles of pH and Ω_{Ar} clearly supports the case that pH		Formatted: Font color: Text 1
			Tomatted. Full COLOR: Text T

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380	alone is not an adequate measure of ocean acidification, in accordance with the C-CAN			
381	recommendation that "measurements should facilitate determination of Ω_{Ar} and a complete			
382	description of the carbonate system, including pH and pCO ₂ " (McLaughlin et al., 2015).			
383	The above described seasonal variations in pH(sst) and $\Omega_{Ar}(sst)$ are spatially more or less			
384	coherent within the whole study area, except for the slight south-north gradient during May-			
385	September, with highest values south of 60°N (see Fig. 3a,c). All in all, during summertime			
386	the study area embody embodied warm surface water with high $\Omega_{Ar}(sst)$ and intermediate		Formatted: Font color: Text 1	
387	pH(sst) values. During winter, the surface water is cold with low $\Omega_{Ar}(sst)$ and pH(sst) values.			
388	3.3 Controls of seasonal variability and trends			
389	To investigate the seasonal variability more thoroughly, we arranged the data intocomputed		Formatted: Font color: Text 1	
390	monthly averages of pH(sst), SST, Ω_{Ar} (sst), and nDIC(sst) for one composite year. Then we			
391	quantified the effect of DIC, TA, SST and SSS on the monthly changes of pH(sst) and Ω_{Ar}			
392	(sst) in order to gain more insight into the processes governing the seasonal variations and			
393	their relative importance- <u>(Fig. 4).</u>		Formatted: Font color: Text 1	
394	For pH(sst) we used the decomposition method described in {Lauvset et al., (2015) to		Formatted: Font color: Text 1	
395	quantify the importance of different parameters. This method estimates the monthly pH		Formatted: Font color: Text 1	
396	changes expected from corresponding changes observed in SST, SSS, DIC, and TA as well as			
397	their sum. The results are shown on Fig. 4 (left panelsa-e) where it can be seen that DIC is the		Formatted: Font color: Text 1	
398	most important driver followed by SST and TA (Fig. 4), whereas SSS had a negligible effect		Formatted: Font color: Text 1	
399	(not shown) on the seasonal pH variations. We also note that the effects of SST and TA		Formatted: Font color: Text 1	
400	combined are nearly equal to, but opposite to that of DIC (Fig. 5e4 c.d.e). As a result, the sum		Formatted: Font color: Text 1	
401	of all effects is <0.06 pH units, and compares well to the observed amplitudes (Fig. $\frac{5a4a}{a}$).		Formatted: Font color: Text 1	
402	meaning that the decomposition model is able to account for the observed seasonal changes.			
403	Note also the TA control is identical to that of SST (Fig. $\frac{5e4c}{e}$). The reason for this is that		Formatted: Font color: Text 1	
404	TA values used here are obtained from SSS(sst) and SST using Eq. 2, which in effect means			
405	that they are based on SST. This emphasizes the need for measured SSS and TA values when			
406	the objective is to analyze the controls of pH and $\Omega_{Ar}(sst)$ variations.		Formatted: Font color: Text 1	
407	For $\Omega_{Ar}(sst)$ we investigated the importance of different drivers by letting one of the			
408	driverscontrols (DIC, TA, SST, SSS) to varyby varying them independently over itstheir		Formatted: Font color: Text 1	
409	observed range, while holding all other drivers constant, and re-computing $\Omega_{Ar}(sst)$. The	$\overline{\ }$	Formatted: Font color: Text 1	
410	magnitude of the standard deviation of the results is indicative for of the importance of the		Formatted: Font color: Text 1	
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411	varying driverdrivers. The result of this exercise is shown on Fig. 4(right panels).4f-i.		Formatted: Font color: Text 1
412	Evidently, the variations of SST and SSS are the least important drivers for $\Omega_{Ar}(sst)$ seasonal		Formatted: Font color: Text 1
413	changes, since varying these parameters induceinduces changes that are about an order of		Formatted: Font color: Text 1
414	magnitude less than the observed seasonal amplitude in $\Omega_{Ar}(sst)$. On the other hand, changing		Formatted: Font color: Text 1
415	DIC and TA (Fig. 4h,i) induceinduces changes that are comparable to the seasonal amplitude		Formatted: Font color: Text 1
416	observed in $\Omega_{Ar}(sst)$ (Fig. 4a). We therefore conclude that seasonal changes in DIC is and TA		Formatted: Font color: Text 1
417	are the most important driver for changes in $\Omega_{Ar}(sst)$ followed by TA.).		Formatted: Font color: Text 1
			Formatted: Font color: Text 1
418	From the above we conclude that the main drivers of $\Omega_{Ar}(sst)$ are DIC and TA, whereas for	\backslash	Formatted: Font color: Text 1
419	pH(sst). SST also have has a significant impact. In terms of processes, this. This means that		Formatted: Font color: Text 1
420	the formation and destruction of argonic matter together with veryalling of each on righ	<	Formatted: Font color: Text 1
420	the formation and destruction of organic matter together with upwening of carbon- <u>fich</u>		Formatted: Font color: Text 1
421	coastal water, seasonal warming and cooling, and runoff <u>inputs</u> , are the processes that govern		Formatted: Font color: Text 1
422	most of the seasonal variability of OA parameters within the study area. It then follows that		
423	interannual variability in the above processes would lead to corresponding variations in		
424	pH(sst) and Ω_{Ar} (sst-). Such interannual changes are evident from the monthly time series (Fig.		Formatted: Font color: Text 1
425	S1), where the rate of seasonal ehangeschange differs between the years, both for SST and		Formatted: Font color: Text 1
426	DIC normalized to the mean salinity (nDIC-) according to Friis et al. (2003), Additionally, for		Formatted: Font color: Text 1
		~	
427	SST, the extreme values also change between years. These changes are in turn reflected in the		Formatted: Font color: Text 1
427 428	SST, the extreme values also change between years. These changes are in turn reflected in the pH(sst) and $\Omega_{Ar}(sst)$. However, the) for which the amplitude of the interannual variability		Formatted: Font color: Text 1
427 428 429	SST, the extreme values also change between years. These changes are in turn reflected in the pH(sst) and $\Omega_{Ar}(sst)$. However, the) for which the amplitude of the interannual variability (IAV), calculated as the temporal standard deviation, is presented in Table 4. For pH, IAV		Formatted: Font color: Text 1
427 428 429 430	SST, the extreme values also change between years. These changes are in turn reflected in the pH(sst) and $\Omega_{Ar}(sst)$. However, the) for which the amplitude of the interannual variability (IAV), calculated as the temporal standard deviation, is presented in Table 4. For pH, IAV was normally much lower than the seasonal changes and ranged between 0.01 and 0.02		Formatted: Font color: Text 1
427 428 429 430 431	SST, the extreme values also change between years. These changes are in turn reflected in the pH(sst) and $\Omega_{Ar}(sst)$. However, the) for which the amplitude of the interannual variability (IAV), calculated as the temporal standard deviation, is presented in Table 4. For pH, IAV was normally much lower than the seasonal changes and ranged between 0.01 and 0.02 although higher changes were observed during the months April (0.04), and July and October		Formatted: Font color: Text 1
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443 year differences were not systematic, and no multiyear temporal trend was apparent from the 4-year time series analyzed in this study. 444 Formatted: Font color: Text 1 Formatted: Font color: Text 1 3.4 Inference of OA parameters from VOS underway data 445 446 Changes in the oceanic CO₂-system variables are related through ratios called Buffer Factors. Specifically, changes in Ω_{Ar} and pH in response to CO₂ variations can be quantified by partial 447 Formatted: Font color: Text 1 derivatives $(\gamma_{DIC}, \beta_{DIC}, \text{and}, \omega_{DIC}, \gamma_{DIC}, \beta_{DIC}, \text{and}, \omega_{DIC})$, which have been defined by Egleston 448 et al. (2010, their table 1), and the slope of these relationships can be expressed 449 450 mathematically by: $\frac{\partial \ln \Omega}{\partial \ln CO^2} = \frac{\gamma_{DIC}}{\omega_{DIC}} = \frac{DIC - Alk_c^2/S}{DIC - Alk_c P/HCO_3^2}$ 451 $\partial \ln \Omega / \partial \ln CO2 = \gamma_{DIC} / \omega_{DIC} = \frac{DIC - Alk_c^2 / S}{DIC - Alk_c P / HCO_3^2}$ Formatted: Font color: Text 1 452 (3)Formatted: Font: Times New Roman, 12 pt. Font color: Text 1 Formatted: Font color: Text 1 $\frac{\partial \ln H^+ / \partial \ln CO2}{\partial \ln CO2} = \frac{\gamma_{DIC}}{\beta_{DIC}} = \frac{(DIC - Alk_c^2) / S}{(DICS - Alk_c^2) / Alk_c}$ 453 Formatted: Font color: Text 1 $\partial \ln H^+ / \partial \ln CO2 = \gamma_{DIC} / \beta_{DIC} = \frac{(DIC - Alk_c^2) / S}{(DICS - Alk_c^2) / Alk_c}$ 454 (4) 455 where expressions for the carbonate alkalinity Alk_C and the parameters P and S are Formatted: Font: Italic, Font color: Text 1 Formatted: Font color: Text 1 givendefined in Egleston et al. (2010). We have evaluated the right hand sides of Eqs. 3 and 456 Formatted: Font: Italic, Font color: Text 1 4, using the CS cruise data, and the results showed that these quantities change only a few per 457 Formatted: Font color: Text 1 cents (1.3 and 3.4 %, respectively) due to seasonal changes in the various variables. The ratio Formatted: Font color: Text 1 458 Formatted: Font color: Text 1 $\gamma_{DIC} \gamma_{DIC} / \omega_{DIC} \omega_{DIC}$ changed by 1-6 % and ranged from -1.08 to -0.980, while $\gamma_{DIC} \gamma_{DIC} / \beta_{DIC}$ 459 Formatted: Font color: Text 1 Formatted: Font color: Text 1 460 $\beta_{\rm DIC}$ changed by 0.5-3 % and ranged from 0.84 to 0.88. This, together with the fact that Formatted: Font color: Text 1 Formatted: Font color: Text 1 461 equations 43 and 54 can be defined in terms of $ln(fCO_2)$ instead of ln(CO2) (Egleston et al., Formatted: Font color: Text 1 462 2010; Takahashi et al., 1993), suggests that in situations where underway surface fCO_2 and Formatted: Font color: Text 1 463 SST are frequently measured, while the CO_2 system is fully determined only occasionally, an easy way of interpolating the seasonality in pH and Ω_{Ar} is to predict them from fCO₂. We 464 have implemented this alternative way of estimating pH and Ω_{Ar} using the CS cruise data. For 465 the estimation of Ω_{Ar} we used $\frac{fCO_{2ts}fCO_{2@meanSST_{e}}}{fCO_{2@meanSST_{e}}}$ which is fCO₂ adjusted to constant 466 Formatted: Font color: Text 1 temperature and salinity (i.e. at mean SST and SSS), because these 467 Formatted: Font color: Text 1 Formatted: Font color: Text 1 468 normalizationsnormalization, improved the regression significantly. Since we were interested

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470	$ln(fCO_{24})$ (CO _{24@meanser}). The results are shown in Fig. 5 and conform to tight relationships		Formatted: Font color: Text 1]
170	hat we have a supervised of the results are shown in Fig. 5 and comparison of the result of the results of the	$\overline{}$	Formatted: Font color: Text 1	J
4/1	between computed pH and $ln(ICO_2)$ values (Fig. 5a), and between computed Ω_{Ar} and		Formatted: Font color: Text 1	Ĺ
472	$ln(fCO_{2ts}fCO_{2t@meanSST})$ (Fig. 5b). Further, by using linear curve fitting we determined the		Formatted: Font color: Text 1	J
473	relationships according to:			
474	$pH = -0.389 \ln fCO_{2ts} + 10.354_{77} pH = -0.389 \ln fCO_2 + 10.354_{2} R^2 = 0.99; n = 28;$		Formatted: Font color: Text 1)
475	<u>rmsrmse</u> =0. 0047<u>005.</u> (5) _ <	<	Formatted: Font color: Text 1	כ
			Formatted: Font color: Text 1	۲
476	$\frac{\Omega_{Ar} = \exp(-0.738 \ln fCO_{2ts} + 5.010)}{\Omega_{Ar}}, \Omega_{Ar} = \exp(-0.6741 \ln fCO_{2ts} + 4.6422)$		Formatted: Font color: Text 1	J
477	$R^2 = 0.9894; n = 28; rmsrmse = 0.05907.$ (6).		Formatted: Font color: Text 1	ר
		$\langle -$	Formatted: Font color: Text 1	ๅ
478	The magnitude of the residuals (computed – estimated) associated with pH and Ω_{Ar} values		Formatted: Font color: Text 1	j
479	obtained from the above relationships were 0.0005000 +/- 0.0049005 and 0.01201 +/-		Formatted: Font color: Text 1	
480	0.06106, respectively, which is comparable to the residuals associated with pH(sst) and	$\overline{}$	Formatted: Font color: Text 1]
181	$\Omega_{\rm c}$ (set) (section Table 3.3) An advantage of this procedure, however, is that it utilizes much	$\overline{\ }$	Formatted: Font color: Text 1	Ĺ
401	Search (section 1 able 5.5). All advantage of this procedure, nowever, is that it duringes much	\sim	Formatted: Font color: Text 1	۲
482	tighter empirical relationships and, involves fewer computational steps, and thus is based on	\backslash	Formatted: Font color: Text 1	┥
483	<u>UW</u> data, which are much more numerous than station data from oceanography cruises. Thus,	$\overline{\ }$	Formatted: Font color: Text 1	۲
484	it minimizes errors introduced by intermediate results such as the TA-SSS/SST regression in		Formatted: Font color: Text 1	ๅ
485	Eq. 22 and/or seasonal data coverage, Furthermore, a direct comparison revealed that values		Formatted: Font color: Text 1	Ĵ
486	obtained from Eqs. 5 and 6 were almost identical withto those of -pH(sst) and $\Omega_{Ar}(sst)$ (Fig.		Formatted: Font color: Text 1	
487	S2).) with values for R ² , p-value, and rmse of 1, 0, and 0.003 for pH; and 1, 0, and 0.02 for		Formatted: Font color: Text 1	J
488	$\Omega_{Ar_{2}}$ However, it is important to realize that for the above procedure too, a representative full		Formatted: Font color: Text 1	
489	description of the carbonate system is necessary for up-to-date determinations of Eqs. 5 and 6.			
490	Further, this calibration data ideally should include high frequency time series observations,			
491	since the slopes (i.e. Eqs. 3 and 4) change slightly with the carbonate system variables (e.g.			
492	DIC and TA, see Eqs. 3 and 4), which vary on multiple time scales (hours-days-years).			
493	Furthermore, the procedure is based on measurements of only one of the four master			
494	parameters constituting the carbonate system (i.e. fCO ₂). Therefore, it only provides a way to			
495	interpolate pH and Ω_{Ar} values, but cannot support the analyses of controls that have been			
496	provided in the proceeding section.			
497	From Fig. 5b we note that lowest Ω_{Ar} values are associated with the highest			

498 fCO_{2ts}fCO_{2@meanSST} values, which occur during late fall and winter. Monitoring of these

499 extreme values are of special interest because: (i) during late fall and early winter the

in pH and Ω_{Ar} we plotted these parameters directly against ln-(fCO₂) or

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500	upwelling of carbonrich water to the surface occurs so that the and surface water also reflects	Formatted: Font color: Text 1
501	the properties of the deeper water properties, and (ii) the rate of change at this point (lowest	Formatted: Font color: Text 1
502	$\Omega_{\rm Ar}$, highest fCO ₂₀₀ fCO ₂₀₀ mean SST) indicates the time when under-saturation of calcium	Formatted: Font color: Text 1
503	carbonate can be expected in these waters. To estimate this for the current data we used Eq. 5	Formatted: Font color: Text 1
505	and the charaction that the class (i.e. Eq. 2) and interpret descended have been 0,000 and	Formatted: Font color: Text 1
504	and the observation that the slope (i.e. Eq. 3) and intercept decreased by about 0.0008 and	
505	0.004 for every 1 μ atm increase in mean $\frac{fCO_{2ts}}{FCO_{2@meanSST}}$. We also took into account	
506	<u>an uncertainty of ± 0.2 in the Ω_{Ar} estimates and found that Ω_{Ar} becomes undersaturated (<1)</u>	Formatted: Font color: Text 1
507	when mean $\frac{fCO_{2ts}annual fCO_{2@meanSST_i}}{fCO_{2ts}annual fCO_{2@meanSST_i}}$ is about $\frac{300310\pm70}{2000}$ µatm higher than its present value.	Formatted: Font color: Text 1
508	(310 µatm), For business as usual emission scenario (RCP 8.5), this is equivalent to about	Formatted: Font color: Text 1
509	year 2070 ± 10 if we assume that the development in the ocean follows that of the atmosphere	Formatted: Font color: Text 1
510	(i.e. constant disequilibrium between ocean and atmosphere)	Formatted: Font color: Text 1
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511	4. Summary and concluding remarks:	Formatted: Font color: Text 1
512	On the basis of four years of weekly underway fCO_2 and SST data combined with less	
513	frequentsporadic data from research cruises, the ocean acidification parameters pH(sst) and	Formatted: Font color: Text 1
514	$\Omega_{Ar}(sst)$ have been estimated and analyzed for western Norway fjords stretching over more	
515	than 60 km from the Korsfjord, through Langenuen strait, to southern parts of the	
516	Hardangerfjord. The total errors associated with the estimated values, ± 0.009301 and ± 0.0001	Formatted: Font color: Text 1
517	for pH and Ω_{Ar} , were an order of magnitude lessabout 50% lower than the maximum target	Formatted: Font color: Text 1
518	uncertainties developed by the Global Ocean Acidification Network.	Formatted: Font color: Text 1
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519	Strong seasonal variations, more or less spatially coherent over the whole study area, were	
520	found for OA parameters in the surface waters of the fjords. These changes were governed	
521	mainly by the formation and decay of organic matter, vertical mixing with deeper carbon rich	
522	coastal water, and the seasonal changes in SST and SSS. The annual mean pH was 8.13, and	
523	this parameter varies typically between minimum values (≈ 8.05) around January to maximum	Formatted: Font color: Text 1
524	values of around 8.2, which occur during the spring and/or late winter (March-April) as a	
525	consequence of the phytoplankton spring bloom, which that reduces DIC levels. However,	Formatted: Font color: Text 1
526	sometime during April/May, the effect of warming starts to dominate, and pH(sst) starts	Formatted: Font color: Text 1
527	decreasing. Later during fall, deepening of the mixed layer brings upallows carbon-rich	Formatted: Font color: Text 1
528	coastal water tomix into the surface and reinforces the decrease in pH_which continuous	Formatted: Font color: Text 1
520	through out follow due as all until the law winter volues are reached assir	Formatted: Font color: Text 1
529	throughout tanreduces priguntil the low winter values are reached again.	Formatted: Font color: Text 1
530	The mean value of $\Omega_{Ar}(sst)$ was found to be 2.21, and it reached its maximum (>2.5) in mid to	Formatted: Font color: Text 1
531	late summer (July to September) when the spring bloom is over and pH has started to	

532	decrease. The lowest $\Omega_{Ar}(sst)$ values (\approx 1.3-1.6), on the other hand, occurred during winter	
533	(January-March), when both pH and SST are low, and DIC is at its highest.	
534	Within the study area, Strong correlations of pH and Ω_{Ar} have been found to correlate strongly	Formatted: Font color: Text 1
535	with fCO ₂ and $\frac{fCO_{2ts}fCO_{2}@meanSST_{4}}{fCO_{2}}$ adjusted to the mean temperature and salinity),	Formatted: Font color: Text 1
536	respectively. These correlations provide an approach to interpolate pH and Ω_{Ar} over large	Formatted: Font color: Text 1
537	areas in the fiords of western Norway where underway measurements of fCO ₂ , SST, and SSS	Formatted: Font color: Text 1
538	are available. However, both the slopes and the intercepts of these correlations vary slightly	Formatted: Font color: 1 ext 1
530	with DIC and TA. Therefore, the most accurate interpolations will be achieved if the	
535	relationships are calibrated with high frequency observations of the complete carbonate	
540	relationships are canorated with high nequency observations of the complete carbonate	
541	system, measured at few strategically placed fixed stations.	
542	The Ω_{Ar} - $\frac{fCO_{2ts}fCO_{2@meanSST_{x}}}{fCO_{2@meanSST_{x}}}$ relationship, and the rate of change of its slope and intercept	Formatted: Font color: Text 1
543	with DIC, have been used to project the time when under-saturation of calcium carbonate	
544	eancould be expected to occur in the study area. This is expected to occur in the year 2070, if	Formatted: Font color: Text 1
545	we assume business as usual emission scenario (RCP 8.5), and that oceanic development of	Formatted: Font color: Text 1
546	fCO ₂ followsCO ₂ concentrations follow, that of the atmosphere (i.e. constant disequilibrium	Formatted: Font color: Text 1
547	between ocean and atmosphere).	
548	5. Acknowledgements	
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556	and E. MJones) that have improved the manuscript.	Formatted: Font color: Text 1
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1	Table 1: Details aboutof, the CarboSchools (CS) and Raunefjord (RF) cruise datasets. The	Formatted: Font color: Text 1
2	plus sign denotes the parameters for which sampling/measurement were carried out. For the	Formatted: Font color: Text 1
3	RF dataset, each data point represents the average of five measurements acquired in the upper	
4	five meters.	Formatted: Font color: Text 1
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04/25/2007	5.19	60.34	1	+	+	+	+			
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A	08/25/2009	5.19	60.34	1	+	+	+	+	/
	08/25/2009	5.2	60.34	11	+	+	+	+	/
	08/25/2009	5.19	60.33		+	+	+	+	/
	08/27/2009	5.19	60.33	1	+	+	+	+	/
	08/27/2009	5.19	60.33	1	+	+	+	+	/
_	08/27/2009	5.18	60.17	1	+	+	+	+	/
A	08/27/2009	5.18	60.17	1	+	+	+	+	/
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2015; Korsfjord	09/29/2015			5	+	+	+	+	Omar
									I. Skjelvan / A.
2015; Langenuen	09/29/2015			5	+	+	+	+	Omar
									I. Skjelvan / A.
2015; Hardangerfjord	09/29/2015			5	+	+	+	+	Omar

Table 1(continuecontinued)

Datasett; area	Date(m/d/y)	Lon_E Lat_N	Depth (m)	DIC	ТА	SST	SSS	Reference/ originator
								S. R. Erga /
RF; Raunefjord	01/03/2007		1-5			+	+	Egge
	01/23/2007		1-5			+	+	//
A	02/13/2007		1-5			+	+	
	02/27/2007		1-5			+	+	//
A	03/07/2007		1-5			+	+	
A	03/13/2007		1-5			+	+	/
	03/27/2007		1-5			+	+	
	04/10/2007		1-5			+	+	
A	04/17/2007		1-5			+	+	
	04/23/2007		1-5			+	+	/`
	05/08/2007		1-5			+	+	
	05/19/2007		1-5			+	+	
	06/05/2007		1-5			+	+	
	06/12/2007		1-5			+	+	
	06/19/2007		1-5			+	+	
<u> </u>	08/31/2007		1-5			+	+	
	09/04/2007		1-5			+	+	
	09/11/2007		1-5			+	+	
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10/31/2007	1-5	+ +		
11/27/2007	1-5	+ +		
12/11/2007	1-5	+ +	/ \	
01/02/2008	1-5	<u>+</u> +		
02/05/2008	1-5	+ +		
02/21/2008	1-5	+ +		
03/05/2008	1-5	+ +		
03/11/2008	1-5	+ +		
03/25/2008	1-5	+ +		
03/31/2008	1-5	+ +		
04/08/2008	1-5	+ +		
04/22/2008	1-5	+ +		
04/29/2008	1-5	+ +		
05/06/2008	1-5	+ +		
05/13/2008	1-5	+ +		
05/20/2008	1-5	+ +		
05/27/2008	1-5	+. +		
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10/21/2008	1-3	+ +		
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Table 32: Overview of the symbols used for quantities estimated and/or derived from the measurement-based

variables SSS, SST, TA, pH, DIC, fCO_2, and Ω_{Ar}

Symbol	Meaning
TA(sss)	TA values estimated from measured SSS and SST using Eq. 2.
$pH(sss), \Omega_{Ar}(sss)$	pH, and Ω_{Ar} values estimated by combining TA (sss) and fCO ₂ .
SSS(sst)	SSS values estimated from SST using Eq. 1.

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	TA value	es determined	from estin	nated SSS	(sst) and SST using Eq. 2	·	Formatted: Font color: Text 1		
$DH(sst), \Omega_{Ar}(sst), DIC(sst)$	Values o	f pH, Ω_{Ar} and	d DIC that h	have been	obtained by combining T	A (sst),	Formatted: Font color: Text 1		
	fCO ₂ and	l ancillary va	riables.						
fCO _{2t}	fCO ₂ at t	he mean tem	perature				Formatted: Font color: Text 1		
fCO _{2ts}	CO _{2ts} fCO ₂ at the mean temperature and salinity								
nDIC	DIC norr	nalized to the	e mean salii	nity			Formatted: Font color: Text 1		
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as standard deviations, in the study area for the period 2005-2009.

		<u>Jan</u>	<u>Feb</u>	<u>Mar</u>	<u>Apr</u>	<u>May</u>	<u>Jun</u>	<u>Jul</u>	Aug	<u>Sep</u>	<u>Oct</u>	Nov	<u>Dec</u>
pH(sst)	Mean	<u>8.08</u>	<u>8.10</u>	<u>8.16</u>	<u>8.19</u>	<u>8.18</u>	<u>8.15</u>	<u>8.15</u>	<u>8.17</u>	<u>8.14</u>	<u>8.11</u>	<u>8.10</u>	<u>8.08</u>
	IAV	<u><0.01</u>	<u>0.01</u>	<u>0.04</u>	<u>0.01</u>	<u>0.02</u>	<u>0.02</u>	<u>0.03</u>	<u>0.02</u>	<u>0.02</u>	<u>0.03</u>	<u>0.02</u>	<u>0.02</u>
$\Omega_{Ar}(sst)$	<u>Mean</u>	<u>1.7</u>	<u>1.7</u>	<u>1.9</u>	<u>2.1</u>	<u>2.3</u>	<u>2.4</u>	<u>2.6</u>	<u>2.7</u>	<u>2.4</u>	<u>2.2</u>	<u>1.9</u>	<u>1.8</u>
	IAV	<u>0.1</u>	<u>0.1</u>	<u>0.1</u>	<u>0.1</u>	<u>0.1</u>	<u>0.2</u>	<u>0.1</u>	<u>0.1</u>	<u>0.1</u>	<u>0.1</u>	<u><0.05</u>	<u><0.05</u>

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775 Figure texts:

Fig. 1: An overview map of western Norway with a detailed map of the study area showing the positions from whichwhere cruise and underway data have been acquired. The <u>tickthick</u> grey arrow indicates the approximate position of the Norwegian Coastal Current (NCC).

779Fig. 2: A) RF SSS as a function of SST (filled symbols) with the regression line described by Eq.1. Sampling780month is indicated by the color of the data points. The CS (dots), 2015 (squares), and sensor (stars) datasets are781also shown for comparison with the regression line. B) Compares RF SST with chronologically co-located UW782SST acquired from the whole study area during 2008 (blue) and 2007 (red). C) Compares pH(sst) with pH values783that have been measured or computed from TA and DIC. Symbols are as in Fig. 1. D) Compares $\Omega_{Ar}(sst)$ with784 Ω_{Ar} values that have been computed from measured TA and DIC or from measured pH and UW fCO2. Symbols785are as in Fig. 1.

Fig. 3: <u>A)</u> Estimated pH(sst) and <u>)</u>, B) UW SST, C) estimated Ω_{Ar}(sst), and D) estimated DIC which have been normalized to the mean salinity of 30.5 as a function of latitude and time of the year. All data from 2005-2009 have been collapsedcondensed into one virtual year to resolve the spatial and seasonal variations.

789Fig. 4: left panel: Monthly pH changes (Δ pH) as observed (A) and expected due to: sum of all derivers (B), SST790changes (C), DIC changes (D) and by TA changes (E). right panel: Standard deviations in monthly mean Ω_{Ar} as791a result of variations in all parameters (F) or only in SST (G) in DIC (H) in TA (I).

Fig. 5: A) and B) pH and Ω_{Ar} from CS (dots) and 2015 (red squares) cruises plotted as a function of $ln(fCO_2)$ and $ln(\frac{nfCO_{2e}fCO_2@meansST})$, respectively.

FigS1: Time series of monthly variations in **A**) pH(sst), **B**) SST, **C**) $\Omega_{Ar}(sst)$ and **D**) nDIC for the whole study area in 2005-2009.

FigS2: A) compares pH(sst) with pH values obtained from Eq. 5 (y-axis). B) Compares Ω_{Ar}(sst) with values
 obtained from Eq. 6 (y-axis).

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