



1 **Aragonite saturation states and pH in western Norway fjords:**
2 **seasonal cycles and controlling factors, 2005-2009**
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8 **Abstract:**

9 The uptake of anthropogenic CO₂ by the ocean leads to a process known as ocean
10 acidification (OA) which lowers aragonite saturation state (Ω_{Ar}) and pH, and this is poorly
11 documented in coastal environments including fjords due to lack of appropriate observations.

12 Here we use weekly underway data from Voluntary Observing Ships (VOS) covering the
13 period 2005-2009 combined with data from research cruises to estimate Ω_{Ar} and pH values in
14 several adjacent western Norwegian fjords, and to evaluate how seawater CO₂ chemistry
15 drives their variations in response to physical and biological factors.

16 The OA parameters in the surface waters of the fjords are characterized by strong seasonal
17 and spatially coherent variations. These changes are governed by the seasonal changes in
18 temperature, salinity, formation and decay of organic matter, and vertical mixing with deeper,
19 carbon rich coastal water. Annual mean pH and Ω_{Ar} values were 8.13 and 2.21, respectively.
20 The former varies from minimum values (≈ 8.05) in late December - early January to
21 maximum values of around 8.2 during early spring (March-April) as a consequence of the
22 phytoplankton spring bloom, which reduces Dissolved Inorganic Carbon (DIC). In the
23 following months, pH decreases in response to warming. This thermodynamic decrease in pH
24 is reinforced by the deepening of the mixed layer, which brings up carbon rich coastal water
25 to the surface, and this trend continues until the low winter values are reached again. Ω_{Ar} , on
26 the other hand, reaches its seasonal maximum (>2.5) in mid to late summer (July -Sept), when
27 the spring bloom is over and pH is decreasing. The lowest Ω_{Ar} values ($\approx 1.3-1.6$) occur during
28 winter (Jan-Mar), when both pH and Sea Surface Temperature (SST) are low and DIC is
29 highest. Consequently, seasonal Ω_{Ar} variations align with those of SST and salinity
30 normalized DIC.

31 We demonstrate that underway measurements of fugacity of CO₂ in seawater (fCO_2), SST,
32 and SSS from VOS lines combined with high frequency observations of the complete



33 carbonate system at strategically placed fixed stations provide an approach to interpolate OA
34 parameters over large areas in the fjords of western Norway.

35 **1. Introduction:**

36 The continued emissions of carbon dioxide (CO₂) (Peters et al., 2013) are of global concern,
37 not only because they are the main drivers of anthropogenic global warming, but also because
38 of the changes in the ocean chemistry they incur (Ciais et al., 2013). The increase in the
39 atmospheric CO₂ concentration drives a net ocean CO₂ uptake, which leads to higher proton
40 (H⁺) concentration i.e. lower pH, lower concentration of carbonate ion (CO₃²⁻) and, lower
41 saturation state (Ω) for calcium carbonate in seawater. This process is known as ocean
42 acidification (OA) (e.g. Royal Society, 2005), and it has direct and indirect effects on
43 biological activity in the ocean (e.g. Gattuso and Hansson, 2011) including reported inhibition
44 of biogenic calcification by marine organisms which precipitate 0.5–2.0 Gt of carbon as
45 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
47 during the last decade. Observations from time series stations and volunteer observing ships
48 in different oceanic regions consistently show systematic changes in surface ocean chemistry
49 that result from OA. Specifically, long-term negative trends of pH and saturation state for
50 aragonite (Ω_{Ar}) have been observed (e.g. Lauvset et al., 2015; Bates et al., 2014).

51 For coastal regions observed rates of pH change largely differ from those expected from
52 oceanic CO₂ uptake alone, as variations in other biogeochemical processes, related for
53 example to changes in nutrient loading and eutrophication, are important as well (Clargo et al.
54 2015; Provoost et al., 2010; Wootton et al., 2008).

55 The Norwegian west coast (Fig. 1) is dominated by fjords, narrow and deep estuaries, carved
56 by glacial processes, with a sill in the mouth where they connect to the coastal North Sea.
57 Apart from being important recreation areas and marine pathways, these fjords are important
58 ecosystems and their physics, marine life, and associated environmental pressures have been
59 relatively well studied (e.g. Matthews and Sands, 1973; Erga and Heimdal, 1984; Asplin et
60 al., 2013; Brattgard et al., 2011; Stigebrandt, 2012; and references therein).

61 However, only a few studies on the carbon cycle of Norwegian fjords exist in the literature.
62 This hampers our understanding of the natural variability and controls of seawater carbonate
63 chemistry, which is prerequisite for sound estimates of OA in these ecosystems. Generally, in



64 the Northern Hemisphere, high latitudes coastal regions are thought to be sinks for
65 atmospheric CO₂, while low-latitude regions are thought to be sources (Borges et al., 2005;
66 Cai et al., 2006; Chavez et al., 2007; Chen and Borges, 2009). The few existing studies of
67 Norwegian fjords confirm the above picture; they act as an annual net sink for atmospheric
68 CO₂ (Fransson et al., 2014; Omar et al., 2005).

69 The carbon cycle of the northern North Sea, to which the western Norway fjords are
70 connected, has been well studied (Thomas et al., 2004; 2005; 2007; 2008; Bozec et al., 2005;
71 2006; Omar et al., 2010). However, observation-based OA estimates are still scarce. Recently,
72 Clargo et al. (2015) observed a rapid pH decrease in the North Sea, but after accounting for
73 biological processes, they estimated an ocean acidification rate consistent with concurrent
74 atmospheric and open ocean CO₂ increases over the period they studied, 2001-2011.

75 OA (and generally the carbon cycle) in fjords in western Norway has not been described
76 previously. Filling this knowledge gap is important because the fjords are important spawning
77 grounds for different fish species (Salvanes and Noreide, 1993; Johannessen et al., 2014),
78 productions sites for pelagic calcifiers (Berge, 1962; Erga and Heimdal, 1984; Frette et al.,
79 2004), the home for some coral reefs (e. g. Fosså et al., 2002), and significant food sources
80 due to the aquaculture industry which operates there.

81 In this study, we present estimates of OA parameters in the surface waters of several adjacent
82 western Norway fjords (Fig. 1), based mainly on weekly underway data from Voluntary
83 Observing Ships (VOS) covering the period 2005-2009. We combine the underway data with
84 available station data from research cruises to facilitate a complete description of the seawater
85 CO₂ chemistry in accordance with the recommendations of OA core principles by
86 McLaughlin et al. (2015). We focus on analyses of Ω_{Ar} and pH values and evaluate their
87 variations in response to the physical and biological factors: summer warming and
88 stratification, spring phytoplankton bloom, and deep mixing during fall and winter. First we
89 present mean distribution across the different fjords (Korsfjord-Langenuen-Hardangerfjord) to
90 understand the spatiotemporal patterns, then we collapse all data into a monthly time series to
91 analyze the seasonal controls and resolve any interannual or multiyear temporal patterns.

92 **1.1 The study area**

93 The study area covers, from North to South, the interconnected Raunefjord (centered around
94 60.27°N; 5.17°E), the Korsfjord (centered around 60.17°N; 5.21°E), Langenuen, and southern
95 parts of the Hardangerfjord, which are all situated at the western coast of mainland Norway



96 (Fig. 1). The area stretches over some 60 km, but the main focus here will be on the area from
97 the Korsfjord to the Hardangerfjord from which the vast majority of the data has been
98 acquired.

99 The bathymetry and hydrographic conditions of the fjords have been described elsewhere
100 (Helle, 1978; Mathew and Sands, 1973; Bakke and Sands, 1977; Erga and Heimdal, 1984;
101 Asplin et al., 2014). In the following only a brief account, based on the above studies, is
102 given.

103 The Korsfjord is 690 m deep in its main basin and situated about 25 km south of the
104 Norway's second largest city, Bergen. To the west it is relatively well connected to the open
105 coastal ocean of the northern North Sea through a 250 m deep sill at Marsteinen. To the north
106 it connects with the Raunefjord through the 100 m deep strait Lerøysundet - between the
107 islands Sotra and Lerøy. At the eastern end it branches into the smaller and shallower fjords
108 Lysefjord and Fanafjord. To the southwest it connects with the open coast through the
109 Selbjørnsfjord, which has a sill depth of 180 m at Selbjørn. To the south it connects to the
110 Hardangerfjord through the 25 km long and 300 m deep strait Langenuen.

111 The Hardangerfjord is a 179 km long fjord ranking as the fourth longest fjord in the world. It
112 stretches from the coastal open ocean in the southwest to the mountainous interior of Norway.
113 Our study includes the southern parts of the fjord. This is bounded by the larger islands Stord
114 and Tysnesøya in the north, the Haugaland peninsula in the south, and the smaller islands
115 Fjellbergøya and Halsnøya on the south/east side. This part of the fjord is over 300 m deep in
116 its basin (around 59.76N; 5.55E) and connects with the smaller fjords Ålfjord and Bjoafjord
117 in the south.

118 In the fjord system run-off from land mix with salty water originating from the northward
119 flowing Norwegian Coastal Current (NCC, to produce a typically salinity stratified water
120 column with a complex circulation, forced both by external and internal factors. In particular,
121 the coastal wind have a profound influence on the circulation of western Norwegian fjords
122 producing episodic renewal of the deep water that follows periods of prolonged northerly
123 winds (Svendsen, 1981; Erga and Heimdal, 1984).

124 Besides wind, the hydrography of the fjords is also influenced by winter cooling, summer
125 warming, snow melt and run-off. Thus, on seasonal time scales, the fjords are generally
126 salinity stratified during spring-summer and more homogenous during winter. Additionally,
127 Asplin et al. (2014) reported regular episodes of water exchange between Hardangerfjord and
128 the NCC that homogenized the upper 50 m of the fjord by mixing with coastal water. During



129 these events the temperature inside the Hardangerfjord regularly becomes identical with that
130 of the coastal open ocean (Asplin et al., 2014).
131 The water exchange with the NCC is important for the fjord ecosystems as it supplies
132 nutrients and oxygen (Aure and Stigebrandt, 1989). In response, the fjords enhance their
133 primary production which enables them to host rich and diverse marine life (Erga and
134 Heimdal, 1984; Erga, 1989; Salvanes and Noreide, 1993).
135 Erga and Heimdal (1984) studied the dynamics of the spring bloom in the Korsfjord and
136 estimated a total primary production of 74 g C m^{-2} during the period February – June.
137 Further, they reported light regime and water column stability to be decisive for the onset of
138 the bloom. They also pointed out that changes in the alongshore wind component are
139 important for the bloom dynamics, with persistent northerly winds inducing upwelling of
140 nutrient rich coastal water that promotes blooming while the opposite situation follows
141 persistent southerly winds. During calm periods strong stratification develops, which can
142 ultimately lead to nutrient exhaustion in the water column.
143 The study area with its adjacent waters is ecologically and economically important because it
144 covers spawning grounds for a number of different fish species (Lie et al., 1978; Johannessen
145 et al., 2014). Additionally, the largest concentration of coral reefs in western Norway is found
146 in the Langenuen strait (Fosså, 2015).

147 **2. Data and methods**

148 **2.1 Weekly underway VOS data**

149 Weekly underway measurements of fugacity of CO_2 in seawater ($f\text{CO}_2$) and SST were
150 obtained aboard the containership MS Trans Carrier (operated by Seatrans AS, Norway,
151 www.seatrans.no). During the study period, the ship sailed from Bergen to ports in
152 southwestern Norway on a weekly basis. It passed through several fjords including the
153 Korsfjord and the Hardangerfjord (Fig. 1). It then crossed the North Sea mostly along a
154 transect roughly at 5°E longitude to Amsterdam, Netherlands, and then back on the same
155 route (Omar et al., 2010). The measurement method used aboard MS Trans Carrier was
156 described in Omar et al. (2010). The instrument recorded one $f\text{CO}_2$ and SST measurement
157 about every three minutes and automatically shut off when the ship approached ports in
158 Bergen (20-30 km from port $\approx 60.2^\circ\text{N}$) and Amsterdam, in order to protect the inlet filter from
159 potentially polluted seawater. Between February and December 2006 the VOS line was



160 serviced by a sister ship, MS Norcliff, which was equipped with the same measurement
161 system in that period.

162 Underway data acquired between 59.74°N – 60.16°N and 5.17°E – 5.58°E (The Korsfjord,
163 Langenuen, and southern parts of the Hardangerfjord) from September 2005 to September
164 2009 are used for the current analyses. This dataset will be referred to as the UW (e.g. UW
165 fCO₂ and UW SST) which stands for underway. The UW data from the years 2005, 2006, and
166 2007 are available from the SOCAT database (<http://www.socat.info/>).

167 **2.2 Cruise and fixed station data**

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

172 Five of the cruises were conducted in the Korsfjord and the Raunefjord (Fig. 1, Table 1)
173 onboard RV Hans Brattstrøm as part of the EU FP7 educational project CarboSchools (CS) in
174 2007-2010. The CS dataset covers mainly the spring and summer seasons reflecting the
175 somewhat opportunistic nature of the sampling campaign. The 2015 cruise took place during
176 fall (September 24) as part of the Ocean Acidification project funded by the Norwegian
177 Environment Agency, and measurements were taken at three stations in the Korsfjord,
178 Langenuen and southern Hardangerfjord (Fig.1, red squares).

179 During each of the above cruises water samples were collected for analyses of parameters
180 including DIC, total alkalinity (TA), salinity and temperature at 1-2 stations. The DIC
181 concentrations were determined by the coulometric method (e.g. Johnson et al., 1993) with a
182 precision of $\pm 1 \mu\text{mol kg}^{-1}$. TA was measured by potentiometric titration with strong acid
183 (HCl), and a precision of $\pm 2 \mu\text{mol kg}^{-1}$. Accuracy was checked by using Certified Reference
184 Material supplied by A. Dickson (SIO). Only surface data (depth $\leq 4\text{m}$) from within the
185 geographical rectangle 59.74-60.34°N and 5.17-5.55°E were used in the current study.

186 The Department of Biology, UoB has acquired CTD (SAIV) data from a fixed station in
187 Raunefjord (RF) during 27 days in 2007 and 35 days in 2008 as part of a monitoring program
188 close to the Marine Biological field Station at Espegrend.

189 **2.3 In situ pH sensor data**



190 In January 2012 we carried out an evaluation of two pH sensors of the type Submersible
191 Autonomous Moored Instruments (SAMI_pH, second generation) at the Marine Biological
192 field Station at the eastern shore of the Raunefjord. The sensors were suspended from a
193 wooden frame attached to the floating docks around a raft-house in the fjord –some hundred
194 meters from land. The instruments were submersed at about one meter depth in the fjord and
195 were left for 50 hours starting 24.01.2012 10:00 GMT, recording one measurement each hour.
196 A full description of the measurement method for these instruments is found at
197 <http://www.sunburstsensors.com/>. In addition to pH, these instruments also recorded the
198 seawater temperature. During the test, salinity was also recorded using a Seaguard RCM from
199 Aanderaa Data Instruments. These sensor data were used to assess the uncertainty in our
200 estimated pH values.

201 **2.4 Methods**

202 2.4.1 Complete seawater CO₂ chemistry from SST and fCO₂

203 A complete description of the seawater CO₂ chemistry from the UW SST and UW fCO₂ data
204 collected onboard MS Trans Carrier has been obtained through a 3-step procedure. This is
205 similar to the procedure described in Nondal et al. (2009) with the main modification being
206 that in the current study, sea surface salinity (SSS) was determined from empirical
207 relationship.

208 First, the RF dataset has been used to determine the regional SSS versus SST relationship.
209 The RF data was chosen for this purpose because it covered all seasons well, both in 2007 and
210 2008. The identified regional SSS-SST relationship allowed us to estimate a SSS value for
211 each UW SST observation from MS Trans Carrier. This step was necessary because the total
212 number of measured SSS values were less than 150 data points, while the available underway
213 SST and fCO₂ data were much more numerous (> 9900 data points), covering most of the
214 study area during the years 2005-2009. The remaining SST and SSS data (CS, and from
215 sensors) were used for evaluation to verify that SST-SSS relationship is valid for the whole
216 study area (section 3.1). Salinity values estimated from SST will be denoted as SSS(sst).

217 Second, we determined TA from SSS(sst) and SST using an algorithm we identified for the
218 region using the CS dataset. This allows us to estimate a corresponding alkalinity value for
219 each UW fCO₂ observation obtained from MS Trans Carrier. Alkalinity values estimated from



220 measured SSS and SST data will be denoted as TA(sss), whereas TA values estimated from
221 SSS(sst) and SST values will be denoted as TA(sst).

222 The UW fCO₂ together with TA (sst) and other ancillary parameters were then used to
223 characterize the full seawater CO₂ chemistry using CO2SYS (Lewis and Wallace, 1998; van
224 Heuven et al., 2011), with K1 and K2 constants from Lueker et al. (2000). The CO2SYS
225 calculation also gives DIC, pH, Ω_{Ar} and all other seawater CO₂ chemistry variables. The data
226 estimated using this three stage procedure will be denoted pH(sst) and Ω_{Ar}(sst) and are the
227 main focus of this study.

228 pH and Ω_{Ar} values based on TA(sss) and fCO₂ will be denoted as pH(sss) and Ω_{Ar}(sss),
229 whereas values that are either measured or computed from measured TA and DIC will be
230 denoted as simply pH and Ω_{Ar}. An overview of the symbols used for estimated and derived
231 quantities used in this study is given in Table 2.

232 3. Results and discussion

233 3.1 Correlations and validations

234 The regional SST-SSS relationship obtained from the RF dataset is given by Eq. 1 and is
235 depicted in Fig. 2a (filled symbols). Despite a clear covariation between SST and SSS, there
236 is a lot of scatter in data and the statistics of the regression equation is not particularly strong
237 (Eq. 1). The observed correlation most probably arises from the annual cycles; during summer
238 the study area embodies warm water diluted by runoff, whereas during winter the surface
239 water is colder and saltier due to little or no runoff. The magnitude of these annual variations
240 varies with time and space and this is reflected by the high scatter in the relationship.
241 Consequently, the identified regression model is able to explain only 27% of the salinity
242 variations. Nonetheless, the independent station and sensor data (dots, squares, and stars),
243 which have been acquired from the whole study area, fits well into the relationship described
244 by Eq.1 with an RMS of 0.81 psu. Thus, these data confirm that Eq. 1 indeed is representative
245 for the whole region.

$$246 \quad SSS = -0.142SST + 31.09, \text{ for } SSS > 29; R^2 = 0.27; n = 61; \text{rms} = 1.2 \text{ psu.} \quad (1)$$

247 As a further verification of the spatial representativeness of the RF SST dataset, we compared
248 it with the chronologically co-located UW SST that have been acquired onboard Trans Carrier
249 in the whole study area. The two datasets are found to be almost identical (Fig. 2b).



250 The relationship between TA, SSS and SST is given by Eq. 2 according to:

251

$$252 \quad TA = 30.84SSS - 4.689SST + 3625.4, R^2=0.91; n=25; rms=13.0 \mu\text{molkg}^{-1}. \quad (2)$$

253

254 Alkalinity is a semi-conservative parameter and is normally modelled as linear functions of
255 salinity (e.g. Millero et al., 1998; Bellerby et al., 2005; Nondal et al, 2009). However,
256 including SST as a second fit parameter improved the regression statics considerably. This is
257 probably because SST functions as indicator of the effect of nutrient cycling on TA in
258 agreement to what has been reported for the open Atlantic Ocean (Lee et al, 2006).

259 In order to assess the error introduced in $\text{pH}(\text{sst})$ and $\Omega_{\text{Ar}}(\text{sst})$ we compared them with values
260 based on the cruise and sensor data i.e. $\text{pH}(\text{sss})$ or pH and $\Omega_{\text{Ar}}(\text{sss})$. First, we computed
261 $\text{pH}(\text{sss})$ and $\Omega_{\text{Ar}}(\text{sss})$ by combining all available measured SSS, estimated TA(sss) from Eq. 2,
262 and co-located UW SST and UW fCO_2 . Then we repeated the calculation, but this time we
263 replaced the measured SSS with estimated SSS(sst) from Eq. 1 to compute $\text{pH}(\text{sst})$ and
264 $\Omega_{\text{Ar}}(\text{sst})$. The very strong linear relationships between the resulting values in Figs. 2c and 2d
265 (circles) confirms that the estimated $\text{pH}(\text{sst})$ and $\Omega_{\text{Ar}}(\text{sst})$ reproduce very well the
266 measurement-based values of $\text{pH}(\text{sss})$ and $\Omega_{\text{Ar}}(\text{sss})$ for the whole study area.

267 To quantify the total error associated with the $\text{pH}(\text{sst})$ and $\Omega_{\text{Ar}}(\text{sst})$ estimates, we considered
268 two main sources for error. First we computed the residuals (estimated – measurement-based)
269 using the data show in Figs. 2c and 2d. The mean difference for the whole study area was
270 0.0018 ± 0.0043 and 0.0048 ± 0.0777 for pH and Ω_{Ar} , respectively. Thus, the maximum
271 probable error from this source is 0.0061 and 0.083 for pH and Ω_{Ar} , respectively. Additionally,
272 we estimated that the computed and/or measured pH values included an error of 0.007 pH
273 units, which under the current conditions (mean TA, fCO_2 , SST, and SSS) would give an error
274 of 0.038 in Ω_{Ar} . These two error estimates were propagated to determine the total error in our
275 estimates, which were found to be ± 0.0093 and ± 0.091 for pH and Ω_{Ar} , respectively. These
276 error estimates are well within maximum target uncertainties developed by the Global Ocean
277 Acidification Network (GOA-ON) and the California Current Acidification Network (C-
278 CAN) of ± 0.2 for Ω_{Ar} , which is indicative for maximum uncertainty of ± 0.02 in pH
279 (McLaughlin et al., 2015).

280 3.2 Spatiotemporal variations



281 Fig. 2 shows the data collapsed into one virtual year to present the mean distribution across
282 the different fjords and throughout the annual cycle. There is a clear seasonality in both
283 pH(sst) and Ω_{Ar} (sst) (Fig. 3). The former varies between minimum values (≈ 8.05) around
284 New Year to the maximum values of around 8.2, which occur during the late winter and/or
285 spring (March-April). This increase is due to the reduction of DIC (Fig. 3d), induced by the
286 phytoplankton spring bloom. This clearly outweighs the negative effect on pH of warming the
287 water column during this period. However, during April/May, the latter processes begins to
288 dominate and pH(sst) starts decreasing. By September SSTs starts decreasing, while pH
289 continues to drop. This is due to the effect of the fall mixing, which brings up carbon rich
290 coastal water to the surface, as mentioned in section 1, and as reflected by the increasing DIC
291 during this period (Fig. 3d).

292 The mean distribution of Ω_{Ar} (sst) also shows a significant seasonal variation. There are three
293 factors that drive this: (i) reduced concentrations of DIC by the spring bloom drives up the
294 concentration of the carbonate ion, (ii) Ω_{Ar} (sst) increases with rising temperature so that
295 warming during the summer actually reinforces the increase of Ω_{Ar} initiated by biological
296 carbon uptake, and (iii) reduced TA due to runoffs also reinforces the Ω_{Ar} (sst) increase. Thus,
297 Ω_{Ar} (sst) reaches its maximum (>2.5) in July-September, when the spring bloom is over and
298 pH has already started decreasing (Fig. 3a, c). The lowest Ω_{Ar} (sst) values ($\approx 1.3-1.6$), on the
299 other hand, occur during winter (January-March) when both pH and SST are low, and TA is
300 high. The mismatch in the seasonal cycles of pH and Ω_{Ar} clearly supports the case that pH
301 alone is not an adequate measure of ocean acidification, in accordance with the C-CAN
302 recommendation that “measurements should facilitate determination of Ω_{Ar} and a complete
303 description of the carbonate system, including pH and pCO_2 ” (McLaughlin et al., 2015).

304 The above described seasonal variations in pH(sst) and Ω_{Ar} (sst) are spatially more or less
305 coherent within the whole study area, except for the slight south-north gradient during May-
306 September, with highest values south of $60^\circ N$ (see Fig. 3a,c). All in all, during summertime
307 the study area embody warm surface water with high Ω_{Ar} (sst) and intermediate pH(sst)
308 values. During winter, the surface water is cold with low Ω_{Ar} (sst) and pH(sst) values.

309 3.3 Controls of seasonal variability and trends

310 To investigate the seasonal variability more thoroughly, we arranged the data into monthly
311 averages of pH(sst), SST, Ω_{Ar} (sst), and nDIC(sst) for one composite year. Then we
312 quantified the effect of DIC, TA, SST and SSS on the monthly changes of pH(sst) and Ω_{Ar}



313 (sst) in order to gain more insight into the processes governing the seasonal variations and
314 their relative importance.

315 For pH(sst) we used the decomposition method described in (Lauvset et al., 2015) to quantify
316 the importance of different parameters. The results are shown on Fig. 4 (left panels) where it
317 can be seen that DIC is the most important driver followed by SST and TA (Fig. 4), whereas
318 SSS had a negligible effect (not shown) on the seasonal pH variations. We also note that the
319 effects of SST and TA combined are nearly equal to, but opposite to that of DIC (Fig. 5c,d,e).
320 As a result, the sum of all effects is <0.06 pH units, and compares well to the observed
321 amplitudes (Fig. 5a), meaning that the decomposition model is able to account for the
322 observed seasonal changes. Note also the TA control is identical to that of SST (Fig. 5c,e).
323 The reason for this is that TA values used here are obtained from SSS(sst) and SST using Eq.
324 2, which in effect means that they are based on SST. This emphasizes the need for measured
325 SSS and TA values.

326 For $\Omega_{Ar}(sst)$ we investigated the importance of different drivers by letting one of the drivers
327 (DIC, TA, SST, SSS) to vary over its observed range, while holding all other drivers constant,
328 and re-computing $\Omega_{Ar}(sst)$. The magnitude of the standard deviation of the results is indicative
329 for the importance of the varying driver. The result of this exercise is shown on Fig. 4(right
330 panels). Evidently, the variations of SST and SSS are the least important drivers for $\Omega_{Ar}(sst)$
331 seasonal changes, since varying these parameters induce changes that are about an order of
332 magnitude less than the observed seasonal amplitude in $\Omega_{Ar}(sst)$. On the hand, changing DIC
333 and TA (Fig. 4h,i) induce changes that are comparable to the seasonal amplitude observed in
334 $\Omega_{Ar}(sst)$ (Fig. 4a). We therefore, conclude that DIC is the most important driver for $\Omega_{Ar}(sst)$
335 followed by TA.

336 From the above we conclude that the main drivers of $\Omega_{Ar}(sst)$ are DIC and TA, whereas for
337 pH(sst), SST also have a significant impact. In terms of processes, this means that the
338 formation and destruction of organic matter together with upwelling of carbon rich coastal
339 water, seasonal warming and cooling, and runoff, are the processes that govern most of the
340 seasonal variability of OA parameters within the study area. It then follows that interannual
341 variability in the above processes would lead to corresponding variations in pH(sst) and
342 $\Omega_{Ar}(sst)$. Such interannual changes are evident from the monthly time series (Fig. S1), where
343 the rate of seasonal changes differs between the years, both for SST and nDIC. Additionally,
344 for SST, the extreme values also change between years. These changes are in turn reflected in



345 the pH(sst) and Ω_{Ar} (sst). However, the year-to-year differences were not systematic, and no
 346 multiyear temporal trend was apparent from the time series.

347 3.4 Inference of OA parameters from VOS underway data

348 Changes in the oceanic CO₂-system variables are related through ratios called Buffer Factors.
 349 Specifically, changes in Ω_{Ar} and pH in response to CO₂ variations can be quantified by partial
 350 derivatives (γ_{DIC} , β_{DIC} , and ω_{DIC}), which have been defined by Egleston et al. (2010, their
 351 table 1), and the slope of these relationships can be expressed mathematically by:

$$352 \quad \partial \ln \Omega / \partial \ln CO_2 = \gamma_{DIC} / \omega_{DIC} = \frac{DIC - Alk_C^2 / S}{DIC - Alk_C P / HCO_3^-} \quad (3)$$

$$353 \quad \partial \ln H^+ / \partial \ln CO_2 = \gamma_{DIC} / \beta_{DIC} = \frac{(DIC - Alk_C^2) / S}{(DIC - Alk_C^2) / Alk_C} \quad (4)$$

354 where expressions for the carbonate alkalinity Alk_C and the parameters P and S are given in
 355 Egleston et al. (2010). We have evaluated the right hand sides of Eqs. 3 and 4, using the CS
 356 cruise data, and the results showed that these quantities change only a few per cents (1.3 and
 357 3.4 %, respectively) due to seasonal changes in the various variables. The ratio $\gamma_{DIC} / \omega_{DIC}$
 358 changed by 1-6 % and ranged from -1.08 to -0.980, while $\gamma_{DIC} / \beta_{DIC}$ changed by 0.5-3 % and
 359 ranged from 0.84 to 0.88. This, together with the fact that equations 4 and 5 can be defined
 360 in terms of $\ln(fCO_2)$ instead of $\ln(CO_2)$ (Egleston et al., 2010; Takahashi et al., 1993),
 361 suggests that in situations where underway surface fCO₂ and SST are frequently measured,
 362 while the CO₂ system is fully determined only occasionally, an easy way of interpolating the
 363 seasonality in pH and Ω_{Ar} , is to predict them from fCO₂. We have implemented this
 364 alternative way of estimating pH and Ω_{Ar} using the CS cruise data. For the estimation of Ω_{Ar}
 365 we used fCO_{2ts}, which is fCO₂ adjusted to constant temperature and salinity (i.e. at mean SST
 366 and SSS), because these normalizations improved the regression significantly. Since we were
 367 interested in pH and Ω_{Ar} we plotted these parameters directly against $\ln(fCO_2)$ or $\ln(fCO_{2ts})$.
 368 The results are shown in Fig. 5 and conform tight relationships between computed pH and
 369 $\ln(fCO_2)$ values (Fig. 5a), and between computed Ω_{Ar} and $\ln(fCO_{2ts})$ (Fig. 5b). Further, by
 370 using linear curve fitting we determined the relationships according to:

$$371 \quad pH = -0.389 \ln fCO_{2ts} + 10.354, \quad R^2=0.99; \quad n=28; \quad rms=0.0047 \quad (5)$$



$$372 \quad \Omega_{Ar} = \exp(-0.738 \ln fCO_{2ts} + 5.010) , R^2=0.98; n=28; rms=0.059 \quad (6).$$

373 The magnitude of the residuals (computed – estimated) associated with pH and Ω_{Ar} values
374 obtained from the above relationships were 0.0005 +/- 0.0049 and 0.012 +/- 0.061,
375 respectively, which is comparable to the residuals associated with pH(sst) and Ω_{Ar} (sst)
376 (section 3.3). An advantage of this procedure, however, is that it utilizes much tighter
377 empirical relationships and involves fewer computational steps, and thus minimizes errors
378 introduced by intermediate results such as the TA-SSS/SST regression in Eq. 2. Furthermore,
379 a direct comparison revealed that values obtained from Eqs. 5 and 6 were almost identical
380 with those of pH(sst) and Ω_{Ar} (sst) (Fig. S2). However, it is important to realize that for the
381 above procedure too, a representative full description of the carbonate system is necessary for
382 up-to-date determinations of Eqs. 5 and 6. Further, this calibration data ideally should include
383 high frequency time series observations, since the slopes (i.e. Eqs. 3 and 4) change slightly
384 with the carbonate system variables (e.g. DIC and TA, see Eqs. 3 and 4), which vary on
385 multiple time scales (hours-days-years). Furthermore, the procedure is based on
386 measurements of only one of the four master parameters constituting the carbonate system
387 (i.e. fCO_2). Therefore, it only provides a way to interpolate pH and Ω_{Ar} values, but cannot
388 support the analyses of controls that have been provided in the proceeding section.

389 From Fig. 5b we note that lowest Ω_{Ar} values are associated with the highest fCO_{2ts} values,
390 which occur during late fall and winter. Monitoring of these extreme values are of special
391 interest because: (i) during late fall and early winter the upwelling of carbon rich water to the
392 surface occurs so that the surface water also reflects the deeper water properties, and (ii) the
393 rate of change at this point (lowest Ω_{Ar} , highest fCO_{2ts}) indicates the time when under-
394 saturation of calcium carbonate can be expected in these waters. To estimate this for the
395 current data we used Eq. 5 and the observation that the slope (i.e. Eq. 3) and intercept
396 decreased by about 0.0008 and 0.004 for every 1 μatm increase in mean fCO_{2ts} . We found that
397 Ω_{Ar} becomes undersaturated (<1) when mean fCO_{2ts} is about 300 μatm higher than its present
398 value. For business as usual emission scenario (RCP 8.5), this is equivalent to about year
399 2070 if we assume that the development in the ocean follows that of the atmosphere (i.e.
400 constant disequilibrium between ocean and atmosphere).

401 **4. Summary and concluding remarks:**



402 On the basis of four years of weekly underway $f\text{CO}_2$ and SST data combined with less
403 frequent data from research cruises, the ocean acidification parameters $\text{pH}(\text{sst})$ and $\Omega_{\text{Ar}}(\text{sst})$
404 have been estimated and analyzed for western Norway fjords stretching over more than 60 km
405 from the Korsfjord, through Langenuen strait, to southern parts of the Hardangerfjord. The
406 errors associated with the estimated values, ± 0.0093 and ± 0.090 for pH and Ω_{Ar} , were an
407 order of magnitude less than the maximum target uncertainties developed by the Global
408 Ocean Acidification Network.

409 Strong seasonal variations, more or less spatially coherent over the whole study area, were
410 found for OA parameters in the surface waters of the fjords. These changes were governed
411 mainly by the formation and decay of organic matter, vertical mixing with deeper carbon rich
412 coastal water, and the seasonal changes in SST and SSS. The annual mean pH was 8.13, and
413 this parameter varies between minimum values (≈ 8.05) around January to maximum values of
414 around 8.2, which occur during the spring and/or late winter (March-April) as a consequence
415 of phytoplankton spring bloom, which reduces DIC levels. However, sometime during
416 April/May, the effect of warming starts to dominate, and $\text{pH}(\text{sst})$ starts decreasing. Later
417 during fall, deepening of the mixed layer brings up carbon rich coastal water to the surface,
418 and reinforces the decrease in pH , which continuous throughout fall until the low winter
419 values are reached again.

420 The mean value of $\Omega_{\text{Ar}}(\text{sst})$ was found to be 2.21, and it reached its maximum (>2.5) in mid to
421 late summer (July to September), when the spring bloom is over and pH has started to
422 decrease. The lowest $\Omega_{\text{Ar}}(\text{sst})$ values (≈ 1.3 -1.6), on the other hand, occurred during winter
423 (January-March), when both pH and SST are low, and DIC is at its highest.

424 Within the study area, pH and Ω_{Ar} have been found to correlate strongly with $f\text{CO}_2$ and $f\text{CO}_{2\text{ts}}$
425 ($f\text{CO}_2$ adjusted to the mean temperature and salinity), respectively. These correlations
426 provide an approach to interpolate pH and Ω_{Ar} over large areas in the fjords of western
427 Norway where underway measurements of $f\text{CO}_2$, SST, and SSS are available. However, both
428 the slopes and the intercepts of these correlations vary slightly with DIC and TA. Therefore,
429 the most accurate interpolations will be achieved if the relationships are calibrated with high
430 frequency observations of the complete carbonate system, measured at few strategically
431 placed fixed stations.

432 The $\Omega_{\text{Ar}} - f\text{CO}_{2\text{ts}}$ relationship, and the rate of change of its slope and intercept with DIC, have
433 been used to project the time when under-saturation of calcium carbonate can be expected in



434 the study area. This is expected to occur in the year 2070, if we assume business as usual
435 emission scenario (RCP 8.5), and that oceanic development of fCO₂ follows that of the
436 atmosphere (i.e. constant disequilibrium between ocean and atmosphere).

437 **5. Acknowledgements**

438 We are grateful for financial supports by the Research Council of Norway (RCN) through the
439 project FME SUCCESS, and by the Norwegian Environment Agency through the project
440 *Havforsuring*. This work would not have been possible without the generosity and help of
441 liner company SeaTrans AS and the captains and crew of MS Trans Carrier. We are grateful
442 for the technical assistance provided by Tor de Lange, Kristin Jackson and Tomas Sørлие.

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595 **Tables:**

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598 **Table 1:** Details about the CarboSchools (CS) and Raunefjord (RF) cruise datasets. The plus
 599 sign denotes the parameters for sampling/measurement were carried out.

Dataset; area	Date(m/d/y)	Lon (E)	Lat (N)	Depth (m)	DIC	TA	SST	SSS	Reference/ originator
CarboSchools (CS); Korsfjord/Raunefjord	04/13/2007	5.19	60.34	1	+	+	+	+	I. Skjelvan
	04/13/2007	5.18	60.17	1	+	+	+	+	
	04/25/2007	5.19	60.34	1	+	+	+	+	
	04/25/2007	5.17	60.17	1	+	+	+	+	
	09/04/2008	5.18	60.33	1	+	+	+	+	
	03/12/2009	5.17	60.15	1	+	+	+	+	
	03/12/2009	5.18	60.32	1	+	+	+	+	
	03/12/2009	5.17	60.17	1	+	+	+	+	
	03/12/2009	5.18	60.33	1	+	+	+	+	
	08/25/2009	5.17	60.17	1	+	+	+	+	
	08/24/2009	5.18	60.16	1	+	+	+	+	
	08/24/2009	5.18	60.16	1	+	+	+	+	
	08/24/2009	5.19	60.34	1	+	+	+	+	
	08/25/2009	5.19	60.34	1	+	+	+	+	
	08/25/2009	5.2	60.34	1	+	+	+	+	
	08/25/2009	5.19	60.33	1	+	+	+	+	
	08/27/2009	5.19	60.33	1	+	+	+	+	
	08/27/2009	5.19	60.33	1	+	+	+	+	
	08/27/2009	5.18	60.17	1	+	+	+	+	
	08/27/2009	5.18	60.17	1	+	+	+	+	
	08/27/2009	5.18	60.17	1	+	+	+	+	
	08/27/2009	5.2	60.33	1	+	+	+	+	
	09/08/2010	5.2	60.33	1	+	+	+	+	
2015; Korsfjord	09/29/2015			5	+	+	+	+	I. Skjelvan / A. Omar
2015; Langenuen	09/29/2015			5	+	+	+	+	I. Skjelvan / A. Omar
2015; Hardangerfjord	09/29/2015			5	+	+	+	+	I. Skjelvan / A. Omar

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603 Table 1(continue)

Datsett; area	Date(m/d/y)	Lon_E	Lat_N	Depth (m)	DIC	TA	SST	SSS	Reference/ originator
RF; Raunefjord	01/03/2007			1-5				+	S. R. Erga / J. Egge
	01/23/2007			1-5				+	
	02/13/2007			1-5				+	
	02/27/2007			1-5				+	
	03/07/2007			1-5				+	
	03/13/2007			1-5				+	
	03/27/2007			1-5				+	
	04/10/2007			1-5				+	
	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				+	
	08/31/2007			1-5				+	
	09/04/2007			1-5				+	
	09/11/2007			1-5				+	
	09/18/2007			1-5				+	
	09/26/2007			1-5				+	
	10/02/2007			1-5				+	
	10/09/2007			1-5				+	
	10/18/2007			1-5				+	
	10/31/2007			1-5				+	
	11/27/2007			1-5				+	
	12/11/2007			1-5				+	
	01/02/2008			1-5				+	
	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				+	
	06/04/2008			1-5				+	
	06/11/2008			1-5				+	
	06/17/2008			1-5				+	
	06/24/2008			1-5				+	
	07/01/2008			1-5				+	
	07/08/2008			1-5				+	
	07/16/2008			1-5				+	
	08/12/2008			1-5				+	
	08/19/2008			1-5				+	



08/26/2008	1-5	+
09/02/2008	1-5	+
09/09/2008	1-5	+
09/16/2008	1-5	+
09/23/2008	1-5	+
09/30/2008	1-5	+
10/07/2008	1-5	+
10/14/2008	1-5	+
10/21/2008	1-5	+
11/04/2008	1-5	+
11/20/2008	1-5	+
12/19/2008	1-5	+

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607 **Table 3:** Overview of the symbols used for quantities estimated and/or derived from the measurement-based
 608 variables SSS, SST, TA, pH, DIC, fCO₂, and Ω_{Ar}.

Symbol	Meaning
TA(sss)	TA values estimated from measured SSS and SST using Eq. 2.
pH(sss), Ω _{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.
TA(sst)	TA values determined from estimated SSS(sst) and SST using Eq. 2.
pH(sst), Ω _{Ar} (sst), DIC(sst)	Values of pH, Ω _{Ar} and DIC that have been obtained by combining TA (sst) , fCO ₂ and ancillary variables.
fCO _{2t}	fCO ₂ at the mean temperature
fCO _{2ts}	fCO ₂ at the mean temperature and salinity
nDIC	DIC normalized to the mean salinity

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618 **Figure texts:**

619 **Fig. 1:** An overview map of western Norway with a detailed map of the study area showing the positions from
620 which cruise and underway data have been acquired. The tick grey arrow indicates the approximate position of
621 the Norwegian Coastal Current (NCC).

622 **Fig. 2:** **A)** RF SSS as a function of SST (filled symbols) with the regression line described by Eq.1. Sampling
623 month is indicated by the color of the data points. The CS (dots), 2015 (squares), and sensor (stars) datasets are
624 also shown for comparison with the regression line. **B)** Compares RF SST with chronologically co-located UW
625 SST acquired from the whole study area during 2008 (blue) and 2007 (red). **C)** Compares pH(sst) with pH values
626 that have been measured or computed from TA and DIC. Symbols are as in Fig. 1. **D)** Compares $\Omega_{Ar}(sst)$ with
627 Ω_{Ar} values that have been computed from measured TA and DIC or from measured pH and UW fCO_2 . Symbols
628 are as in Fig. 1.

629 **Fig. 3:** Estimated pH(sst) and $\Omega_{Ar}(sst)$ as a function of latitude and time of the year. All data from 2005-2009
630 have been collapsed into one virtual year to resolve the spatial and seasonal variations.

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

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

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