



1                   **Seasonal Carbonate Chemistry Variability in Marine Surface Waters**  
2   **of the Pacific Northwest**

3  
4 Andrea J. Fassbender<sup>1,2,\*</sup>, Simone R. Alin<sup>2</sup>, Richard A. Feely<sup>2</sup>, Adrienne J. Sutton<sup>2</sup>, Jan A.  
5 Newton<sup>3</sup>, Christopher Krembs<sup>4</sup>, Julia Bos<sup>4</sup>, Mya Keyzers<sup>4</sup>, Allan Devol<sup>5</sup>, Wendi Ruef<sup>5</sup>, and Greg  
6 Pelletier<sup>4</sup>

7  
8 <sup>1</sup>Monterey Bay Aquarium Research Institute, Moss Landing, CA 95039, USA

9 <sup>2</sup>Pacific Marine Environmental Laboratory, National Oceanic and Atmospheric Administration,  
10 Seattle, WA 98115, USA

11 <sup>3</sup>Applied Physics Laboratory, University of Washington, Seattle, WA 98105, USA

12 <sup>4</sup>Washington State Department of Ecology, Olympia, WA 98504, USA

13 <sup>5</sup>School of Oceanography, University of Washington, Seattle, WA 98195, USA

14 \*Corresponding author: [fassbender@mbari.org](mailto:fassbender@mbari.org)

15  
16 **Abstract**

17       Fingerprinting ocean acidification (OA) in U.S. West Coast waters is extremely challenging  
18 due to the large magnitude of natural carbonate chemistry variations common to these regions.  
19 Additionally, quantifying a change requires information about the initial conditions, which is not  
20 readily available in most coastal systems. In an effort to address this issue, we have collated high-  
21 quality, publicly-available data to characterize the modern seasonal carbonate chemistry variability  
22 in marine surface waters of the Pacific Northwest. Underway ship data from Version 4 of the  
23 Surface Ocean CO<sub>2</sub> Atlas, discrete observations from various sampling platforms, and sustained  
24 measurements from regional moorings were incorporated to provide ~100,000 inorganic carbon  
25 observations from which modern seasonal cycles were estimated. Underway ship and discrete  
26 observations were merged and gridded to a 0.1°×0.1° scale. Eight unique regions were identified  
27 and seasonal cycles from grid cells within each region were averaged. Data from nine surface  
28 moorings were also compiled and used to develop robust estimates of mean seasonal cycles for  
29 comparison with the eight regions. This manuscript describes our methodology and the resulting  
30 mean seasonal cycles for multiple OA metrics in an effort to provide large-scale, environmental  
31 context for ongoing research, adaptation, and management efforts throughout the Pacific



32 Northwest. Major findings include the identification of unique chemical characteristics across the  
33 study domain. There is a clear increase in the ratio of dissolved inorganic carbon (DIC) to total  
34 alkalinity (TA) and in the seasonal cycle amplitude of carbonate system parameters when moving  
35 from the open ocean North Pacific into the Salish Sea. Due to the logarithmic nature of the pH  
36 scale ( $\text{pH} = -\log_{10}[\text{H}^+]$ , where  $[\text{H}^+]$  is the hydrogen ion concentration), lower annual mean pH  
37 values (associated with elevated DIC:TA) coupled with larger magnitude seasonal pH cycles  
38 results in seasonal  $[\text{H}^+]$  ranges that are  $\sim 27$  times larger in Hood Canal than in the neighboring  
39 North Pacific open ocean. Organisms living in the Salish Sea are thus exposed to much larger  
40 seasonal acidity changes than those living in nearby open ocean waters. Additionally, our findings  
41 suggest that lower buffering capacities in the Salish Sea make these waters less efficient at  
42 absorbing anthropogenic carbon than open ocean waters at the same latitude.

43

44 All data used in this analysis are publically available at the following websites:

- 45 • Surface Ocean CO<sub>2</sub> Atlas Version 4 coastal data,  
46 <https://doi.pangaea.de/10.1594/PANGAEA.866856>;
- 47 • National Oceanic and Atmospheric Administration (NOAA) West Coast Ocean Acidification  
48 cruise data, [10.3334/CDIAC/otg.CLIVAR\\_NACP\\_West\\_Coast\\_Cruise\\_2007](https://doi.org/10.3334/CDIAC/otg.CLIVAR_NACP_West_Coast_Cruise_2007);  
49 [10.3334/CDIAC/OTG.COAST\\_WCOA2011](https://doi.org/10.3334/CDIAC/OTG.COAST_WCOA2011); [10.3334/CDIAC/OTG.COAST\\_WCOA2012](https://doi.org/10.3334/CDIAC/OTG.COAST_WCOA2012);  
50 [10.7289/V5C53HXP](https://doi.org/10.7289/V5C53HXP);
- 51 • University of Washington (UW) and Washington Ocean Acidification Center cruise data,  
52 <https://doi.org/10.5281/zenodo.1184657>;
- 53 • Washington State Department of Ecology seaplane data,  
54 <https://doi.org/10.5281/zenodo.1184657>;
- 55 • NOAA Moored Autonomous pCO<sub>2</sub> (MAPCO<sub>2</sub>) Buoy data,  
56 [https://doi.org/10.3334/CDIAC/OTG.TSM\\_LAPUSH\\_125W\\_48N](https://doi.org/10.3334/CDIAC/OTG.TSM_LAPUSH_125W_48N);  
57 [https://doi.org/10.3334/CDIAC/OTG.TSM\\_WA\\_125W\\_47N](https://doi.org/10.3334/CDIAC/OTG.TSM_WA_125W_47N);  
58 [https://doi.org/10.3334/CDIAC/OTG.TSM\\_DABOB\\_122W\\_478N](https://doi.org/10.3334/CDIAC/OTG.TSM_DABOB_122W_478N);  
59 [https://doi.org/10.3334/CDIAC/OTG.TSM\\_TWANOH\\_123W\\_47N](https://doi.org/10.3334/CDIAC/OTG.TSM_TWANOH_123W_47N);
- 60 • UW Oceanic Remote Chemical/Optical Analyzer Buoy data,  
61 <https://doi.org/10.5281/zenodo.1184657>;
- 62 • NOAA Pacific Coast Ocean Observing System cruise data,  
63 <https://doi.org/10.5281/zenodo.1184657>.

64

## 65 1. Introduction

66 Changes in seawater chemistry caused by the continuous absorption of anthropogenic carbon  
67 dioxide (CO<sub>2</sub>) from the atmosphere are altering the marine environment in ways that are often  
68 invisible to the human eye but may resonate throughout coastal communities in the form of altered



69 ecosystem health and economic vulnerabilities associated with food security (e.g., Adelman and  
70 Binder, 2012; Barton et al., 2015; Ekstrom et al., 2015; Somero et al., 2016; Wong et al., 2014).  
71 Rising CO<sub>2</sub> partial pressures ( $p\text{CO}_2$ ) and hydrogen ion concentrations ( $[\text{H}^+]$ ), commonly referred  
72 to as “Ocean Acidification” (OA; Caldeira and Wickett, 2003; Doney et al., 2009), have been  
73 observed throughout the global open ocean (e.g., Bates et al., 2014; Brewer, 1978; Feely et al.,  
74 2004, 2009; Sabine, 2004). Yet, in the coastal zone it remains challenging to attribute carbonate  
75 chemistry changes to anthropogenic factors due to the much larger and more sporadic natural  
76 chemical variations that take place near shore (Borges, 2011; Evans et al., 2011; Fassbender et al.,  
77 2016; Feely et al., 2016; Harris, 2013; Takeshita et al., 2015; Waldbusser and Salisbury, 2014).  
78 Unlike the open ocean, the coastal zone is complicated by land-sea interactions (e.g., river input,  
79 upwelling, point source pollution, as well as tidal and estuarine mixing) that influence water and  
80 chemical residence times as well as carbon transformation processes, and these may also be  
81 changing over time due to human activities (Bauer et al., 2013; Cai et al., 2011; Feely et al., 2016;  
82 Raymond et al., 2008; Raymond and Cole, 2003; Regnier et al., 2013). Baseline observations  
83 capturing the range of modern variability in the coastal ocean will therefore be required to  
84 accurately fingerprint the ongoing carbonate system changes associated with human perturbations  
85 to the global climate system.

86

87 Washington State has an expansive coastline where a diversity of land-sea interfaces results in  
88 regional patchiness, requiring local assessments of OA that cannot be inferred from the broader  
89 North Pacific Ocean (e.g., Feely et al., 2010). Monthly pH observations have been collected  
90 throughout Washington waters since 1989 by the Washington State Department of Ecology (ECY)  
91 using electrode-based pH measurements with known accuracy and precision limitations. The use  
92 of a consistent method has allowed ECY to perform intensive quality assurance (Bos et al., 2016),  
93 leading to the detection of interannual variability and secular trends in pH over the data record.  
94 Still, great uncertainty remains in how other carbonate system parameters have changed  
95 throughout the region. In 2006, the National Oceanic and Atmospheric Administration (NOAA)  
96 and the University of Washington (UW) began sustained high-frequency (3-hour) observations of  
97 carbonate chemistry in Washington waters with the deployment of an autonomous CO<sub>2</sub> sensor on  
98 a surface mooring. Three additional surface moorings have been outfitted with CO<sub>2</sub> sensors since  
99 2009 (Alin et al., 2015). Routine discrete sampling of two carbonate system parameters conducted



100 by NOAA and regional partners started in 2007 (Feely et al., 2008, 2010, 2016), and the  
101 deployment of autonomous pH sensors on regional surface moorings was initiated by the UW and  
102 NOAA in 2010. In addition to these targeted carbon observations, CO<sub>2</sub> measurements collected on  
103 ships of opportunity while they are underway have amassed throughout the region since the 1970s  
104 (Bakker et al., 2016). Altogether these data, in addition to measurements made by a large  
105 community of agencies, institutions, and stakeholders, constitute an impressive network of  
106 observations that makes Washington one of the most data-rich states to tackle research questions  
107 associated with OA in the coastal zone (Alin et al., 2015).

108

109 Building upon these observing efforts, we have characterized the average seasonal cycles of  
110 numerous carbonate system parameters throughout Pacific Northwest marine surface waters.  
111 Comprised of publicly-available, high-quality data, the resulting data products are meant to  
112 provide large-scale environmental context for OA research (Andersson and MacKenzie, 2012;  
113 Hofmann et al., 2011; McElhany and Busch, 2013; Takeshita et al., 2015; Wahl et al., 2016),  
114 adaptation and management strategy development (Boehm et al., 2015; Ekstrom et al., 2015), and  
115 water quality assessment (Bednaršek et al., 2017; Weisberg et al., 2016). Here we describe our  
116 methodology and the resulting carbonate chemistry seasonal cycles, reproduced in the netCDF  
117 format as supplementary material to this article (contact the author for similar information in the  
118 MATLAB format).

119

## 120 2. Data sources

### 121 2.1. Surface Ocean CO<sub>2</sub> Atlas version 4

122 The Surface Ocean CO<sub>2</sub> Atlas (SOCAT; <http://www.socat.info/>) is the result of a data synthesis  
123 activity carried out by >100 members of the carbon research community who compile and quality  
124 control sea surface (upper ~5 m) measurements of CO<sub>2</sub> fugacity (*f*CO<sub>2</sub>) made across the globe  
125 (Bakker et al., 2016; Pfeil et al., 2013; Sabine et al., 2013), where *f*CO<sub>2</sub> is similar to *p*CO<sub>2</sub> but takes  
126 into account the non-ideal nature of the gas (Dickson et al., 2007). The SOCAT database is updated  
127 annually to include the most recent observations as well as historical data files that have been  
128 discovered and validated. For this analysis, SOCAT version 4 (SOCAT-v4) *f*CO<sub>2</sub> data collected  
129 within 45.5° N to 49° N and 127° W to 122° W (**Fig. 1a**) were downloaded from the coastal  
130 SOCAT database (**Table 1**) along with the accompanying sea surface salinity (SSS) and sea



131 surface temperature (SST) observations. World Ocean Circulation Experiment quality control  
132 flags of 2 (“good”) and SOCAT metadata flags of A through D were applied such that the  $f\text{CO}_2$   
133 observations included have an accuracy of 5  $\mu\text{atm}$  or better (Bakker et al., 2016). Mooring  
134 observations found within the study region were removed from the SOCAT-v4 dataset and are  
135 treated separately. The resultant dataset includes ~57,000 quality-controlled  $f\text{CO}_2$  observations  
136 collected between 1976 and 2015 (**Table 2**). Because these observations span four decades, a  
137 seawater anthropogenic  $p\text{CO}_2$  trend of 1.5  $\mu\text{atm yr}^{-1}$  was used to normalize the  $f\text{CO}_2$  observations  
138 to the reference year 2010, following *Takahashi et al.*, [2009, 2014]. Although a trend in  $p\text{CO}_2$   
139 rather than  $f\text{CO}_2$  was used, the difference in trend between the two parameters is negligible over a  
140 40-year period ( $\sim 0.006 \mu\text{atm yr}^{-1}$ ), resulting in a 0.2  $\mu\text{atm}$  absolute difference between parameters.  
141

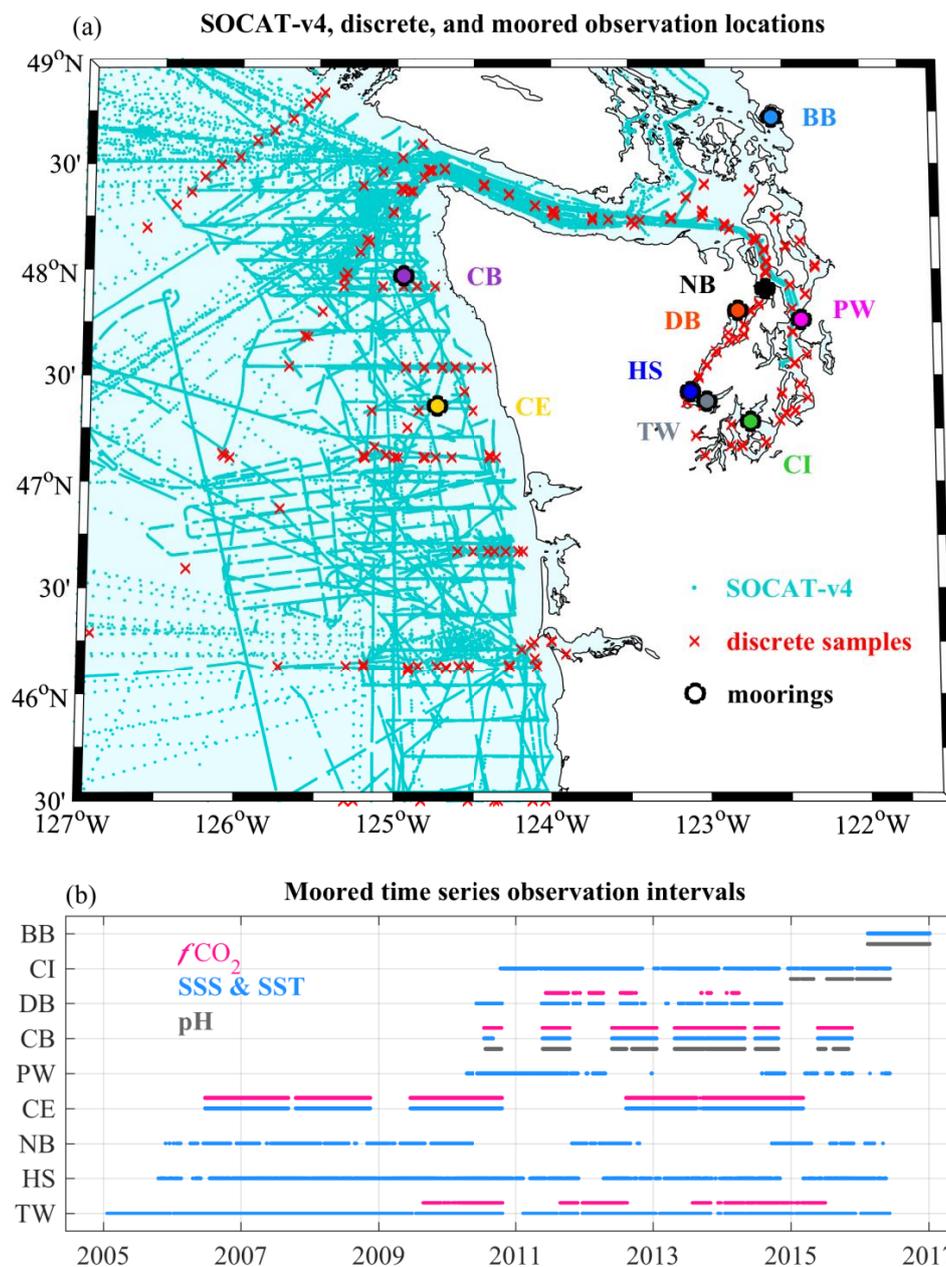
## 142 **2.2. Discrete carbon data**

143 Discrete samples collected from the top 10 m of water during recurring oceanographic cruises  
144 conducted between May 2007 and May 2015 near Washington State were compiled for this  
145 analysis (**Fig. 1a**). These cruises include: NOAA Ocean Acidification Program (OAP) West Coast  
146 Ocean Acidification (WCOA) cruises; Pacific Coast Ocean Observing System (PacOOS) cruises;  
147 and UW cruises associated with the Puget Sound Regional Synthesis Model (PRISM) program,  
148 Washington Ocean Acidification Center (WOAC), and Northwest Association of Networked  
149 Ocean Observing Systems (NANOOS) as well as its Chá bá mooring. Additionally, ECY collected  
150 discrete water samples via seaplane from ~5 m depth during 2014 and 2015 (Keyzers, 2014, 2016).  
151 Each data subset used herein can be accessed online using information from **Table 1**.  
152

153 Discrete samples with quality control flags of 2 or 6 (2 = good data, 6 = replicate samples)  
154 were included, yielding ~900 observations. **Table 2** provides a detailed list of the cruises, number  
155 of dissolved inorganic carbon (DIC), total alkalinity (TA), total pH ( $\text{pH}_T$ ),  $f\text{CO}_2$ , salinity (S), and  
156 temperature (T) observations relied on from each cruise. Information about the silicate and  
157 phosphate observations that were also used in this analysis can be found in **Appendix A**. All of  
158 the DIC and TA bottle samples were analyzed at the Pacific Marine Environmental Laboratory via  
159 coulometric (Dickson et al., 2007; Johnson et al., 1998) and potentiometric titration (Dickson et  
160 al., 2007; Millero et al., 1993), respectively. For all cruises, the accuracy of DIC and TA relative  
161 to Certified Reference Materials (Dickson et al., 2007) is  $\pm 0.1$  % of the measurement value and



162 the precision is  $< \pm 0.1$  % for TA and  $\sim 1 \mu\text{mol kg}^{-1}$  for DIC. This is equal to an accuracy of  
163 approximately  $2 \mu\text{mol kg}^{-1}$  for both parameters in Washington surface waters. For simplicity, we  
164 use  $2 \mu\text{mol kg}^{-1}$  as the total uncertainty for DIC and TA samples and discuss the role of  
165 measurement error further in Sect. 4.2.  $\text{pH}_T$  measurements were also made during the 2011 and  
166 2013 WCOA cruises (**Table 2**). These samples were analyzed by spectrophotometry (Byrne et al.,  
167 2010; Liu et al., 2011) and have measurement accuracies of 0.004.  
168



169

170 **Figure 1:** (a) Locations of coastal SOCAT-v4  $f\text{CO}_2$  observations, discrete sample stations, and  
 171 moorings. The moorings are labeled with two-letter acronyms as follows: Bellingham Bay (BB),  
 172 Carr Inlet (CI), Dabob Bay (DB), Chá bã (CB), Point Wells (PW), Cape Elizabeth (CE), North  
 173 Buoy (NB), Hoodsport (HS), and Twanoth (TW). (b) Intervals of available sensor data from each  
 174 of the nine moorings. All moorings include salinity (SSS) and temperature (SST) observations  
 175 (blue). Some moorings also include pH (gray) and/or  $f\text{CO}_2$  (pink) observations.



176 **Table 1.** Data attribution table listing the SOCAT and discrete datasets (with observing periods)  
 177 incorporated into the analysis. Data curating organization URLs and data DOIs are provided.  
 178

| DISCRETE DATA         | Obs. Period | Data Curating Organization URL  | Data DOI  |
|-----------------------|-------------|---|---|
| SOCAT-v4 <sup>1</sup> | 1970-2015   | <a href="https://doi.pangaea.de/10.1594/PANGAEA.866876">https://doi.pangaea.de/10.1594/PANGAEA.866876</a>   | <a href="https://doi.pangaea.de/10.1594/PANGAEA.866856">https://doi.pangaea.de/10.1594/PANGAEA.866856</a>   |
| NOAA WCOA             | 05/2007     | <a href="https://www.nodc.noaa.gov/ocads/data/0083685.xml">https://www.nodc.noaa.gov/ocads/data/0083685.xml</a>                                     | <a href="https://doi.org/10.3334/CDIAC/otg.CLIVAR_NACP_West_Coast_Cruise_2007">https://doi.org/10.3334/CDIAC/otg.CLIVAR_NACP_West_Coast_Cruise_2007</a> |
| UW PRISM*             | 02/2008     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW PRISM/EPA*         | 08/2008     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| PacOOS*               | 08/2009     |   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW PRISM*             | 09/2009     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| PacOOS*               | 05/2010     |   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| PacOOS*               | 08/2010     |   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW PRISM*             | 11/2010     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW/Chá bã*            | 05/2011     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| NOAA WCOA             | 08/2011     | <a href="https://www.nodc.noaa.gov/ocads/data/0157458.xml">https://www.nodc.noaa.gov/ocads/data/0157458.xml</a>                                     | <a href="https://doi.org/10.3334/CDIAC/OTG.COAST_WCOA2_011">https://doi.org/10.3334/CDIAC/OTG.COAST_WCOA2_011</a>                                       |
| UW PRISM*             | 10/2011     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW/NANOOS*            | 05/2012     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| NOAA WCOA             | 09/2012     | <a href="https://www.nodc.noaa.gov/ocads/data/0157445.xml">https://www.nodc.noaa.gov/ocads/data/0157445.xml</a>                                     | <a href="https://doi.org/10.3334/CDIAC/OTG.COAST_WCOA2_012">https://doi.org/10.3334/CDIAC/OTG.COAST_WCOA2_012</a>                                       |
| UW/Chá bã*            | 01/2013     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW/NANOOS*            | 04/2013     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| NOAA WCOA             | 08/2013     | <a href="https://www.nodc.noaa.gov/oceanacidification/data/0132_082.xml">https://www.nodc.noaa.gov/oceanacidification/data/0132_082.xml</a>         | <a href="https://doi.org/10.7289/V5C53HXP">10.7289/V5C53HXP</a>   |
| UW/NANOOS*            | 09/2013     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW/NANOOS*            | 06/2014     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| ECY                   | 06/2014     | <a href="https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html">https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| WOAC*                 | 07/2014     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| ECY                   | 07/2014     | <a href="https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html">https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| ECY                   | 08/2014     | <a href="https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html">https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| WOAC*                 | 09/2014     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| ECY                   | 09/2014     | <a href="https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html">https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW/Chá bã*            | 10/2014     | <a href="http://nvs.nanoos.org/CruiseSalish">http://nvs.nanoos.org/CruiseSalish</a>   | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| ECY                   | 10/2014     | <a href="https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html">https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| ECY                   | 11/2014     | <a href="https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html">https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| ECY                   | 01/2015     | <a href="https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html">https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| ECY                   | 02/2015     | <a href="https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html">https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| ECY                   | 03/2015     | <a href="https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html">https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| ECY                   | 04/2015     | <a href="https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html">https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| ECY                   | 05/2015     | <a href="https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html">https://fortress.wa.gov/ecy/publications/SummaryPages/1603032.html</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |

179 <sup>1</sup>The coastal subset of SOCAT-v4 data can be easily accessed here: <https://www.socat.info/index.php/version-4/>

180 \*Full dataset in preparation for submission to a long-term data repository.



182 **Table 2.** SOCAT and discrete datasets incorporated into the analysis. The observing period and  
 183 the number of  $f\text{CO}_2$ , DIC, TA, pH, S, and T values that were observed (Obs.), calculated (Calc.),  
 184 and estimated (Est.) for each cruise are provided.  
 185

| DISCRETE<br>DATA | Obs.<br>Period | $f\text{CO}_2$ |            | DIC        |               | TA         |               | pH <sub>T</sub> |               | S             | T             |
|------------------|----------------|----------------|------------|------------|---------------|------------|---------------|-----------------|---------------|---------------|---------------|
|                  |                | Obs.           | Calc.      | Obs.       | Calc.         | Obs.       | Est.          | Obs.            | Calc.         | Obs.          | Obs.          |
| SOCAT-v4         | 1976-2015      | 57,425         | 0          | 0          | 52,521        | 0          | 52,524        | 0               | 52,521        | 52,528        | 57,422        |
| NOAA WCOA        | 05/2007        | 0              | 11         | 11         | 0             | 11         | 0             | 0               | 11            | 11            | 11            |
| UW PRISM         | 02/2008        | 0              | 80         | 80         | 0             | 75         | 5             | 0               | 80            | 80            | 80            |
| UW PRISM/EPA     | 08/2008        | 0              | 122        | 122        | 0             | 120        | 2             | 0               | 122           | 122           | 122           |
| PacOOS           | 08/2009        | 0              | 8          | 8          | 0             | 8          | 0             | 0               | 8             | 8             | 8             |
| UW PRISM         | 09/2009        | 0              | 38         | 38         | 0             | 38         | 0             | 0               | 38            | 38            | 38            |
| PacOOS           | 05/2010        | 0              | 13         | 13         | 0             | 13         | 0             | 0               | 13            | 13            | 13            |
| PacOOS           | 08/2010        | 0              | 22         | 22         | 0             | 22         | 0             | 0               | 22            | 22            | 22            |
| UW PRISM         | 11/2010        | 0              | 40         | 40         | 0             | 37         | 3             | 0               | 40            | 40            | 40            |
| UW/Chá bã        | 05/2011        | 0              | 1          | 1          | 0             | 1          | 0             | 0               | 1             | 1             | 1             |
| NOAA WCOA        | 08/2011        | 0              | 37         | 37         | 0             | 32         | 5             | 32              | 5             | 37            | 37            |
| UW PRISM         | 10/2011        | 0              | 60         | 60         | 0             | 60         | 0             | 0               | 60            | 60            | 60            |
| UW/NANOOS        | 05/2012        | 0              | 4          | 4          | 0             | 4          | 0             | 0               | 4             | 4             | 4             |
| NOAA WCOA        | 09/2012        | 0              | 43         | 43         | 0             | 39         | 3             | 0               | 43            | 43            | 43            |
| UW/Chá bã        | 01/2013        | 0              | 2          | 2          | 0             | 2          | 0             | 0               | 2             | 2             | 2             |
| UW/NANOOS        | 04/2013        | 0              | 19         | 19         | 0             | 18         | 1             | 0               | 19            | 19            | 19            |
| NOAA WCOA        | 08/2013        | 0              | 59         | 59         | 0             | 47         | 9             | 45              | 14            | 59            | 59            |
| UW/NANOOS        | 09/2013        | 0              | 19         | 19         | 0             | 19         | 0             | 0               | 19            | 19            | 19            |
| UW/NANOOS        | 06/2014        | 0              | 10         | 10         | 0             | 10         | 0             | 0               | 10            | 10            | 10            |
| ECY              | 06/2014        | 0              | 17         | 17         | 0             | 17         | 0             | 0               | 17            | 17            | 17            |
| WOAC             | 07/2014        | 0              | 68         | 68         | 0             | 68         | 0             | 0               | 68            | 68            | 68            |
| ECY              | 07/2014        | 0              | 12         | 12         | 0             | 12         | 0             | 0               | 12            | 12            | 12            |
| ECY              | 08/2014        | 0              | 18         | 18         | 0             | 18         | 0             | 0               | 18            | 18            | 18            |
| WOAC             | 09/2014        | 0              | 64         | 64         | 0             | 64         | 0             | 0               | 64            | 64            | 64            |
| ECY              | 09/2014        | 0              | 14         | 14         | 0             | 14         | 0             | 0               | 14            | 14            | 14            |
| UW/Chá bã        | 10/2014        | 0              | 19         | 19         | 0             | 19         | 0             | 0               | 19            | 19            | 19            |
| ECY              | 10/2014        | 0              | 9          | 9          | 0             | 9          | 0             | 0               | 9             | 9             | 9             |
| ECY              | 11/2014        | 0              | 9          | 9          | 0             | 9          | 0             | 0               | 9             | 9             | 9             |
| ECY              | 01/2015        | 0              | 13         | 13         | 0             | 13         | 0             | 0               | 13            | 13            | 13            |
| ECY              | 02/2015        | 0              | 15         | 15         | 0             | 15         | 0             | 0               | 15            | 15            | 15            |
| ECY              | 03/2015        | 0              | 20         | 20         | 0             | 20         | 0             | 0               | 20            | 20            | 20            |
| ECY              | 04/2015        | 0              | 15         | 15         | 0             | 15         | 0             | 0               | 15            | 15            | 15            |
| ECY              | 05/2015        | 0              | 6          | 6          | 0             | 6          | 0             | 0               | 6             | 6             | 6             |
| <b>TOTAL</b>     |                | <b>57,425</b>  | <b>887</b> | <b>887</b> | <b>52,521</b> | <b>855</b> | <b>52,552</b> | <b>77</b>       | <b>53,331</b> | <b>53,415</b> | <b>58,309</b> |

186

187 **2.3. Mooring carbon data**

188 In addition to discrete data, time-series observations from nine surface moorings in the region  
 189 are included in the analysis (**Fig. 1a**). These moorings are maintained by NOAA, UW, WOAC,  
 190 and NANOOS. Moored time-series data can be accessed online using information from **Table 3**.  
 191 All of the moorings carry sensors that measure SST and SSS, and most include sensors for surface



192 ocean  $f\text{CO}_2$  or  $\text{pH}_T$ , while the Chá bã mooring measures both of these carbon parameters (**Fig. 1b**).  
193 Presently, quality-controlled observations from the moorings range from January 2005 through  
194 January 2017, with the longest carbon record from the Cape Elizabeth mooring that started in 2006.  
195 Assuming the same anthropogenic  $f\text{CO}_2$  trend of  $1.5 \mu\text{atm yr}^{-1}$  that was used to normalize the  
196 SOCAT-v4 data (Takahashi *et al.*, 2009, 2014), the Cape Elizabeth time series may include a  $\sim 12$   
197  $\mu\text{atm}$  anthropogenic increase in the mean  $f\text{CO}_2$  over the 8-year record. This is much smaller than  
198 the potential  $\sim 60 \mu\text{atm}$  increase over the  $\sim 40$  year SOCAT-v4 record. Additionally, since all other  
199 moorings have much shorter carbon time series (some with pH rather than  $f\text{CO}_2$  records), we do  
200 not remove the anthropogenic  $f\text{CO}_2$  trend from the mooring data.

201

202 The Chá bã, Cape Elizabeth, Dabob Bay, and Twanoh surface moorings each carry a Moored  
203 Autonomous  $p\text{CO}_2$  (MAPCO2) system that makes 3-hour measurements of surface air and sea  
204  $p\text{CO}_2$  with an accuracy of  $\leq 2 \mu\text{atm}$  (Sutton *et al.*, 2014). The Chá bã mooring is also outfitted with  
205 a Sunburst SAMI pH sensor with reported field accuracy of 0.018, and a Sea-Bird Electronics  
206 (SBE) 16plus V2 Sea-CAT conductivity-temperature-depth (CTD) sensor with SST accuracy  $<$   
207 0.01 and SSS accuracy  $< 0.05$ , as described in Sutton *et al.* (2016). The Cape Elizabeth mooring  
208 carries an SBE 37 MicroCAT CTD sensor with SST accuracy  $< 0.01$  and SSS accuracy  $< 0.05$ .  
209 These sensors are programed to sample every 3 hours in tandem with the MAPCO2 system. The  
210 other seven moorings, Oceanic Remote Chemical/Optical Analyzer (ORCA) buoys (Dunne *et al.*,  
211 2002), are equipped with profiling sensors that include SBE19 or 19Plus CTD sensors (SST  
212 accuracy of 0.005 and SSS accuracy of 0.005), SBE 43 dissolved oxygen sensors, and WETLabs  
213 WETStar or FLNTUS chlorophyll fluorometers. Temperature and salinity observations collected  
214 from the top 3 m of the water column were binned and averaged to serve as SST and SSS for this  
215 analysis. The Carr Inlet and Bellingham Bay surface buoys also carry Satlantic SeaFET pH sensors  
216 with a reported accuracy of  $\pm 0.05$  pH. All raw SeaFET measurements were converted to pH using  
217 custom code adapted from Martz *et al.* (2015) and equations from Bresnahan *et al.* (2014). ORCA  
218 mooring data were quality controlled according to protocols outlined in the Quality Assurance  
219 Project Plan ([http://nwem.ocean.washington.edu/ORCA\\_QAPP.pdf](http://nwem.ocean.washington.edu/ORCA_QAPP.pdf); Newton and Devol, 2012).  
220 All sensors on the surface buoys sample the water column at  $\sim 1$  m depth, excluding the profiling  
221 CTDs. Details about the number of observations collected at each mooring location are provided  
222 in **Table 4**. It is important to note that many of the moored time series include significant data gaps



223 and/or times when  $f\text{CO}_2$  and  $\text{pH}_T$  measurements are not available (**Fig. 1b**). The observing periods  
 224 listed in **Table 3** represent the time range over which quality-controlled, *in situ* observations exist  
 225 at the mooring site at the time of this manuscript.

226

227 **Table 3.** Data attribution table listing the mooring datasets (with observing periods) incorporated  
 228 into the analysis. Data curating organization URLs and data DOIs are provided.

229

| MOORING DATA        | Obs. Period | Data Curating Organization URL  | Data DOI  |
|---------------------|-------------|---|---|
| Chá bã <sup>1</sup> | 07/10-10/15 | <a href="https://www.nodc.noaa.gov/ocads/data/010007_2.xml">https://www.nodc.noaa.gov/ocads/data/010007_2.xml</a>       | <a href="https://doi.org/10.3334/CDIAC/OTG.TSM_LAPUSH_1_25W_48N">https://doi.org/10.3334/CDIAC/OTG.TSM_LAPUSH_1_25W_48N</a> |
| Cape Elizabeth      | 06/06-09/13 | <a href="https://www.nodc.noaa.gov/ocads/data/011532_2.xml">https://www.nodc.noaa.gov/ocads/data/011532_2.xml</a>       | <a href="https://doi.org/10.3334/CDIAC/OTG.TSM_WA_125W_47N">https://doi.org/10.3334/CDIAC/OTG.TSM_WA_125W_47N</a>           |
| Dabob Bay*          | 06/10-11/14 | <a href="https://www.nodc.noaa.gov/ocads/data/011671_5.xml">https://www.nodc.noaa.gov/ocads/data/011671_5.xml</a>       | <a href="https://doi.org/10.3334/CDIAC/OTG.TSM_DABOB_12_2W_478N">https://doi.org/10.3334/CDIAC/OTG.TSM_DABOB_12_2W_478N</a> |
| Twanoh*             | 01/05-06/16 | <a href="https://www.nodc.noaa.gov/ocads/data/015760_0.xml">https://www.nodc.noaa.gov/ocads/data/015760_0.xml</a>       | <a href="https://doi.org/10.3334/CDIAC/OTG.TSM_TWANOH_123W_47N">https://doi.org/10.3334/CDIAC/OTG.TSM_TWANOH_123W_47N</a>   |
| Carr Inlet          | 10/10-06/16 | <a href="http://nwem.ocean.washington.edu/prod_Data_Req.shtml">http://nwem.ocean.washington.edu/prod_Data_Req.shtml</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>                                 |
| Bellingham Bay      | 02/16-01/17 | <a href="http://nwem.ocean.washington.edu/prod_Data_Req.shtml">http://nwem.ocean.washington.edu/prod_Data_Req.shtml</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>                                 |
| Hoodspport          | 10/05-05/16 | <a href="http://nwem.ocean.washington.edu/prod_Data_Req.shtml">http://nwem.ocean.washington.edu/prod_Data_Req.shtml</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>                                 |
| Point Wells         | 04/10-06/16 | <a href="http://nwem.ocean.washington.edu/prod_Data_Req.shtml">http://nwem.ocean.washington.edu/prod_Data_Req.shtml</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>                                 |
| North Buoy          | 11/05-05/16 | <a href="http://nwem.ocean.washington.edu/prod_Data_Req.shtml">http://nwem.ocean.washington.edu/prod_Data_Req.shtml</a> | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>                                 |

230

<sup>1</sup>The Chá bã mooring is referred to as the La Push mooring in the NOAA data repository.

231

\*Temperature and salinity data for the Dabob Bay and Twanoh moorings can be found here:

232

<https://doi.org/10.5281/zenodo.1184657>

233

234

**Table 4.** Mooring datasets incorporated into the analysis. The observing period and number of  
 235  $f\text{CO}_2$ , DIC, TA, pH, S, and T values that were observed (Obs.), calculated (Calc.), and estimated  
 236 (Est.) are provided.

237

| MOORING DATA   | Obs. Period | $f\text{CO}_2$ |              | DIC      |               | TA       |                | $\text{pH}_T$ |               | S              | T              |
|----------------|-------------|----------------|--------------|----------|---------------|----------|----------------|---------------|---------------|----------------|----------------|
|                |             | Obs.           | Calc.        | Obs.     | Calc.         | Obs.     | Est.           | Obs.          | Calc.         | Obs.           | Obs.           |
| Chá bã         | 07/10-11/15 | 9,681          | 0            | 0        | 9,330         | 0        | 9,341          | 7,438         | 2,291         | 9,341          | 9,717          |
| Cape Elizabeth | 06/06-03/15 | 17,617         | 0            | 0        | 17,617        | 0        | 17,667         | 0             | 17,617        | 17,667         | 18,009         |
| Dabob Bay      | 06/10-11/14 | 2,076          | 0            | 0        | 2,076         | 0        | 4,662          | 0             | 2,076         | 4,662          | 4,662          |
| Twanoh         | 01/05-06/16 | 9,221          | 0            | 0        | 9,221         | 0        | 28,466         | 0             | 9,221         | 28,466         | 28,466         |
| Carr Inlet     | 10/10-06/16 | 0              | 1,848        | 0        | 1,848         | 0        | 12,369         | 1,848         | 0             | 12,369         | 12,369         |
| Bellingham Bay | 02/16-01/17 | 0              | 0            | 0        | 0             | 0        | 0              | 45,583        | 0             | 44,895         | 44,895         |
| Hoodspport     | 10/05-05/16 | 0              | 0            | 0        | 0             | 0        | 17,147         | 0             | 0             | 17,186         | 17,186         |
| Point Wells    | 04/10-06/16 | 0              | 0            | 0        | 0             | 0        | 4,315          | 0             | 0             | 4,315          | 4,315          |
| North Buoy     | 11/05-05/16 | 0              | 0            | 0        | 0             | 0        | 10,199         | 0             | 0             | 10,199         | 10,199         |
| <b>TOTAL</b>   |             | <b>38,595</b>  | <b>1,848</b> | <b>0</b> | <b>40,092</b> | <b>0</b> | <b>104,166</b> | <b>54,869</b> | <b>31,205</b> | <b>149,100</b> | <b>149,818</b> |

238

### 239 3. Methods

#### 240 3.1. Leveraging the carbon datasets



241 Seawater carbonate chemistry can be fully characterized with information about salinity,  
242 temperature, pressure, weak acids that contribute to TA (e.g., phosphoric and silicic acid), and two  
243 of the four commonly-measured carbonate system parameters: DIC, TA,  $p\text{CO}_2$  (or  $f\text{CO}_2$ ), and pH  
244 (Dickson and Riley, 1978; Millero, 2007). In order to leverage the numerous SOCAT-v4  $f\text{CO}_2$   
245 observations that lack a second carbonate system parameter, a previously characterized  
246 relationship between near-surface ( $\leq 25$  m) salinity and TA ( $\text{TA} = 47.7 \times \text{Salinity} + 647$ ) for marine  
247 surface waters near Washington (Fassbender et al., 2017a) was used to estimate TA from the  
248 SOCAT-v4 SSS observations. This relationship yields TA estimates that have an uncertainty  $\leq$   
249  $\pm 34 \mu\text{mol kg}^{-1}$  at the 95% confidence level ( $2\sigma$ ) for waters with salinity  $\geq 20$  (Fassbender et al.,  
250 2017). Uncertainties may be larger in the salinity range of 20 to 27 ( $2\sigma = \pm 94 \mu\text{mol kg}^{-1}$ ); however,  
251 only ~2% of the SOCAT-v4 SSS observations are below salinity 27. TA was not estimated when  
252 SOCAT-v4 SSS values were below 20 ( $n=4$ ). The estimated TA and 2010-normalized SOCAT-v4  
253  $f\text{CO}_2$  data were then used to calculate other carbonate system parameters of interest (“calculated”  
254 values in **Table 2**). All carbonate system calculations herein were made using the program  
255 CO2SYS Version 1.1 (van Heuven et al., 2011; Lewis and Wallace, 1998) with the equilibrium  
256 constants of Lueker et al. (2000) and Dickson (1990) while applying the boron-to-chlorinity ratio  
257 of Uppstrom, (1974), following the recommendations of Orr et al. (2015). SOCAT-v4 does not  
258 include nutrient observations, so all of the associated CO2SYS calculations were made with  
259 phosphate and silicate concentrations set to zero. The omission of nutrients when using the TA-  
260  $f\text{CO}_2$  pair to calculate other carbonate system parameters results in small errors. These errors were  
261 previously assessed for the TA- $p\text{CO}_2$  pair within Washington waters (Fassbender et al., 2017a),  
262 and will be discussed further in Sect. 3.2.

263

264 TA and DIC were measured during discrete sampling efforts in addition to temperature,  
265 salinity, and pressure, and these data were used to calculate other carbonate system parameters of  
266 interest. Some of the discrete sample datasets lack nutrient observations, so phosphate and silicate  
267 concentration were set to zero for the associated CO2SYS calculations. Nutrient omission when  
268 using the TA-DIC pair has a larger influence on calculated carbonate system parameters than the  
269 TA- $p\text{CO}_2$  pair (Fassbender et al., 2017), which will be addressed further in Sect. 3.2. In some  
270 cases, only DIC was measured. For these instances, TA was estimated from the previously  
271 mentioned regional TA-salinity relationship when salinity was  $\geq 20$  (true of all discrete samples),



272 making it possible to fully characterize the carbonate system as long as DIC, SST, and SSS data  
273 were available. Finally, on cruises in which  $\text{pH}_T$  was measured directly, missing  $\text{pH}_T$  observations  
274 were calculated from TA and DIC. Details about the specific number of measured, estimated, and  
275 calculated values for each parameter can be found in **Table 2**.

276

277 Sustained, autonomous  $f\text{CO}_2$  and pH measurements made on some of the surface moorings  
278 located throughout the region (**Fig. 1a**) were also used to evaluate carbonate chemistry. The TA-  
279 salinity relationship was applied to the moored SSS observations to estimate TA for pairing with  
280 the *in situ*  $f\text{CO}_2$  or pH data (**Fig. 1b**; **Table 4**), allowing calculation of other carbonate system  
281 parameters of interest. For the Chá bã mooring, where  $f\text{CO}_2$  and pH measurements have been made  
282 simultaneously since 2010,  $f\text{CO}_2$  and TA estimates were used to calculate additional carbonate  
283 system parameters due to challenges associated with constraining the carbonate system with the  
284  $f\text{CO}_2$ -pH pair (Dickson and Riley, 1978; Fassbender et al., 2015; Gray et al., 2011). At moorings  
285 limited to SSS and SST observations, TA was the only carbonate system parameter determined.  
286 Importantly, salinity at the Twanoh and Hoodspout moorings occasionally dropped below 20  
287 throughout the observational record, and salinity at the Bellingham Bay mooring was often  $< 20$ .  
288 These samples were not used to estimate TA. Because the occurrence of salinity  $< 20$  was  
289 infrequent at Twanoh and Hoodspout (1.7% and 0.2% of samples, respectively), the exclusion of  
290 these observations from seasonal cycle evaluations likely has a negligible impact. At Bellingham  
291 Bay, regular occurrences of salinity  $< 20$  (14% of samples) throughout all seasons renders the TA-  
292 salinity relationship unusable for a substantial fraction of the time series. Therefore, we do not  
293 calculate TA, or any other carbonate system parameters, and evaluate only *in situ*  $\text{pH}_T$  observations  
294 from the Bellingham Bay mooring.

295

296 For this study, the carbonate system parameters of interest (either measured, estimated, or  
297 calculated) include: TA, DIC,  $\text{pH}_T$ ,  $f\text{CO}_2$ , aragonite saturation state ( $\Omega_{\text{Ar}}$ ), and the Revelle Factor  
298 (RF).  $\Omega_{\text{Ar}}$  describes the thermodynamic stability of aragonite in solution, where aragonite is a  
299 polymorph of the mineral calcium carbonate ( $\text{CaCO}_3$ ). This stability is dependent on temperature,  
300 pressure, salinity, and the concentrations of calcium and carbonate ions (Mucci, 1983). By  
301 definition, when  $\Omega_{\text{Ar}} \geq 1$ , the mineral is thermodynamically stable. When  $\Omega_{\text{Ar}} < 1$ , the mineral is  
302 unstable and will begin to dissolve. This is relevant to the study of OA because many marine



303 calcifying organisms make their shells out of aragonite and are susceptible to dissolution when  
304 exposed to waters with low (often  $\Omega_{Ar} < 1$ , but occasionally higher) aragonite saturation states  
305 (Barton et al., 2012; Bednaršek et al., 2014; Doney et al., 2009; Feely et al., 2008). The Revelle  
306 Factor is analogous to the buffer capacity of the ocean (Broecker et al., 1979; Revelle and Suess,  
307 1957; Sundquist et al., 1979; Takahashi et al., 1980) and provides information about how the  
308 carbonate system responds to change. Formally, RF is equal to the fractional change in  $p\text{CO}_2$  (or  
309  $f\text{CO}_2$ ) divided by the fractional change in DIC resulting from a given perturbation. Lower RF  
310 values equate to more buffered systems and result in a larger DIC change per percentage increase  
311 in  $p\text{CO}_2$  due to more efficient conversion of  $\text{CO}_2$  into other molecular forms of DIC (e.g.,  
312 bicarbonate and carbonate ion), which enables additional seawater  $\text{CO}_2$  uptake. Thus, low RF  
313 regions are associated with larger anthropogenic carbon uptake through air-sea exchange relative  
314 to high RF regions (e.g., Fassbender et al., 2017b; Sabine et al., 2004).

315

### 316 **3.2. Errors associated with nutrient omission and estimating TA from salinity**

317 Silicate and phosphate contribute to TA (Dickson, 1981) and when omitted from CO2SYS  
318 carbonate system calculations can have a small but non-negligible influence on the calculated  
319 parameters. This was evaluated previously by Fassbender et al. (2017a) for many of the cruise data  
320 used herein; however, RF was not included in the analysis and the TA- $p\text{CO}_2$  pair was used rather  
321 than TA- $f\text{CO}_2$  pair. Observations from regional cruises (**Table 2**) that include complete  
322 information about TA, DIC, phosphate, silicate, salinity, and temperature were used to calculate  
323 carbonate system parameters of interest using the observed nutrient concentrations and with  
324 nutrient concentrations set to zero. The analysis was conducted with both the TA-DIC pair and the  
325 TA- $f\text{CO}_2$  pair as input parameters, where  $f\text{CO}_2$  values were initially calculated from TA and DIC  
326 using the observed nutrient concentrations. Differences between the computations made with and  
327 without nutrients are given in **Table 5**, displaying the larger influence of nutrient omission on  
328 calculations made from the TA-DIC pair than the TA- $f\text{CO}_2$  pair for pH and  $\Omega_{Ar}$ , though not for RF  
329 due to its strong dependence to the DIC-TA ratio.

330

331 In addition to missing nutrient data, there is uncertainty in the Washington TA-salinity  
332 relationship (Fassbender et al., 2017a), which characterizes a static mean condition from which  
333 deviations can occur in both space and time. Biases in the estimated TA value caused by departures



334 from the mean condition may therefore create biases in the carbonate system parameters derived  
 335 from it. This is of particular relevance in regard to leveraging the SOCAT-v4 and mooring datasets  
 336 due to the explicit use of estimated TA to constrain the carbonate system. To evaluate the  
 337 magnitude of these potential biases, the same ship data discussed in the previous paragraph were  
 338 used (with nutrient concentrations set to zero) to compute carbonate system parameters of interest  
 339 from the TA- $f\text{CO}_2$  pair after adding and subtracting the  $\pm 2\sigma$  TA-salinity regression uncertainty ( $34$   
 340  $\mu\text{mol kg}^{-1}$ ) to the TA values. Absolute differences between the resulting values and original  
 341 estimates from the TA- $f\text{CO}_2$  pair (without altering the TA) were then averaged (**Table 5**). The  
 342 errors associated with using the Washington TA-salinity relationship are similar in magnitude to  
 343 those caused by nutrient omission when using the DIC-TA pair and much larger than those  
 344 resulting from nutrient omission when using the TA-  $f\text{CO}_2$  pair, excluding for RF. These results  
 345 suggest that  $\Omega_{\text{Ar}}$  is more sensitive to the TA-salinity regression uncertainty than nutrient omission  
 346 and RF is more sensitive to nutrient omission in the TA- $f\text{CO}_2$  pair than uncertainty in TA-salinity  
 347 regression within Washington waters. Notably, most of the errors are small relative to sensor  
 348 accuracies and/or natural variability (including for  $f\text{CO}_2$  and DIC), which will be addressed in Sect.  
 349 4.2.

350

351 **Table 5.** Errors associated with calculating carbonate system parameters in the absence of nutrient  
 352 data and when using salinity-based estimates of TA for marine surface waters near Washington.  
 353 Results for the TA-DIC and TA- $f\text{CO}_2$  pairs of input parameters are shown.

354

| INPUT ↓  | OUTPUT → | pH          | $\Omega_{\text{Ar}}$ | RF         | $f\text{CO}_2$<br>( $\mu\text{atm}$ ) | DIC<br>( $\mu\text{mol kg}^{-1}$ ) |
|--|----------|-------------|----------------------|------------|---------------------------------------|------------------------------------|
| TA & DIC (w/ - w/o Nutrients)                        |          | -0.007      | -0.02                | -0.009     | 11                                    | -                                  |
| TA & $f\text{CO}_2$ (w/ - w/o Nutrients)             |          | -0.0004     | -0.002               | -0.07      | -                                     | -2                                 |
| TA & $f\text{CO}_2$ ( $\pm 2\sigma$ on Estimated TA) |          | $\pm 0.007$ | $\pm 0.05$           | $\pm 0.02$ | -                                     | $\pm 30$                           |

355

356 Finally, SOCAT-v4 and mooring SSS data used herein were not rigorously quality controlled  
 357 beyond the identification of large outliers. Because TA is estimated directly from these salinity  
 358 observations, we performed a simple evaluation of the TA-salinity regression sensitivity. Modern,  
 359 generic conductivity sensors can achieve salinity measurements with an accuracy of  $\sim 0.02$ . To be  
 360 conservative, we propagate a salinity error ten times this value ( $\pm 0.2$ ) through the regression  
 361 calculations. This yields a mean error of  $\pm 10 \mu\text{mol kg}^{-1}$  in TA, which is well within the TA  
 362 regression 95% confidence interval of  $\pm 34 \mu\text{mol kg}^{-1}$ . In order to exceed the regression uncertainty,



363 a salinity error of ~0.7, which is 35 times the presumed accuracy of 0.02, would be required. As a  
364 result, salinity measurement errors are likely secondary to uncertainties associated with the  
365 regression relationship.

366

### 367 *3.3. New data products of monthly averaged carbonate chemistry*

368 Discrete sample and SOCAT-v4 datasets (including estimated and calculated values) were  
369 combined and gridded to a  $0.1^\circ \times 0.1^\circ$  scale. From here on, this merged product will be referred to  
370 as the discrete data product. **Figure 2a** shows the total number of times that two carbon parameters  
371 were available to fully characterize the carbonate system within each  $0.1^\circ \times 0.1^\circ$  grid cell. Monthly  
372 means, unique to each year ( $mm_{yr}$ ), were calculated for each parameter then averaged over the  
373 number of years ( $n$ ) with data to get the overall mean value ( $\overline{mm}$ ) for each month of the seasonal  
374 cycle.

375

$$376 \quad \overline{mm} = \frac{\sum_{yr=1}^n mm_{yr}}{n} \quad (1)$$

377

378 The same approach was applied to the mooring data so that monthly observations from each year  
379 were weighted equally, regardless of the number of available samples. Average seasonal cycles of  
380 all parameters for each region and mooring are tabulated in **Appendices B** and **C**.

381

382 Monthly variance values, unique to each year ( $m\sigma^2_{yr}$ ), were calculated for each parameter then  
383 averaged over the number of years with data before the square root was taken to determine the  
384 mean standard deviation ( $\overline{m\sigma}$ ) for each month of the seasonal cycle.

385

$$386 \quad \overline{m\sigma} = \sqrt{\frac{\sum_{yr=1}^n m\sigma^2_{yr}}{n}} \quad (2)$$

387

388 **Fig. 3** shows monthly averaged SST data from each annual Cape Elizabeth (CE) mooring  
389 deployment ( $mm_{yr}$  values) as well as the multi-year average seasonal cycle ( $\overline{mm}$  values). Shading  
390 bounds the mean standard deviation (**Eq. 2; Fig. 3a**) and the standard deviation of monthly mean  
391 values (**Fig. 3b**), as expressed in **Eq. 3**:

392



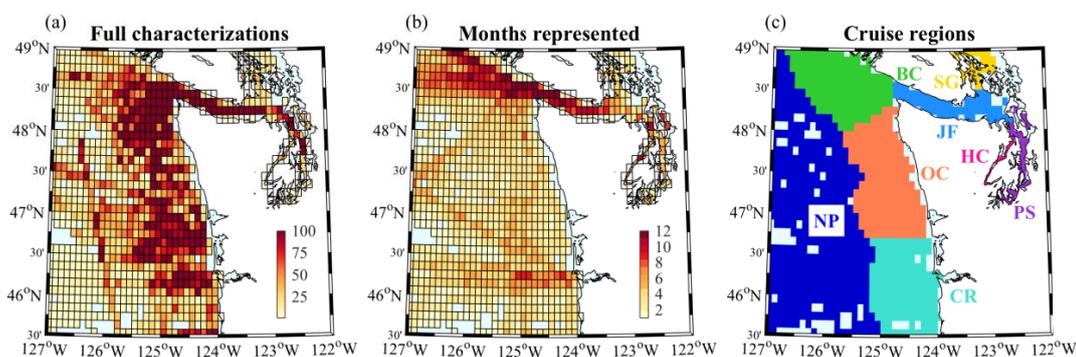
393

$$\sigma = \sqrt{\frac{\sum_{yr=1}^n (mm_{yr} - \bar{m})^2}{n-1}} \quad (3)$$

394

395 The former (**Eq. 2**) provides an estimate of the range of variability about the mean that is observed  
 396 in a given month, while the latter (**Eq. 3**) provides an estimate of interannual variability in the  
 397 mean. Both methods deliver equally useful information; however, nearly all of the moored time  
 398 series are shorter than a decade, which hinders making robust estimates of interannual variability.  
 399 Thus, standard deviations presented herein (and tabulated in **Appendix C**) reflect the average  
 400 range of variability about the mean during each month (i.e., **Eq. 2; Fig. 3a**).

401



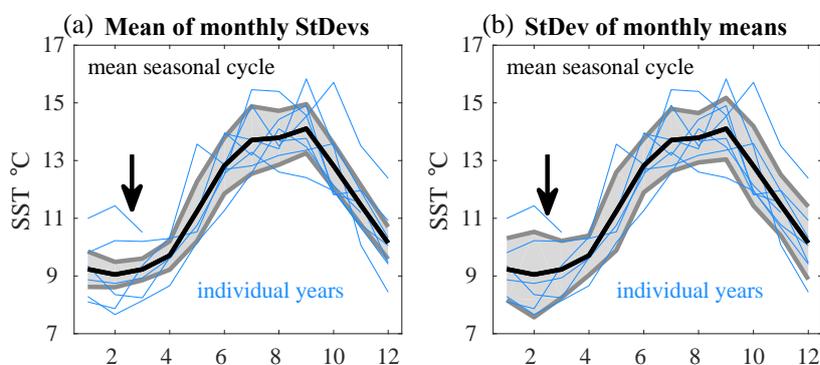
402

403 **Figure 2:** Maps showing (a) the total number of times the carbonate system was fully characterized  
 404 (two parameters available) in each grid cell, (b) the number of months represented in the seasonal  
 405 cycle for each grid cell, and (c) the eight unique regions identified, including: North Pacific (NP),  
 406 British Columbia (BC), Outer Coast (OC), Columbia River (CR), Juan de Fuca (JF), Strait of  
 407 Georgia (SG), Hood Canal (HC), and Puget Sound (PS).  
 408

409 **Figure 2b** shows the number of months represented in the seasonal cycle estimate for each  
 410 grid cell, highlighting that all 12 months are rarely resolved within a single grid cell. It is important  
 411 to note, however, that neighboring grid cells with, for example, six months of the year represented  
 412 may have observations from different months, making it possible to fully resolve the annual cycle  
 413 by combining the seasonal cycles of neighboring grid cells with similar characteristics. Relying on  
 414 this approach to accommodate data paucity, salinity and topographic features (e.g., the Juan de  
 415 Fuca Canyon and continental shelf) were used to identify eight unique regions (**Fig. 2c**) in which  
 416 seasonal cycles from individual grid cells were averaged to create regional estimates. These  
 417 regions include: North Pacific (NP), British Columbia coast (BC), Washington Outer Coast (OC),



418 Columbia River (CR), Strait of Juan de Fuca (JF), Strait of Georgia (SG), Hood Canal (HC), and  
 419 Puget Sound (PS). Importantly, some of the major physical features that result in regionally distinct  
 420 characteristics, such as the Columbia and Fraser River plumes and the Juan de Fuca Eddy, vary in  
 421 extent seasonally. The nonuniformity of available data in space and time means that these features  
 422 and their seasonal transitions may not be adequately captured in all grid cells. In addition, the lines  
 423 between regional boundaries can be blurred by interannual variability that may be inconsistently  
 424 represented in neighboring grid cells. Thus, the dynamic nature of the coastal zone should be  
 425 considered when applying the regional seasonal cycles characterized herein, which are presented  
 426 in a static-boundary framework. Due to the unequal distribution of discrete observations in space  
 427 and time, monthly mean standard deviations for the regional seasonal cycle estimates do not  
 428 provide reliable information and are not included in **Appendix B**.  
 429



430

431 **Figure 3.** Monthly averaged SST values for each year (2006-2015) of Cape Elizabeth mooring  
 432 data ( $\text{mm}_{\text{yr}}$ ; blue lines) and the average seasonal cycle of SST across all deployments ( $\overline{\text{mm}}$ ; black  
 433 line). Blue and black lines are the same in plots **a** and **b**. Gray shading represents **(a)** the mean  
 434 monthly standard deviation ( $\overline{\text{m}\sigma}$ ) and **(b)** the standard deviation of monthly means ( $\sigma$ ). Black  
 435 arrows point to a region of notable discrepancy between computations. Here, the average sub-  
 436 monthly SST variability **(a)** is much smaller than the range of interannual SST variability **(b)**,  
 437 indicating that organisms could experience distinct (i.e., non-overlapping) SST conditions during  
 438 two subsequent Februaries.

439

## 440 4. Results and discussion

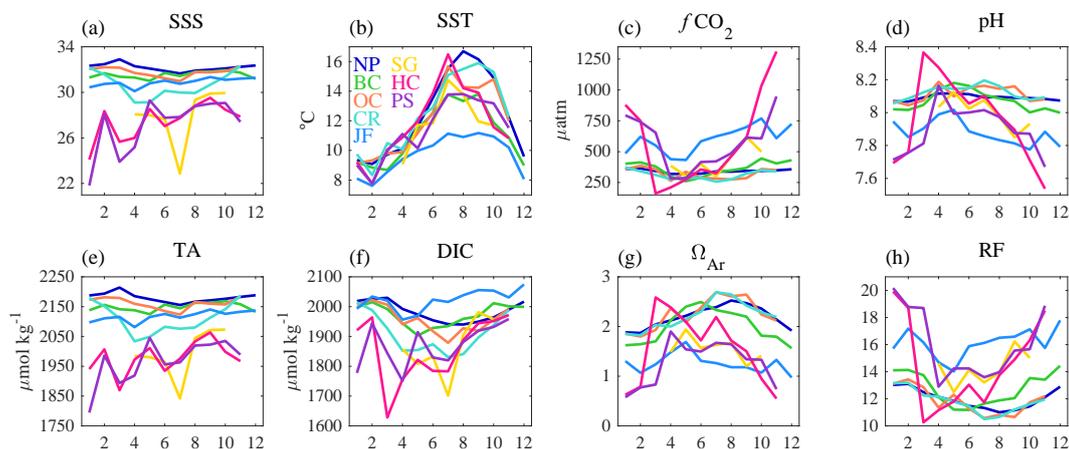
### 441 4.1. Modern seasonal cycles

442 Seasonal cycles for the eight regions, derived using measurements from the top 10 m of the  
 443 water column, are shown in **Fig. 4**. A clear pattern of salinity decrease is observed moving from  
 444 the offshore North Pacific region toward land where rivers contribute freshwater to the marine



445 environment. In particular, the Columbia and Fraser River outflows are evident in the seasonal  
446 cycles for the Columbia River and Strait of Georgia regions, with other small rivers exerting a  
447 strong influence on the semi-enclosed Hood Canal and Puget Sound domains. SST seasonal cycles  
448 are similar in structure across regions, excluding the Strait of Juan de Fuca where vigorous mixing  
449 driven by bottom topography results in much cooler surface temperatures than neighboring areas  
450 throughout the year (Alford and MacCready, 2014; Martin and MacCready, 2011). The mean  
451 seasonal cycles for  $f\text{CO}_2$  and pH are approximate mirror images within each region as a result of  
452 the strong negative correlation between these two parameters (Dickson and Riley, 1978). Annual  
453 mean  $f\text{CO}_2$  values increase and pH values decline when moving from the offshore North Pacific  
454 toward the Salish Sea, and the seasonal cycle amplitudes increase for both parameters. In all  
455 regions, the lowest  $f\text{CO}_2$  values emerge in summer and the highest values in winter, which opposes  
456 expectations associated with temperature-driven solubility changes (Takahashi et al., 1993). This  
457 suggests that physical and biological effects dominate over solubility changes in all regions  
458 (Pelletier et al., 2018), though to varying extents. Due to the logarithmic nature of the pH scale,  
459 declines in the annual mean pH coupled with increases in the seasonal range of pH values means  
460 that organisms in the Salish Sea (Puget Sound, Hood Canal, Strait of Juan de Fuca, and Strait of  
461 Georgia) are exposed to extremely large changes in  $[\text{H}^+]$  on seasonal time scales. For example, the  
462 seasonal range of  $[\text{H}^+]$  in Hood Canal is  $\sim 27$  times larger than in the North Pacific region, which  
463 indicates that organisms are living in starkly different chemical regimes across a relatively small  
464 spatial domain.

465



466



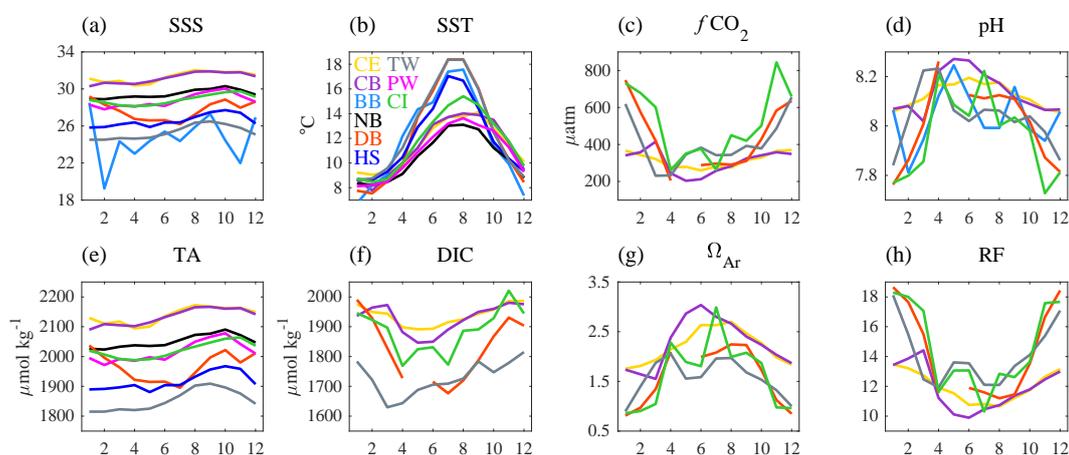
467 **Figure 4.** Near surface (< 10 m) seasonal cycles of (a) SSS, (b) SST, (c)  $f\text{CO}_2$ , (d)  $\text{pH}_T$ , (e) TA,  
468 (f) DIC, (g)  $\Omega_{\text{Ar}}$ , and (h) Revelle Factor (RF) for the eight regions shown in **Fig. 2c**: North Pacific  
469 (NP), British Columbia (BC), Outer Coast (OC), Columbia River (CR), Juan de Fuca (JF), Strait  
470 of Georgia (SG), Hood Canal (HC), and Puget Sound (PS).  
471

472 Most regions exhibit moderate seasonal TA variability with the exception of the Strait of  
473 Georgia where the Fraser River has a significant impact on the TA. Additionally, the Hood Canal  
474 and Puget Sound regions display somewhat erratic seasonal TA variations likely due to sporadic  
475 river influences and/or the small number of available samples that are unevenly distributed in space  
476 and time (**Appendix B**). DIC seasonal cycles are similar to TA but with larger amplitudes.  
477 Generally, DIC and TA are highest during winter and lowest during summer, with DIC:TA being  
478 lowest in summer.  $\Omega_{\text{Ar}}$  is dependent on temperature and the carbonate ion concentration [ $\text{CO}_3^{2-}$ ],  
479 which is inversely proportional to DIC:TA.  $\Omega_{\text{Ar}}$  seasonal cycles closely track (DIC:TA)<sup>-1</sup> changes  
480 in all regions, expressing the highest values during summer and the lowest values during winter.  
481 If SST is held constant in the CO2SYS calculations of  $\Omega_{\text{Ar}}$  for each region, the resulting seasonal  
482 cycle is altered by <10% for all regions. This indicates that local chemistry changes are controlling  
483 the  $\Omega_{\text{Ar}}$  seasonal cycle throughout the study domain, with temperature contributing minimally.  
484 Finally, RF values are elevated during winter and depressed during summer indicating a larger  
485 buffer capacity of waters during summer. RF values in the Salish Sea are ~50% higher than those  
486 in coastal regions, particularly during winter, indicating that Salish Sea waters are significantly  
487 less efficient at absorbing anthropogenic carbon than open ocean waters at the same latitude. In  
488 general, seasonal cycle amplitudes are largest within the Salish Sea for all parameters.  
489

490 Seasonal cycles for the nine moorings, using data collected from ~1 m depth, are shown in **Fig.**  
491 **5** and generally follow the same pattern as the eight regions. SSS decreases when moving inland  
492 from the outer coast toward Puget Sound. SST cycles are similar across moorings, with larger  
493 amplitudes found in shallower basins, such as at the Bellingham Bay and Carr Inlet moorings, as  
494 well as the moorings in Hood Canal where vertical mixing is weaker than in other parts of Puget  
495 Sound (Newton et al., 2003). pH and  $f\text{CO}_2$  display opposing seasonal cycles and TA follows the  
496 seasonality of salinity, as expected, exhibiting lower values during spring and summer and higher  
497 values during fall and winter. DIC displays more pronounced seasonal cycles with the lowest  
498 values in summer and highest values in winter. As seen for the regional discrete sample data,  $\Omega_{\text{Ar}}$



499 is highest in summer and lowest in winter following its inverse relationship with DIC:TA. Finally,  
 500 RF values are lowest in summer and highest in winter, with the largest annual mean values in the  
 501 Salish Sea. In general, seasonal cycle amplitudes for all parameters are higher in the Salish Sea  
 502 than on the outer coast, similar to what was found for the regional evaluations.  
 503



504

505 **Figure 5.** Near surface (~1 m) seasonal cycles of (a) SSS, (b) SST, (c)  $f\text{CO}_2$ , (d)  $\text{pH}_T$ , (e) TA, (f)  
 506 DIC, (g)  $\Omega_{\text{Ar}}$ , and (h) Revelle Factor (RF) for the nine moorings shown in **Fig. 1a**: Cape Elizabeth  
 507 (CE), Chá bã (CB), Carr Inlet (CI), Dabob Bay (DB), Twanoh (TW), Hoodsport (HS), Point Wells  
 508 (PW), North Buoy (NB), and Bellingham Bay (BB).  
 509

510 A shared feature of the region and mooring seasonal cycle results is the seasonal evolution of  
 511 offshore-onshore gradients in numerous parameters. For example, most regions and moorings  
 512 exhibit similar winter SST values; however, large gradients in SST are found between sites during  
 513 summer. Alternatively,  $\text{pH}$ ,  $\Omega_{\text{Ar}}$ , and  $f\text{CO}_2$  are most similar across sites during summer, with the  
 514 largest gradients found during winter. These findings suggest that seasonal resolution is needed to  
 515 accurately describe the spatial pattern of chemical gradients in the Pacific Northwest, particularly  
 516 since different parameters exhibit maximum gradients at dissimilar times of the year. Consistency  
 517 between the large-scale seasonal cycle patterns derived from the nine moorings and eight unique  
 518 regions is somewhat surprising due to differences in the sampling depth, with the moorings  
 519 sampling the top ~1m of the water column and regional seasonal cycles derived from samples  
 520 within the top 10 m. Since deeper waters are generally cooler and have higher salinity,  $f\text{CO}_2$ , TA,  
 521 and DIC values, this implies that seasonal forcing overwhelms any influence of stratification,  
 522 yielding the broad agreement between region and mooring seasonal cycles.



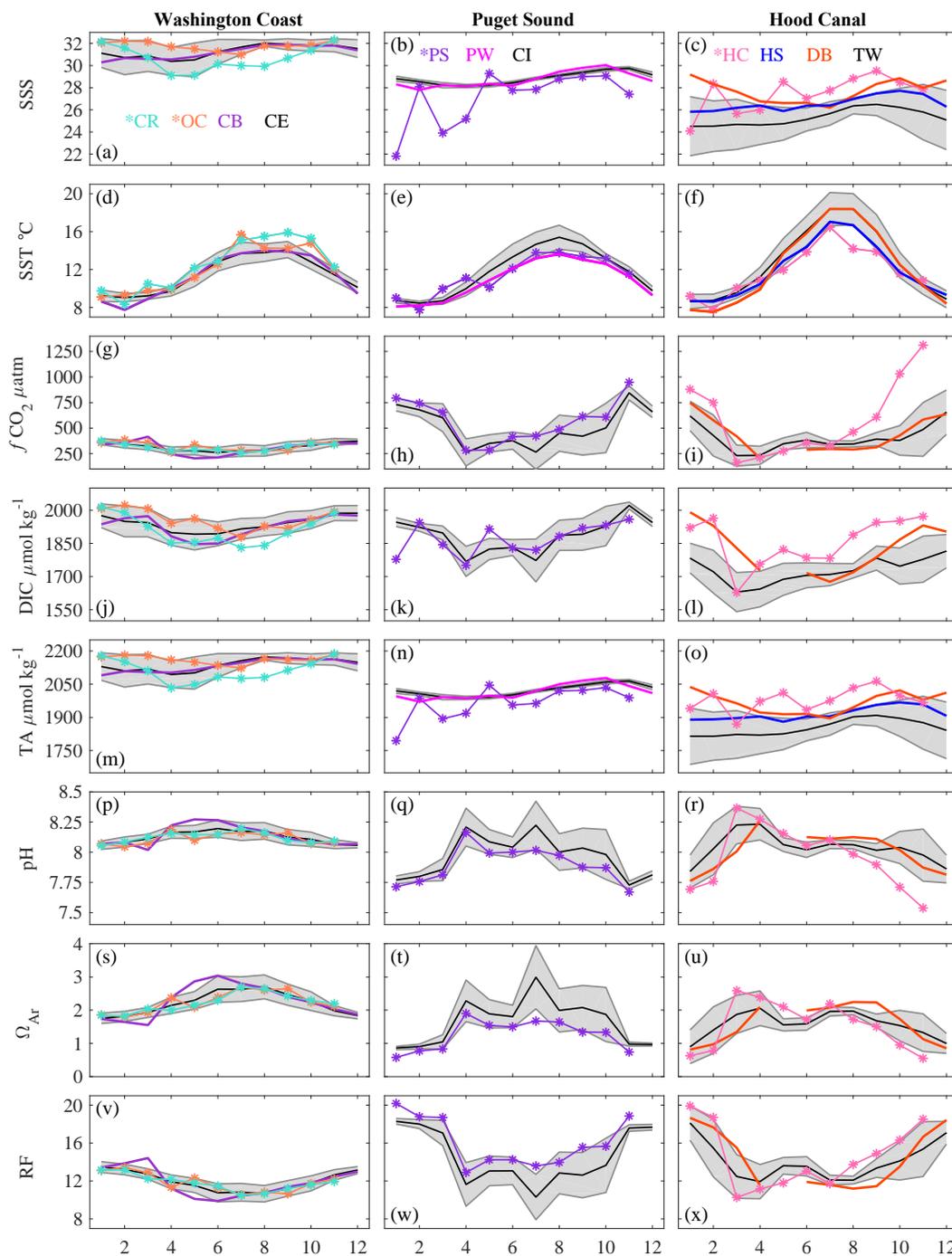
523

524 To more directly compare results from the regions and moorings and evaluate large-scale  
525 patterns of variability, we focus on seasonal cycles from the Washington Coast, Puget Sound, and  
526 Hood Canal domains. The left vertical panel of **Fig. 6** shows mean seasonal cycles from the Cape  
527 Elizabeth and Chá bã moorings and the Columbia River and Outer Coast regions. The two  
528 moorings lie within the Outer Coast region and display similar SSS and SST seasonal cycles,  
529 suggesting that large-scale rather than local physical processes dominate the seasonal cycles at  
530 both locations. However, there are notable differences in the seasonal cycles of carbonate system  
531 variables between moorings, which indicates that carbonate chemistry on the Washington coast is  
532 somewhat heterogeneous and cannot be adequately characterized with observations from a single  
533 mooring. In contrast to the moorings and Outer Coast domain, the Columbia River region exhibits  
534 a SSS and TA depression during spring/early-summer in concert with the maximum in Columbia  
535 River discharge. The fresh water plume is often advected southward due to seasonal winds and  
536 does not greatly influence salinity in the neighboring Outer Coast region or at the Cape Elizabeth  
537 mooring during the summer months.

538

539 Gray shading around the Cape Elizabeth mooring seasonal cycles reflects the average range of  
540 variability about the monthly mean (**Eq. 2**;  $\overline{m\sigma}$ ). Sensors on the Cape Elizabeth and Chá bã  
541 moorings collect samples every 3 hours, which means that one year of data equates to ~3,000  
542 observations, or ~250 observations per month (though there are occasional gaps in the time series;  
543 **Fig. 1b**). With multiple years of observations (**Appendix B**), data from these moorings provide  
544 robust constraints on the average range of sub-monthly variability. General agreement between the  
545 mooring data and relatively poorly-sampled regions is therefore somewhat exceptional, as many  
546 regional carbonate system values fall within the  $\pm 1\sigma$  envelope. This finding underscores that  
547 seasonal variations outcompete the influence of vertical gradients associated with stratification  
548 that would work to create discrepancies between the seasonal cycle data products.

549



550

551 **Figure 6.** Seasonal cycles of (a-c) SSS, (d-f) SST, (g-i)  $f\text{CO}_2$ , (j-l) DIC, (m-o) TA, (p-r) pH, (s-  
552 u)  $\Omega_{\text{Ar}}$ , and (v-x) RF for regions and moorings located along the outer Washington Coast, in Puget



553 Sound, and in Hood Canal. Asterisks are used for the regions. Black lines with gray shading  
554 highlight data from one mooring within each domain (see figure keys in subplots **a** to **c**). Gray  
555 shading shows the average range of variability about the mean during each month (i.e., average  
556 standard deviation across years derived from variance estimates). See also **Appendix D Figures**.  
557

558 Moving inland, the middle vertical panel of **Fig. 6** shows the Carr Inlet and Point Wells  
559 moorings and the Puget Sound region. The two moorings display similar SSS and TA seasonal  
560 cycles in terms of magnitude and timing, and general agreement in SST with a larger amplitude  
561 seasonal cycle at the Carr Inlet mooring due to more stratified conditions relative to the well-mixed  
562 Point Wells site in the main basin of Puget Sound. The Puget Sound region generally follows the  
563 mooring SSS and SST results, with some erratic variations likely caused by the much smaller  
564 number of observations (**Appendix B**). Carbonate chemistry seasonal cycles from the Carr Inlet  
565 mooring and Puget Sound region are similar in pattern but are offset due to the lower TA values  
566 found throughout Puget Sound for nearly all months. Differences in the timing of seasonal  
567 agreement between the carbon variables at Carr Inlet and Puget Sound may be attributable to  
568 stratification and sampling depth; however, it is also important note that the carbonate variables  
569 from Carr Inlet are based on only ~1.5 years of observations.

570

571 Finally, the right vertical panel of **Fig. 6** shows results from the Twanoh, Dabob Bay, and  
572 Hoodsport moorings as well as the Hood Canal region. There are significant differences in SSS  
573 across sites, while SST seasonal cycles are similar for Twanoh and Dabob Bay as well as for  
574 Hoodsport and the Hood Canal region. Seasonal SSS variations at Twanoh and Hoodsport are  
575 similar in timing and amplitude, but SSS is depressed at Twanoh due to its proximity to the  
576 Skokomish River. This influence is muted at the Hoodsport mooring and even more so at the  
577 Dabob Bay mooring (and correspondingly evident in TA), which is located much farther north and  
578 outside of the main channel. The moorings, thus, depict a range of diverse physical conditions  
579 within Hood Canal that depend on the sampling location. This is relevant because seasonal cycles  
580 for the Hood Canal region were determined using far fewer discrete samples that were collected  
581 across the entire domain (**Appendix B**), and thus blend all of the nuanced environments within the  
582 canal. Though observations are not presently available to show the range of variability across the  
583 Puget Sound region, this issue is likely also important when interpreting seasonal cycles from that  
584 domain.



585

586 The seasonal cycles for carbonate system variables are unique across sites within Hood Canal  
587 with somewhat better agreement found between the Twanoh and Dabob Bay moorings. The  
588 pycnocline in Hood Canal is typically strong and often located at ~5-8 m. Thus, the 10 m depth-  
589 integrated water samples used for the regional assessment would include colder, saltier, and more  
590 carbon rich waters than the surface moorings. This influence can be seen in SSS and DIC where  
591 the Hood Canal region displays higher values than the moorings during almost all months. This is  
592 particularly notable for SSS since the mooring estimates come from averaging the top 3 m of  
593 profiling mooring observations. Thus, the region and mooring SSS (and SST) discrepancies would  
594 likely be larger if the mooring values actually came from the top 1 m of water, where the  $f\text{CO}_2$   
595 measurements are made. This is supported by the extremely high  $f\text{CO}_2$  and low pH values observed  
596 in the Hood Canal region during winter that significantly exceed what is found at the moorings.  
597 Better agreement in  $f\text{CO}_2$  and pH exists across sites during summer, and there are fewer  
598 discrepancies for  $\Omega_{\text{Ar}}$  and RF at all locations throughout the year due to the large dependence of  
599 both parameters on DIC:TA, which is similar across sites.

600

601 Having discussed important nuances between the region and mooring seasonal cycles within  
602 these domains, it is important to also consider the large-scale gradients in carbonate chemistry  
603 across the domains. Taking a broader view of **Fig. 6**, greater environmental context is gained by  
604 comparing results from the open North Pacific Ocean and the Salish Sea. The large decline in SSS  
605 and increase in SST as waters become more stratified and heavily influenced by rivers when  
606 moving toward land greatly outcompetes any small differences found among sites within a given  
607 domain. There is an accompanying pattern of DIC decline along this path; however, TA exhibits  
608 even larger declines, resulting in elevated DIC:TA and thus more poorly buffered waters (higher  
609 RF values) within the Salish Sea. This leads to much higher  $f\text{CO}_2$  values and lower pH and  $\Omega_{\text{Ar}}$   
610 values in Hood Canal and Puget Sound than on the outer Washington coast. Since the North Pacific  
611 Ocean is the sole source of marine water to the Salish Sea, elevated carbon content within the  
612 Salish Sea reflects longer retention times and potentially intensified carbon cycling due to local  
613 nutrient and carbon dioxide pollution (Feely et al., 2010; Pelletier et al., 2017).

614



615 In addition to gradients in annual mean values of carbonate system parameters, there is a  
616 significant increase in the amplitude of seasonal cycles and in the range of sub-monthly variability  
617 moving landward. Unexpectedly, the seasonal evolution of sub-monthly variability is unique to  
618 different domains. For example, at the Twanoh mooring  $f\text{CO}_2$ , pH, and  $\Omega_{\text{Ar}}$  vary significantly more  
619 about the mean value during winter than during summer; however, at Carr Inlet and Cape  
620 Elizabeth, the opposite is true. This kind of insight, which emerges from sustained observing,  
621 provides precisely the type of information necessary to streamline OA monitoring, adaption, and  
622 research. For example, understanding how natural variability amplifies and attenuates throughout  
623 the year could improve our interpretations of sparse data, since samples collected during seasons  
624 with low sub-monthly variability may better represent the monthly mean condition. Additionally,  
625 the season exhibiting the lowest range of variability for a given parameter will likely be first to  
626 exit the envelope of natural variability as anthropogenic carbon accumulates (Hauri et al., 2013;  
627 Sutton et al., 2016). This nuanced information may be useful for determining when organisms will  
628 begin to experience persistent anomalous conditions and for designing experiments that accurately  
629 reflect nonstationarity in natural variability throughout the year.

630

#### 631 **4.2. Important limitations to consider**

632 Although the average seasonal cycles herein may help to inform OA monitoring and research  
633 conducted throughout the region, there are important limitations that must be acknowledged. The  
634 region-specific seasonal cycles are meant to provide large-scale, environmental context and an  
635 understanding of how the carbonate system varies across the broader domain. The moorings fill in  
636 temporal observing gaps and give robust statistical estimates of mean conditions at specific  
637 locations, as well as the average variability around those means. General agreement among the  
638 region and mooring seasonal cycles lends confidence to the results, but is not necessarily expected  
639 due to data paucity coupled with natural temporal variability and spatial heterogeneity within the  
640 various regions (e.g., **Fig. 6**). Opportunities for discrepancy were especially evident in the more  
641 stratified domains, such as Hood Canal and Puget Sound, due to differences in sampling depth for  
642 the various datasets. While seasonal forcing appears to dominate over stratification and vertical  
643 gradient issues in all domains, prior work has clearly shown that surface conditions do not reflect  
644 bottom water conditions (Feely et al., 2010; Pelletier et al., 2017). General agreement between



645 data products is likely due to all depths residing within the euphotic zone such that vertical  
646 gradients are minimized relative to those found throughout the full water column.

647

648 A key challenge in developing seasonal cycle estimates for the smaller regional domains (e.g.,  
649 Hood Canal and Puget Sound) is the dearth of data that renders these seasonal cycles more  
650 susceptible to biases that may result from non-homogenous conditions (e.g., proximity to rivers),  
651 interannual variability, or anomalous events (e.g., storms). As discrete observations amass, this  
652 issue will attenuate, but at present is something that needs to be considered when using the  
653 information provided herein. Additionally, regional seasonal cycles were evaluated using rigid  
654 domain boundaries, which we know to be dynamic in reality. Thus, caution should be used when  
655 relying on our estimates to evaluate samples collected near one of the boundaries. The use of a  
656 static regional TA-salinity relationship also has important implications for how carbonate system  
657 seasonal cycles can be interpreted. The TA-salinity relationship homogenizes the influence of  
658 freshwater alkalinity input (organic and inorganic) from numerous rivers in the region. As a result,  
659 the regression may not work perfectly near a specific river mouth. In these cases (e.g., the Twanoh  
660 mooring), the TA estimates may be biased and the bias may change seasonally. Importantly, **Table**  
661 **5** displays how these biases can propagate to other carbonate system variables. For all parameters,  
662 these errors are significantly smaller than the seasonal cycle amplitudes and, in most cases, are  
663 smaller than  $\pm 1\sigma$  values from the moorings (**Appendix B**). This is also true of the *in situ*  
664 measurement errors discussed throughout Sect. 2.

665

#### 666 **4.3. Understanding what the data do and do not reveal**

667 Characterizing average seasonal cycles throughout the study domain reveals important  
668 insights about how different types of observations can be used to better understand the dynamic  
669 coastal region. Discrete measurements made sporadically over space and time can be challenging  
670 to interpret without a vast number of data points, which are both cost- and time-intensive to collect.  
671 This limitation arises from spatial heterogeneity and the range of temporal scales over which  
672 variability can occur in the coastal zone (e.g., < days to > decades). Discrete observations provide  
673 a snapshot in time that may or may not represent the mean condition. Even with multiple years of  
674 observations, and no interannual variability, the true mean can be elusive due to sub-monthly  
675 variations. What discrete observations do extremely well is unveil the connectivity of spatial



676 domains by sampling across regions over a fixed period of time (e.g., research cruise). This too  
677 has uncertainty resulting from environmental heterogeneity and chemical gradients that are  
678 nonstationary over time, but is at present the most effective and efficient method of making dual-  
679 parameter, high-quality carbon observations across a wide spatial area.

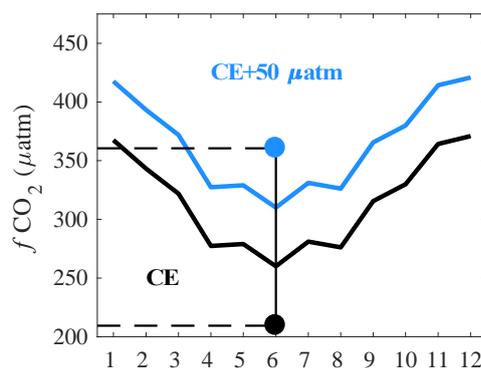
680

681 Moorings provide something altogether different. Sustained, high-frequency observations fill  
682 in temporal uncertainties, integrating over (often) unknown spatial scales that vary with tidal  
683 cycles, surface currents, and depth, allowing for robust determination of the mean condition and  
684 the average variability about the mean within the represented domain. This type of information  
685 lends context to discrete samples by providing bounds on the domain within which discrete  
686 samples may be expected to reside. **Figure 7**, for example, shows the monthly mean  $f\text{CO}_2$  seasonal  
687 cycle from the Cape Elizabeth mooring as well as the same data shifted upward by  $50 \mu\text{atm}$ , which  
688 is meant to represent an anomalous year, where the range of sub-monthly variability (shading) is  
689 identical for each year. Two discrete  $f\text{CO}_2$  samples collected during one “normal” and one  
690 “anomalous” June could span over  $100 \mu\text{atm}$ , hampering determination of which sample best  
691 reflects the mean conditions. Mooring observations collected over the same two years would give  
692 an average seasonal cycle that falls in-between the two lines shown in **Fig. 7**, and the discrete  
693 samples would thus lie outside of the associated  $\pm 1\sigma$  window. This would make it tempting to  
694 conclude that the discrete samples are outliers when the data actually indicate that one of the  
695 sampling years was anomalous. The true climatology would eventually emerge with additional  
696 years of observations. Thus, firm conclusions about what is or isn’t an anomaly or outlier may be  
697 extremely challenging to decipher without a sustained time series and information about the  
698 magnitude of sub-monthly variability.

699

700 To summarize, the array of time and space scales over which chemical variability occurs in  
701 U.S. West Coast waters hinders our ability to adequately describe the coastal system without the  
702 dual effort of ship-based and moored observations that provide complementary information. The  
703 seasonal cycles presented herein are an example of what can be achieved from such efforts and  
704 serve as a starting point from which strategic improvements to observational networks can be  
705 made.

706



707  
708 **Figure 7.** Schematic showing the Cape Elizabeth (CE) mooring monthly mean  $f\text{CO}_2$  seasonal cycle  
709 (black) and the seasonal cycle  $+50 \mu\text{atm}$  (blue), which is meant to reflect an anomalous year. Black  
710 and blue shading are identical in magnitude and show the mean standard deviation ( $\overline{\sigma}$ ). The filled  
711 circles show two hypothetical discrete samples collected during June of each year.  
712

## 713 5. Conclusions

714 Quantifying modern, human-induced changes in surface ocean chemistry requires a baseline  
715 from which to assess alterations. These baselines often do not exist in coastal regions where large-  
716 magnitude and high-frequency natural oscillations can overwhelm the secular anthropogenic  
717 signal. This inhibits our understanding of how coastal environments are changing and will continue  
718 to do so until these domains are adequately characterized. In an effort to galvanize baseline  
719 development for seawater parameters relevant to the study of ocean acidification, we have  
720 compiled  $\sim 100,000$  *in situ* carbonate system observations from marine surface waters near  
721 Washington State to characterize carbonate system variability throughout the region. By merging  
722  $f\text{CO}_2$  observations from the SOCAT-v4 database with dissolved inorganic carbon (DIC), total  
723 alkalinity (TA), and  $\text{pH}_T$  measurements made during discrete sampling efforts, we have estimated  
724 the average seasonal cycles of numerous carbonate system parameters in marine surface waters  
725 from eight distinct regions within the Pacific Northwest. Agreement between these regional  
726 estimates and the average seasonal cycles determined at nine surface moorings throughout  
727 Washington State coastal waters indicates a coherent, large-scale gradient in carbonate chemistry  
728 from the open North Pacific Ocean into the Salish Sea that is seasonally variable. Near surface  
729 salinity, DIC, TA, aragonite saturation state, and pH decline significantly along this path. Larger  
730 declines in TA relative to DIC lead to elevated  $f\text{CO}_2$  and Revelle Factor values within the Salish  
731 Sea, indicating a lower capacity of these inland waters to absorb anthropogenic carbon than  
732 offshore waters at the same latitude. These large-scale gradients persist throughout the year but



733 vary seasonally in magnitude with unique variables expressing larger gradients during different  
734 seasons. For all carbonate system parameters, the seasonal cycle amplitudes and ranges of sub-  
735 monthly variability are larger within the Salish Sea, indicating that organisms living in the Pacific  
736 Northwest experience starkly different chemical environments across the study domain.

737

738 In addition to providing environmental context, this analysis reveals important insights about  
739 the extreme care required to accurately interpret spatially and temporally nonuniform observations  
740 from the coastal zone. Spatial heterogeneity was discovered within all regional domains evaluated,  
741 indicating that chemical conditions, even in semi-enclosed domains, cannot be adequately  
742 characterized using observations from a single mooring. Additionally, the wide range of sub-  
743 monthly variability found at the mooring sites suggests that it may be difficult to determine what  
744 is and isn't an outlier in a discrete dataset, particularly in the context of interannual variability.  
745 By pairing multiple types of datasets (discrete, underway, and sustained time series) our ability to  
746 interpret complex coastal environments can be enhanced by more than what is achievable from  
747 any one observing approach. It is our intent that the insights gained from the seasonal cycle  
748 characterizations herein help to guide strategic monitoring, management, and scientific inquiry  
749 that leads to improved baseline development and water quality assessment in Washington State.

750

## 751 6. Appendix A

752 **Table A1.** Discrete datasets incorporated into the analysis. The observing period and the number  
753 of phosphate, silicate, SSS, and SST observations for each cruise are provided. Data DOIs  
754 matching those in Table 1 are also listed. Phosphate and silicate samples were analyzed following  
755 standard protocols (UNESCO, 1994) during all cruises.

756

| NUTRIENTS             | Obs. Period | Phosphate Obs. | Silicate Obs. | SSS Obs. | SST Obs. | Data DOI  |
|-----------------------|-------------|----------------|---------------|----------|----------|---|
| SOCAT-v4 <sup>1</sup> | 1970-2015   | 0              | 0             | 52,528   | 57,422   | <a href="https://doi.pangaea.de/10.1594/PANGAEA.866856">https://doi.pangaea.de/10.1594/PANGAEA.866856</a>                               |
| NOAA WCOA             | 05/2007     | 11             | 11            | 11       | 11       | <a href="https://doi.org/10.3334/CDIAC/otg.CLIVAR_NACP_West_Coast_Cruise_2007">10.3334/CDIAC/otg.CLIVAR_NACP_West_Coast_Cruise_2007</a> |
| UW PRISM*             | 02/2008     | 74             | 78            | 80       | 80       | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW PRISM/EPA*         | 08/2008     | 105            | 105           | 122      | 122      | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| PacOOS*               | 08/2009     | 8              | 8             | 8        | 8        | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW PRISM*             | 09/2009     | 38             | 38            | 38       | 38       | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| PacOOS*               | 05/2010     | 13             | 13            | 13       | 13       | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| PacOOS*               | 08/2010     | 22             | 22            | 22       | 22       | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW PRISM*             | 11/2010     | 39             | 39            | 40       | 40       | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| UW/Chá bā*            | 05/2011     | 0              | 0             | 1        | 1        | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>   |
| NOAA WCOA             | 08/2011     | 11             | 11            | 37       | 37       | <a href="https://doi.org/10.3334/CDIAC/OTG.COAST_WCOA2011">10.3334/CDIAC/OTG.COAST_WCOA2011</a>   |



|              |         |            |            |               |               |   |
|--------------|---------|------------|------------|---------------|---------------|---|
| UW PRISM*    | 10/2011 | 38         | 38         | 60            | 60            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| UW/NANOOS*   | 05/2012 | 0          | 0          | 4             | 4             | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| NOAA WCOA    | 09/2012 | 40         | 43         | 43            | 43            | <a href="https://doi.org/10.3334/CDIAC/OTG.COAST_WCOA2012">10.3334/CDIAC/OTG.COAST_WCOA2012</a> |
| UW/Chá bã*   | 01/2013 | 0          | 0          | 2             | 2             | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| UW/NANOOS*   | 04/2013 | 1          | 1          | 19            | 19            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| NOAA WCOA    | 08/2013 | 59         | 59         | 59            | 59            | <a href="https://doi.org/10.7289/V5C53HXP">10.7289/V5C53HXP</a>                                 |
| UW/NANOOS*   | 09/2013 | 14         | 14         | 19            | 19            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| UW/NANOOS*   | 06/2014 | 10         | 10         | 10            | 10            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| ECY          | 06/2014 | 8          | 8          | 17            | 17            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| WOAC*        | 07/2014 | 61         | 61         | 68            | 68            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| ECY          | 07/2014 | 0          | 0          | 12            | 12            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| ECY          | 08/2014 | 0          | 0          | 18            | 18            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| WOAC*        | 09/2014 | 64         | 64         | 64            | 64            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| ECY          | 09/2014 | 0          | 0          | 14            | 14            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| UW/Chá bã*   | 10/2014 | 19         | 19         | 19            | 19            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| ECY          | 10/2014 | 0          | 0          | 9             | 9             | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| ECY          | 11/2014 | 6          | 6          | 9             | 9             | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| ECY          | 01/2015 | 13         | 12         | 13            | 13            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| ECY          | 02/2015 | 11         | 11         | 15            | 15            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| ECY          | 03/2015 | 8          | 8          | 20            | 20            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| ECY          | 04/2015 | 5          | 5          | 15            | 15            | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| ECY          | 05/2015 | 2          | 2          | 6             | 6             | <a href="https://doi.org/10.5281/zenodo.1184657">https://doi.org/10.5281/zenodo.1184657</a>     |
| <b>TOTAL</b> |         | <b>680</b> | <b>686</b> | <b>53,415</b> | <b>58,309</b> |   |

757 <sup>1</sup>The coastal subset of SOCAT-v4 data can be easily accessed here: <https://www.socat.info/index.php/version-4/>

758 \*Full dataset in preparation for submission to a long-term data repository.

759

## 760 7. Appendix B

761 **Table B1.** Average monthly SSS seasonal cycles for the eight unique regions. The total number  
762 of observations (# Obs.) contributing to the monthly average estimate is included.

763

| SSS                    | Jan. | Feb. | Mar. | Apr. | May  | June | July | Aug. | Sept. | Oct. | Nov. | Dec. |
|------------------------|------|------|------|------|------|------|------|------|-------|------|------|------|
| North Pacific (NP)     | 32.3 | 32.5 | 32.9 | 32.3 | 32.1 | 31.9 | 31.7 | 31.9 | 32.0  | 32.1 | 32.2 | 32.4 |
| # Obs.                 | 163  | 439  | 250  | 66   | 305  | 2791 | 4024 | 1812 | 504   | 88   | 109  | 29   |
| British Columbia (BC)  | 31.3 | 31.7 | 31.4 | 31.3 | 31.0 | 31.7 | 31.4 | 31.8 | 31.8  | 31.9 | 31.7 | 31.2 |
| # Obs.                 | 162  | 647  | 205  | 160  | 452  | 1107 | 8067 | 1735 | 250   | 229  | 110  | 100  |
| Outer Coast (OC)       | 32.1 | 32.2 | 32.2 | 31.7 | 31.5 | 31.2 | 31.0 | 31.8 | 31.8  | 31.8 | 32.3 |      |
| # Obs.                 | 57   | 65   | 170  | 7    | 34   | 1700 | 6969 | 5585 | 749   | 5    | 13   |      |
| Columbia River (CR)    | 32.1 | 31.6 | 30.7 | 29.1 | 29.1 | 30.1 | 30.0 | 29.9 | 30.7  | 31.4 | 32.3 |      |
| # Obs.                 | 120  | 19   | 98   | 23   | 135  | 3456 | 1545 | 3059 | 719   | 24   | 46   |      |
| Juan de Fuca (JF)      | 30.4 | 30.7 | 30.8 | 30.1 | 30.8 | 31.0 | 30.8 | 31.0 | 31.3  | 31.1 | 31.2 | 31.3 |
| # Obs.                 | 68   | 2099 | 172  | 48   | 274  | 627  | 2046 | 814  | 152   | 44   | 43   | 40   |
| Strait of Georgia (SG) |      | 28.7 |      | 28.1 | 28.0 | 27.7 | 22.9 | 29.3 | 29.9  | 29.9 |      |      |
| # Obs.                 |      | 192  |      | 5    | 58   | 32   | 5    | 35   | 6     | 1    |      |      |
| Hood Canal (HC)        | 24.1 | 28.3 | 25.7 | 26.0 | 28.5 | 27.0 | 27.7 | 28.8 | 29.5  | 28.5 | 27.9 |      |
| # Obs.                 | 1    | 24   | 3    | 3    | 3    | 3    | 27   | 28   | 6     | 71   | 25   |      |



|                  |      |      |      |      |      |      |      |      |      |      |      |
|------------------|------|------|------|------|------|------|------|------|------|------|------|
| Puget Sound (PS) | 21.8 | 28.1 | 23.9 | 25.2 | 29.3 | 27.8 | 27.8 | 28.8 | 29.0 | 29.1 | 27.4 |
| # Obs.           | 12   | 57   | 17   | 12   | 35   | 93   | 287  | 257  | 32   | 49   | 15   |

764

765

766

767

**Table B2.** Average monthly SST seasonal cycles for the eight unique regions. The total number of observations (# Obs.) contributing to the monthly average estimate is included.

| SST (°C)               | Jan. | Feb. | Mar. | Apr. | May  | June | July | Aug. | Sept. | Oct. | Nov. | Dec. |
|------------------------|------|------|------|------|------|------|------|------|-------|------|------|------|
| North Pacific (NP)     | 9.3  | 9.1  | 9.7  | 10.1 | 11.6 | 13.5 | 15.5 | 16.7 | 16.1  | 14.9 | 12.2 | 9.6  |
| # Obs.                 | 163  | 446  | 250  | 66   | 364  | 2791 | 4026 | 1922 | 504   | 88   | 109  | 29   |
| British Columbia (BC)  | 9.2  | 8.8  | 8.7  | 9.8  | 11.5 | 12.5 | 13.8 | 13.3 | 13.8  | 11.9 | 10.9 | 9.0  |
| # Obs.                 | 166  | 644  | 205  | 160  | 567  | 1102 | 8078 | 1938 | 250   | 229  | 110  | 100  |
| Outer Coast (OC)       | 9.1  | 9.3  | 9.8  | 9.9  | 11.2 | 12.6 | 15.7 | 14.3 | 14.2  | 14.8 | 12.0 |      |
| # Obs.                 | 57   | 65   | 170  | 7    | 34   | 1700 | 6969 | 6301 | 750   | 5    | 13   |      |
| Columbia River (CR)    | 9.7  | 8.3  | 10.5 | 10.1 | 12.2 | 12.9 | 15.1 | 15.5 | 15.9  | 15.3 | 12.3 |      |
| # Obs.                 | 120  | 19   | 98   | 23   | 135  | 3456 | 1545 | 3614 | 719   | 24   | 46   |      |
| Juan de Fuca (JF)      | 8.1  | 7.6  | 8.5  | 9.4  | 10.0 | 10.3 | 11.1 | 10.9 | 11.2  | 10.9 | 10.2 | 8.1  |
| # Obs.                 | 71   | 2108 | 172  | 48   | 378  | 628  | 1953 | 816  | 153   | 44   | 43   | 39   |
| Strait of Georgia (SG) |      | 7.2  |      | 9.1  | 11.7 | 11.8 | 14.7 | 13.8 | 12.0  | 11.7 |      |      |
| # Obs.                 |      | 197  |      | 5    | 59   | 33   | 5    | 35   | 6     | 1    |      |      |
| Hood Canal (HC)        | 9.2  | 7.8  | 10.1 | 10.9 | 12.0 | 13.9 | 16.5 | 14.2 | 13.9  | 11.6 | 10.8 |      |
| # Obs.                 | 1    | 24   | 3    | 3    | 3    | 3    | 27   | 28   | 6     | 71   | 25   |      |
| Puget Sound (PS)       | 9.0  | 7.8  | 10.0 | 11.1 | 10.1 | 12.1 | 13.8 | 13.8 | 13.4  | 13.2 | 11.5 |      |
| # Obs.                 | 12   | 57   | 17   | 12   | 35   | 93   | 298  | 257  | 32    | 49   | 15   |      |

768

769

770

771

**Table B3.** Average monthly TA seasonal cycles for the eight unique regions. The total number of observations (# Obs.) contributing to the monthly average estimate is included.

| TA ( $\mu\text{mol kg}^{-1}$ ) | Jan. | Feb. | Mar. | Apr. | May  | June | July | Aug. | Sept. | Oct. | Nov. | Dec. |
|--------------------------------|------|------|------|------|------|------|------|------|-------|------|------|------|
| North Pacific (NP)             | 2187 | 2194 | 2214 | 2185 | 2175 | 2165 | 2155 | 2167 | 2171  | 2176 | 2183 | 2189 |
| # Obs.                         | 163  | 439  | 250  | 66   | 305  | 2791 | 4024 | 1812 | 504   | 88   | 109  | 29   |
| British Columbia (BC)          | 2138 | 2156 | 2141 | 2138 | 2124 | 2157 | 2144 | 2163 | 2163  | 2169 | 2158 | 2133 |
| # Obs.                         | 162  | 647  | 205  | 160  | 452  | 1107 | 8067 | 1735 | 250   | 229  | 110  | 100  |
| Outer Coast (OC)               | 2173 | 2181 | 2179 | 2160 | 2150 | 2135 | 2123 | 2166 | 2160  | 2158 | 2184 |      |
| # Obs.                         | 57   | 65   | 170  | 7    | 34   | 1700 | 6969 | 5584 | 749   | 5    | 13   |      |
| Columbia River (CR)            | 2179 | 2151 | 2111 | 2034 | 2049 | 2082 | 2076 | 2080 | 2113  | 2142 | 2187 |      |
| # Obs.                         | 120  | 19   | 98   | 23   | 135  | 3456 | 1545 | 3059 | 719   | 24   | 46   |      |
| Juan de Fuca (JF)              | 2097 | 2111 | 2116 | 2081 | 2115 | 2125 | 2113 | 2125 | 2140  | 2126 | 2133 | 2137 |
| # Obs.                         | 68   | 2099 | 172  | 48   | 267  | 627  | 2046 | 816  | 152   | 44   | 43   | 40   |
| Strait of Georgia (SG)         |      | 2012 |      | 1984 | 1980 | 1965 | 1842 | 2045 | 2071  | 2073 |      |      |
| # Obs.                         |      | 192  |      | 5    | 58   | 32   | 5    | 35   | 6     | 1    |      |      |
| Hood Canal (HC)                | 1941 | 2007 | 1870 | 1972 | 2011 | 1935 | 1975 | 2033 | 2063  | 1999 | 1967 |      |
| # Obs.                         | 1    | 24   | 3    | 3    | 3    | 3    | 27   | 28   | 6     | 71   | 25   |      |
| Puget Sound (PS)               | 1795 | 1986 | 1894 | 1919 | 2046 | 1956 | 1963 | 2020 | 2023  | 2035 | 1989 |      |



772

773

774

775

|        |    |    |    |    |    |    |     |     |    |    |    |
|--------|----|----|----|----|----|----|-----|-----|----|----|----|
| # Obs. | 12 | 57 | 17 | 12 | 35 | 93 | 287 | 257 | 32 | 49 | 15 |
|--------|----|----|----|----|----|----|-----|-----|----|----|----|

**Table B4.** Average monthly DIC seasonal cycles for the eight unique regions. The total number of observations (# Obs.) contributing to the monthly average estimate is included.

| DIC ( $\mu\text{mol kg}^{-1}$ ) | Jan. | Feb. | Mar.   | Apr. | May  | June | July | Aug. | Sept. | Oct. | Nov. | Dec. |
|---------------------------------|------|------|--------|------|------|------|------|------|-------|------|------|------|
| North Pacific (NP)              | 2019 | 2027 | 2029   | 1992 | 1974 | 1954 | 1941 | 1940 | 1950  | 1964 | 1990 | 2017 |
| # Obs.                          | 163  | 446  | 250    | 66   | 305  | 2791 | 4024 | 1814 | 504   | 88   | 109  | 29   |
| British Columbia (BC)           | 1998 | 2015 | 1992   | 1944 | 1903 | 1928 | 1935 | 1959 | 1967  | 2012 | 2001 | 2000 |
| # Obs.                          | 162  | 627  | 205    | 160  | 462  | 1104 | 8067 | 1732 | 250   | 229  | 110  | 100  |
| Outer Coast (OC)                | 2008 | 2023 | 2006   | 1941 | 1962 | 1918 | 1879 | 1928 | 1919  | 1957 | 1993 |      |
| # Obs.                          | 57   | 65   | 170    | 7    | 34   | 1700 | 6969 | 5585 | 749   | 5    | 13   |      |
| Columbia River (CR)             | 2014 | 1988 | 1926.5 | 1852 | 1855 | 1874 | 1831 | 1841 | 1897  | 1938 | 1989 |      |
| # Obs.                          | 120  | 19   | 98     | 23   | 135  | 3456 | 1545 | 3059 | 719   | 24   | 46   |      |
| Juan de Fuca (JF)               | 1993 | 2034 | 2020   | 1956 | 1970 | 2024 | 2016 | 2038 | 2056  | 2054 | 2031 | 2075 |
| # Obs.                          | 68   | 2103 | 172    | 48   | 301  | 633  | 2044 | 827  | 152   | 44   | 43   | 40   |
| Strait of Georgia (SG)          |      | 1966 |        | 1857 | 1808 | 1833 | 1702 | 1905 | 1981  | 1958 |      |      |
| # Obs.                          |      | 192  |        | 5    | 58   | 32   | 5    | 35   | 6     | 1    |      |      |
| Hood Canal (HC)                 | 1921 | 1964 | 1628   | 1756 | 1822 | 1784 | 1783 | 1888 | 1944  | 1951 | 1972 |      |
| # Obs.                          | 1    | 24   | 3      | 3    | 3    | 3    | 27   | 28   | 6     | 71   | 25   |      |
| Puget Sound (PS)                | 1778 | 1944 | 1844   | 1751 | 1915 | 1830 | 1820 | 1881 | 1919  | 1932 | 1959 |      |
| # Obs.                          | 12   | 57   | 17     | 12   | 35   | 93   | 287  | 257  | 32    | 49   | 15   |      |

776

777

778

779

**Table B5.** Average monthly  $f\text{CO}_2$  seasonal cycles for the eight unique regions. The total number of observations (# Obs.) contributing to the monthly average estimate is included.

| $f\text{CO}_2$ ( $\mu\text{atm}$ ) | Jan. | Feb. | Mar. | Apr. | May | June | July | Aug. | Sept. | Oct. | Nov. | Dec. |
|------------------------------------|------|------|------|------|-----|------|------|------|-------|------|------|------|
| North Pacific (NP)                 | 361  | 363  | 343  | 320  | 320 | 322  | 340  | 338  | 344   | 344  | 349  | 357  |
| # Obs.                             | 163  | 446  | 250  | 66   | 364 | 2791 | 4026 | 1876 | 504   | 88   | 109  | 29   |
| British Columbia (BC)              | 402  | 413  | 380  | 300  | 261 | 290  | 335  | 349  | 368   | 445  | 404  | 432  |
| # Obs.                             | 164  | 648  | 205  | 160  | 562 | 1083 | 8078 | 1922 | 248   | 229  | 110  | 100  |
| Outer Coast (OC)                   | 358  | 385  | 356  | 264  | 337 | 288  | 282  | 275  | 285   | 359  | 351  |      |
| # Obs.                             | 57   | 65   | 170  | 7    | 34  | 1700 | 6969 | 6301 | 750   | 5    | 13   |      |
| Columbia River (CR)                | 373  | 341  | 311  | 277  | 287 | 287  | 257  | 275  | 323   | 350  | 340  |      |
| # Obs.                             | 120  | 19   | 98   | 23   | 135 | 3456 | 1545 | 3614 | 719   | 24   | 46   |      |
| Juan de Fuca (JF)                  | 486  | 621  | 550  | 438  | 433 | 584  | 624  | 655  | 699   | 769  | 609  | 724  |
| # Obs.                             | 69   | 2110 | 172  | 48   | 367 | 638  | 2094 | 832  | 157   | 44   | 43   | 40   |
| Strait of Georgia (SG)             |      | 694  |      | 386  | 304 | 400  | 300  | 466  | 621   | 500  |      |      |
| # Obs.                             |      | 197  |      | 5    | 59  | 33   | 5    | 35   | 6     | 1    |      |      |
| Hood Canal (HC)                    | 879  | 750  | 161  | 211  | 274 | 356  | 325  | 462  | 607   | 1032 | 1311 |      |
| # Obs.                             | 1    | 24   | 3    | 3    | 3   | 3    | 27   | 28   | 6     | 71   | 25   |      |
| Puget Sound (PS)                   | 794  | 742  | 654  | 283  | 286 | 416  | 422  | 487  | 614   | 607  | 948  |      |
| # Obs.                             | 12   | 57   | 17   | 12   | 35  | 93   | 298  | 257  | 32    | 49   | 15   |      |



780

781 **Table B6.** Average monthly pH seasonal cycles for the eight unique regions. The total number of  
782 observations (# Obs.) contributing to the monthly average estimate is included.

783

| pH                     | Jan. | Feb. | Mar. | Apr. | May  | June | July | Aug. | Sept. | Oct. | Nov. | Dec. |
|------------------------|------|------|------|------|------|------|------|------|-------|------|------|------|
| North Pacific (NP)     | 8.07 | 8.07 | 8.09 | 8.12 | 8.12 | 8.11 | 8.09 | 8.10 | 8.09  | 8.09 | 8.08 | 8.07 |
| # Obs.                 | 163  | 446  | 250  | 66   | 358  | 2791 | 4024 | 1770 | 504   | 88   | 109  | 29   |
| British Columbia (BC)  | 8.02 | 8.02 | 8.05 | 8.14 | 8.18 | 8.16 | 8.11 | 8.09 | 8.07  | 8.00 | 8.03 | 8.00 |
| # Obs.                 | 160  | 647  | 205  | 160  | 455  | 1083 | 8067 | 1744 | 248   | 229  | 110  | 100  |
| Outer Coast (OC)       | 8.07 | 8.05 | 8.07 | 8.19 | 8.10 | 8.15 | 8.16 | 8.14 | 8.16  | 8.07 | 8.08 |      |
| # Obs.                 | 57   | 65   | 170  | 7    | 34   | 1700 | 6969 | 5585 | 749   | 5    | 13   |      |
| Columbia River (CR)    | 8.06 | 8.09 | 8.12 | 8.16 | 8.14 | 8.15 | 8.20 | 8.16 | 8.10  | 8.08 | 8.09 |      |
| # Obs.                 | 120  | 19   | 98   | 23   | 135  | 3456 | 1545 | 3059 | 719   | 24   | 46   |      |
| Juan de Fuca (JF)      | 7.95 | 7.85 | 7.90 | 7.99 | 8.02 | 7.89 | 7.86 | 7.83 | 7.81  | 7.78 | 7.88 | 7.79 |
| # Obs.                 | 67   | 2110 | 172  | 48   | 301  | 635  | 2044 | 799  | 152   | 44   | 43   | 40   |
| Strait of Georgia (SG) |      | 7.78 |      | 8.03 | 8.13 | 8.02 | 8.08 | 7.98 | 7.85  | 7.93 |      |      |
| # Obs.                 |      | 192  |      | 5    | 58   | 32   | 5    | 35   | 6     | 1    |      |      |
| Hood Canal (HC)        | 7.69 | 7.76 | 8.37 | 8.27 | 8.15 | 8.05 | 8.10 | 7.98 | 7.90  | 7.71 | 7.54 |      |
| # Obs.                 | 1    | 24   | 3    | 3    | 3    | 3    | 27   | 28   | 6     | 71   | 25   |      |
| Puget Sound (PS)       | 7.71 | 7.76 | 7.81 | 8.16 | 7.99 | 8.00 | 8.02 | 7.97 | 7.88  | 7.87 | 7.67 |      |
| # Obs.                 | 12   | 57   | 17   | 12   | 35   | 93   | 287  | 257  | 32    | 49   | 15   |      |

784

785 **Table B7.** Average monthly  $\Omega_{Ar}$  seasonal cycles for the eight unique regions. The total number of  
786 observations (# Obs.) contributing to the monthly average estimate is included.

787

| $\Omega_{Ar}$          | Jan. | Feb. | Mar. | Apr. | May | June | July | Aug. | Sept. | Oct. | Nov. | Dec. |
|------------------------|------|------|------|------|-----|------|------|------|-------|------|------|------|
| North Pacific (NP)     | 1.9  | 1.9  | 2.0  | 2.1  | 2.2 | 2.3  | 2.4  | 2.5  | 2.5   | 2.4  | 2.1  | 1.9  |
| # Obs.                 | 163  | 446  | 250  | 66   | 338 | 2791 | 4024 | 1814 | 504   | 88   | 109  | 29   |
| British Columbia (BC)  | 1.6  | 1.6  | 1.7  | 2.1  | 2.4 | 2.5  | 2.3  | 2.3  | 2.2   | 1.8  | 1.8  | 1.6  |
| # Obs.                 | 161  | 647  | 205  | 160  | 455 | 1105 | 8067 | 1732 | 248   | 229  | 110  | 100  |
| Outer Coast (OC)       | 1.9  | 1.8  | 1.9  | 2.4  | 2.1 | 2.4  | 2.7  | 2.6  | 2.6   | 2.2  | 2.1  |      |
| # Obs.                 | 57   | 65   | 170  | 7    | 34  | 1700 | 6969 | 5585 | 749   | 5    | 13   |      |
| Columbia River (CR)    | 1.9  | 1.8  | 2.0  | 2.0  | 2.1 | 2.3  | 2.7  | 2.6  | 2.4   | 2.3  | 2.2  |      |
| # Obs.                 | 120  | 19   | 98   | 23   | 135 | 3456 | 1545 | 3059 | 719   | 24   | 46   |      |
| Juan de Fuca (JF)      | 1.3  | 1.1  | 1.2  | 1.5  | 1.7 | 1.3  | 1.3  | 1.2  | 1.2   | 1.1  | 1.3  | 1.0  |
| # Obs.                 | 68   | 2110 | 172  | 48   | 300 | 633  | 2043 | 788  | 150   | 44   | 43   | 39   |
| Strait of Georgia (SG) |      | 0.8  |      | 1.5  | 1.9 | 1.6  | 1.6  | 1.7  | 1.2   | 1.4  |      |      |
| # Obs.                 |      | 192  |      | 5    | 58  | 32   | 5    | 35   | 6     | 1    |      |      |
| Hood Canal (HC)        | 0.6  | 0.8  | 2.6  | 2.4  | 2.1 | 1.7  | 2.2  | 1.7  | 1.5   | 1.0  | 0.5  |      |
| # Obs.                 | 1    | 24   | 3    | 3    | 3   | 3    | 27   | 28   | 6     | 71   | 25   |      |
| Puget Sound (PS)       | 0.6  | 0.8  | 0.8  | 1.9  | 1.5 | 1.5  | 1.7  | 1.6  | 1.3   | 1.3  | 0.7  |      |
| # Obs.                 | 12   | 57   | 17   | 12   | 35  | 93   | 287  | 257  | 32    | 49   | 15   |      |

788



789 **Table B8.** Average monthly Revelle Factor seasonal cycles for the eight unique regions. The total  
790 number of observations (# Obs.) contributing to the monthly average estimate is included.  
791

| Revelle Factor         | Jan. | Feb. | Mar. | Apr. | May  | June | July | Aug. | Sept. | Oct. | Nov. | Dec. |
|------------------------|------|------|------|------|------|------|------|------|-------|------|------|------|
| North Pacific (NP)     | 13.0 | 13.1 | 12.5 | 12.1 | 11.8 | 11.5 | 11.3 | 11.0 | 11.1  | 11.4 | 12.1 | 12.9 |
| # Obs.                 | 163  | 446  | 250  | 66   | 356  | 2791 | 4024 | 1814 | 504   | 88   | 109  | 29   |
| British Columbia (BC)  | 14.1 | 14.1 | 13.7 | 12.1 | 11.2 | 11.2 | 11.6 | 11.9 | 12.0  | 13.5 | 13.4 | 14.4 |
| # Obs.                 | 160  | 647  | 205  | 160  | 459  | 1082 | 8067 | 1727 | 248   | 229  | 110  | 100  |
| Outer Coast (OC)       | 13.1 | 13.4 | 12.9 | 11.3 | 12.3 | 11.3 | 10.6 | 10.8 | 10.7  | 11.7 | 12.2 |      |
| # Obs.                 | 57   | 65   | 170  | 7    | 34   | 1700 | 6969 | 5585 | 749   | 5    | 13   |      |
| Columbia River (CR)    | 13.1 | 13.2 | 12.2 | 12.2 | 11.8 | 11.5 | 10.5 | 10.7 | 11.2  | 11.6 | 11.9 |      |
| # Obs.                 | 120  | 19   | 98   | 23   | 135  | 3456 | 1545 | 3059 | 719   | 24   | 46   |      |
| Juan de Fuca (JF)      | 15.7 | 17.2 | 16.2 | 14.7 | 14.0 | 15.9 | 16.1 | 16.5 | 16.6  | 17.1 | 15.8 | 17.8 |
| # Obs.                 | 67   | 2110 | 172  | 48   | 300  | 635  | 2043 | 795  | 152   | 44   | 43   | 39   |
| Strait of Georgia (SG) |      | 18.6 |      | 14.6 | 12.5 | 14.1 | 13.2 | 13.9 | 16.2  | 15.0 |      |      |
| # Obs.                 |      | 192  |      | 5    | 58   | 32   | 5    | 35   | 6     | 1    |      |      |
| Hood Canal (HC)        | 19.9 | 18.7 | 10.3 | 11.2 | 11.8 | 13.0 | 11.7 | 13.7 | 14.9  | 16.3 | 18.5 |      |
| # Obs.                 | 1    | 24   | 3    | 3    | 3    | 3    | 27   | 28   | 6     | 71   | 25   |      |
| Puget Sound (PS)       | 20.2 | 18.8 | 18.7 | 12.9 | 14.2 | 14.2 | 13.6 | 14.0 | 15.6  | 15.7 | 18.9 |      |
| # Obs.                 | 12   | 57   | 17   | 12   | 35   | 93   | 287  | 257  | 32    | 49   | 15   |      |

792

793 **8. Appendix C**

794 **Table C1.** Average monthly SSS seasonal cycles for the nine Washington State moorings. The  
795 average monthly standard deviation (StDev) and total number of observations (# Obs.)  
796 contributing to the monthly average estimate are included.  
797

| SSS                 | Jan.  | Feb.  | Mar.  | Apr.  | May   | June  | July  | Aug.  | Sept. | Oct.  | Nov.  | Dec.  |
|---------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Chá bá (CB)         | 30.3  | 30.7  | 30.6  | 30.5  | 30.8  | 31.2  | 31.5  | 31.9  | 31.9  | 31.8  | 31.8  | 31.4  |
| StDev               | 1.2   | 1.2   | 1.6   | 1.1   | 0.4   | 1.2   | 0.8   | 0.3   | 0.5   | 0.5   | 0.5   | 0.7   |
| # Obs.              | 398   | 224   | 248   | 280   | 485   | 1,080 | 1,461 | 1,648 | 1,346 | 1,029 | 646   | 496   |
| Cape Elizabeth (CE) | 31.1  | 30.7  | 30.9  | 30.4  | 30.5  | 31.2  | 31.7  | 32.0  | 31.9  | 31.8  | 31.8  | 31.5  |
| StDev               | 1.3   | 1.5   | 1.4   | 1.3   | 1.5   | 1.1   | 0.7   | 0.3   | 0.5   | 0.5   | 0.6   | 0.8   |
| # Obs.              | 1,488 | 1,352 | 1,295 | 1,199 | 1,161 | 1,246 | 1,715 | 1,812 | 1,621 | 1,756 | 1,538 | 1,484 |
| Twanoh (TW)         | 24.5  | 24.5  | 24.7  | 24.6  | 24.7  | 25.1  | 25.7  | 26.4  | 26.5  | 26.2  | 25.8  | 25.1  |
| StDev               | 2.7   | 2.3   | 2.3   | 1.8   | 1.5   | 1.0   | 1.1   | 0.7   | 1.0   | 1.7   | 2.5   | 2.7   |
| # Obs.              | 684   | 913   | 1,443 | 2,678 | 3,332 | 3,234 | 3,169 | 3,664 | 3,993 | 3,220 | 1,545 | 591   |
| Dabob Bay (DB)      | 29.2  | 28.3  | 27.6  | 26.8  | 26.6  | 26.6  | 26.2  | 27.3  | 28.3  | 28.9  | 28.0  | 28.6  |
| StDev               | 0.1   | 0.6   | 0.8   | 0.5   | 0.5   | 0.8   | 1.0   | 0.9   | 0.3   | 0.5   | 1.0   | 0.9   |
| # Obs.              | 34    | 33    | 85    | 109   | 174   | 286   | 767   | 1,167 | 1,138 | 667   | 163   | 39    |
| Carr Inlet (CI)     | 28.8  | 28.5  | 28.2  | 28.1  | 28.2  | 28.4  | 28.8  | 29.1  | 29.4  | 29.7  | 29.7  | 29.2  |
| StDev               | 0.2   | 0.2   | 0.2   | 0.2   | 0.2   | 0.1   | 0.1   | 0.1   | 0.1   | 0.2   | 0.1   | 0.2   |
| # Obs.              | 349   | 347   | 1,008 | 937   | 820   | 1,161 | 1,629 | 1,641 | 1,611 | 1,566 | 716   | 584   |
| Bellingham Bay (BB) | 28.5  | 19.3  | 24.3  | 23.0  | 24.4  | 25.4  | 24.4  | 25.9  | 27.3  | 24.4  | 22.0  | 26.9  |



|                  |               |      |       |       |       |       |       |       |       |       |       |       |       |
|------------------|---------------|------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
|                  | <i>StDev</i>  | 2.9  | 8.5   | 5.1   | 4.9   | 3.3   | 3.3   | 2.9   | 2.2   | 2.6   | 6.7   | 7.7   | 3.9   |
|                  | <i># Obs.</i> | 844  | 2,102 | 4,308 | 3,611 | 4,365 | 4,227 | 4,317 | 3,958 | 4,223 | 4,383 | 4,218 | 4,339 |
| Hoodspport (HS)  |               | 25.8 | 25.9  | 26.2  | 26.4  | 25.9  | 26.4  | 26.4  | 27.0  | 27.5  | 27.7  | 27.4  | 26.3  |
|                  | <i>StDev</i>  | 2.3  | 2.2   | 1.9   | 1.4   | 1.4   | 1.2   | 1.2   | 0.9   | 1.0   | 1.4   | 1.9   | 2.1   |
|                  | <i># Obs.</i> | 346  | 309   | 593   | 1,325 | 1,991 | 1,477 | 2,669 | 3,241 | 2,332 | 1,682 | 850   | 371   |
| Point Wells (PW) |               | 28.3 | 27.8  | 28.2  | 28.1  | 28.4  | 28.2  | 28.8  | 29.4  | 29.8  | 30.0  | 29.3  | 28.6  |
|                  | <i>StDev</i>  | 1.2  | 0.9   | 0.5   | 0.5   | 0.4   | 0.5   | 0.4   | 0.2   | 0.2   | 0.2   | 0.8   | 0.6   |
|                  | <i># Obs.</i> | 128  | 84    | 208   | 275   | 474   | 379   | 631   | 494   | 398   | 751   | 364   | 129   |
| North Buoy (NB)  |               | 28.9 | 28.9  | 29.1  | 29.2  | 29.1  | 29.2  | 29.6  | 29.9  | 30.0  | 30.3  | 29.9  | 29.4  |
|                  | <i>StDev</i>  | 0.6  | 0.5   | 0.4   | 0.2   | 0.3   | 0.3   | 0.3   | 0.2   | 0.2   | 0.3   | 0.5   | 0.5   |
|                  | <i># Obs.</i> | 329  | 474   | 349   | 578   | 884   | 972   | 1,476 | 1,734 | 858   | 1,320 | 693   | 532   |

798

799 **Table C2.** Average monthly SST seasonal cycles for the nine Washington State moorings. The  
800 average monthly standard deviation (*StDev*) and total number of observations (*# Obs.*)  
801 contributing to the monthly average estimate are included.  
802

| SST (°C)            | Jan.          | Feb.  | Mar.  | Apr.  | May   | June  | July  | Aug.  | Sept. | Oct.  | Nov.  | Dec.  |
|---------------------|---------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Chá bã (CB)         | 8.6           | 7.8   | 8.9   | 10.0  | 11.2  | 13.1  | 13.7  | 14.0  | 13.9  | 13.5  | 11.7  | 9.5   |
|                     | <i>StDev</i>  | 0.6   | 0.4   | 0.6   | 0.5   | 0.6   | 0.8   | 1.0   | 0.9   | 0.9   | 0.5   | 0.7   |
|                     | <i># Obs.</i> | 398   | 224   | 248   | 280   | 485   | 1,080 | 1,461 | 1,673 | 1,553 | 1,173 | 646   |
| Cape Elizabeth (CE) | 9.2           | 9.0   | 9.2   | 9.7   | 11.2  | 12.8  | 13.7  | 13.8  | 14.1  | 12.8  | 11.5  | 10.1  |
|                     | <i>StDev</i>  | 0.6   | 0.4   | 0.4   | 0.5   | 1.0   | 1.0   | 1.2   | 0.9   | 0.9   | 0.8   | 0.7   |
|                     | <i># Obs.</i> | 1,488 | 1,352 | 1,295 | 1,199 | 1,161 | 1,246 | 1,715 | 1,812 | 1,621 | 1,829 | 1,778 |
| Twanoh (TW)         | 8.6           | 8.8   | 9.6   | 11.2  | 13.8  | 16.1  | 18.4  | 18.4  | 16.0  | 12.5  | 10.2  | 8.9   |
|                     | <i>StDev</i>  | 0.8   | 0.7   | 0.6   | 1.0   | 1.4   | 1.6   | 1.8   | 1.6   | 1.8   | 1.3   | 0.8   |
|                     | <i># Obs.</i> | 684   | 913   | 1,443 | 2,678 | 3,332 | 3,234 | 3,169 | 3,664 | 3,993 | 3,220 | 1,545 |
| Dabob Bay (DB)      | 7.8           | 7.5   | 8.5   | 9.9   | 13.7  | 15.9  | 18.4  | 18.4  | 16.0  | 12.5  | 10.3  | 8.4   |
|                     | <i>StDev</i>  | 0.2   | 0.3   | 0.4   | 0.5   | 0.9   | 1.5   | 1.8   | 1.7   | 1.5   | 1.1   | 0.6   |
|                     | <i># Obs.</i> | 34    | 33    | 85    | 109   | 174   | 286   | 767   | 1,167 | 1,138 | 667   | 39    |
| Carr Inlet (CI)     | 8.7           | 8.5   | 8.7   | 10.0  | 11.8  | 13.3  | 14.7  | 15.4  | 14.7  | 13.2  | 11.8  | 9.8   |
|                     | <i>StDev</i>  | 0.2   | 0.2   | 0.4   | 0.7   | 1.0   | 1.4   | 1.3   | 1.3   | 0.9   | 0.5   | 0.4   |
|                     | <i># Obs.</i> | 349   | 347   | 1,008 | 937   | 820   | 1,161 | 1,629 | 1,641 | 1,611 | 1,566 | 716   |
| Bellingham Bay (BB) | 6.7           | 8.2   | 9.1   | 12.2  | 14.3  | 14.9  | 17.4  | 17.6  | 14.7  | 11.4  | 9.9   | 7.4   |
|                     | <i>StDev</i>  | 0.7   | 0.7   | 0.6   | 1.2   | 1.2   | 1.2   | 1.6   | 1.3   | 1.3   | 0.9   | 1.0   |
|                     | <i># Obs.</i> | 844   | 2,102 | 4,308 | 3,611 | 4,365 | 4,227 | 4,317 | 3,958 | 4,223 | 4,383 | 4,218 |
| Hoodspport (HS)     | 8.7           | 8.6   | 9.3   | 10.5  | 12.9  | 14.4  | 17.0  | 16.7  | 14.4  | 11.7  | 10.4  | 9.3   |
|                     | <i>StDev</i>  | 0.9   | 0.7   | 0.4   | 0.9   | 1.5   | 1.5   | 2.0   | 1.7   | 1.7   | 1.3   | 0.7   |
|                     | <i># Obs.</i> | 346   | 309   | 593   | 1,325 | 1,991 | 1,477 | 2,669 | 3,241 | 2,332 | 1,682 | 850   |
| Point Wells (PW)    | 8.1           | 8.2   | 8.5   | 9.6   | 10.9  | 12.1  | 13.2  | 13.6  | 13.1  | 12.6  | 11.3  | 9.3   |
|                     | <i>StDev</i>  | 0.3   | 0.3   | 0.2   | 0.3   | 0.6   | 0.7   | 0.6   | 0.5   | 0.3   | 0.3   | 0.6   |
|                     | <i># Obs.</i> | 128   | 84    | 208   | 275   | 474   | 379   | 631   | 494   | 398   | 751   | 364   |
| North Buoy (NB)     | 8.4           | 8.2   | 8.5   | 9.1   | 10.6  | 11.7  | 13.0  | 13.1  | 12.7  | 11.2  | 10.3  | 8.8   |
|                     | <i>StDev</i>  | 0.2   | 0.1   | 0.3   | 0.4   | 0.5   | 0.7   | 0.7   | 0.6   | 0.4   | 0.3   | 0.4   |
|                     | <i># Obs.</i> | 329   | 474   | 349   | 578   | 884   | 972   | 1,476 | 1,734 | 858   | 1,320 | 693   |

803



804 **Table C3.** Average monthly TA seasonal cycles for the eight Washington State moorings with  
 805 appropriate SSS observations (vast majority of SSS >20) to estimate TA. The average monthly  
 806 standard deviation (StDev) and total number of observations (# Obs.) contributing to the monthly  
 807 average estimate are included.  
 808

| TA ( $\mu\text{mol kg}^{-1}$ ) | Jan.  | Feb.  | Mar.  | Apr.  | May   | June  | July  | Aug.  | Sept. | Oct.  | Nov.  | Dec.  |
|--------------------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Chá bã (CB)                    | 2,090 | 2,108 | 2,105 | 2,102 | 2,114 | 2,132 | 2,148 | 2,165 | 2,167 | 2,161 | 2,162 | 2,142 |
| <i>StDev</i>                   | 59    | 58    | 77    | 54    | 20    | 55    | 40    | 12    | 25    | 22    | 22    | 34    |
| <i># Obs.</i>                  | 398   | 224   | 248   | 280   | 485   | 1,080 | 1,461 | 1,648 | 1,346 | 1,029 | 646   | 496   |
| Cape Elizabeth (CE)            | 2,130 | 2,110 | 2,118 | 2,094 | 2,101 | 2,135 | 2,156 | 2,173 | 2,168 | 2,162 | 2,164 | 2,149 |
| <i>StDev</i>                   | 62    | 73    | 67    | 60    | 73    | 53    | 32    | 14    | 24    | 24    | 28    | 38    |
| <i># Obs.</i>                  | 1,488 | 1,352 | 1,295 | 1,199 | 1,161 | 1,246 | 1,715 | 1,812 | 1,621 | 1,756 | 1,538 | 1,484 |
| Twanoh (TW)                    | 1,815 | 1,815 | 1,823 | 1,820 | 1,825 | 1,844 | 1,870 | 1,903 | 1,909 | 1,896 | 1,876 | 1,842 |
| <i>StDev</i>                   | 127   | 109   | 108   | 85    | 71    | 49    | 51    | 34    | 49    | 82    | 119   | 127   |
| <i># Obs.</i>                  | 684   | 913   | 1,443 | 2,678 | 3,332 | 3,234 | 3,169 | 3,664 | 3,993 | 3,220 | 1,545 | 591   |
| Dabob Bay (DB)                 | 2,037 | 1,996 | 1,964 | 1,922 | 1,915 | 1,916 | 1,895 | 1,945 | 1,997 | 2,022 | 1,980 | 2,012 |
| <i>StDev</i>                   | 6     | 29    | 39    | 24    | 24    | 39    | 49    | 41    | 16    | 22    | 47    | 45    |
| <i># Obs.</i>                  | 34    | 33    | 85    | 109   | 174   | 286   | 767   | 1,167 | 1,138 | 667   | 163   | 39    |
| Carr Inlet (CI)                | 2,019 | 2,007 | 1,992 | 1,988 | 1,991 | 2,002 | 2,020 | 2,033 | 2,047 | 2,060 | 2,064 | 2,036 |
| <i>StDev</i>                   | 11    | 8     | 11    | 7     | 7     | 7     | 7     | 5     | 7     | 8     | 7     | 11    |
| <i># Obs.</i>                  | 349   | 347   | 1,008 | 937   | 820   | 1,161 | 1,629 | 1,641 | 1,611 | 1,566 | 716   | 584   |
| Hoodspout (HS)                 | 1,890 | 1,891 | 1,897 | 1,904 | 1,881 | 1,904 | 1,904 | 1,932 | 1,957 | 1,968 | 1,959 | 1,907 |
| <i>StDev</i>                   | 93    | 86    | 82    | 67    | 69    | 55    | 55    | 44    | 48    | 65    | 80    | 91    |
| <i># Obs.</i>                  | 337   | 302   | 590   | 1,323 | 1,991 | 1,477 | 2,669 | 3,241 | 2,332 | 1,681 | 843   | 361   |
| Point Wells (PW)               | 1,995 | 1,972 | 1,992 | 1,986 | 1,998 | 1,989 | 2,020 | 2,050 | 2,066 | 2,078 | 2,042 | 2,010 |
| <i>StDev</i>                   | 58    | 41    | 25    | 22    | 20    | 25    | 19    | 12    | 7     | 11    | 40    | 28    |
| <i># Obs.</i>                  | 128   | 84    | 208   | 275   | 474   | 379   | 631   | 494   | 398   | 751   | 364   | 129   |
| North Buoy (NB)                | 2,026 | 2,023 | 2,033 | 2,038 | 2,036 | 2,038 | 2,055 | 2,073 | 2,076 | 2,090 | 2,072 | 2,047 |
| <i>StDev</i>                   | 29    | 22    | 19    | 12    | 15    | 16    | 15    | 12    | 12    | 15    | 22    | 25    |
| <i># Obs.</i>                  | 329   | 474   | 349   | 578   | 884   | 972   | 1,476 | 1,734 | 858   | 1,320 | 693   | 532   |

809

810 **Table C4.** Average monthly DIC seasonal cycles for the five Washington State moorings with  
 811 sufficient observations to estimate DIC. The average monthly standard deviation (StDev) and total  
 812 number of observations (# Obs.) contributing to the monthly average estimate are included.  
 813

| DIC ( $\mu\text{mol kg}^{-1}$ ) | Jan.  | Feb.  | Mar.  | Apr.  | May   | June  | July  | Aug.  | Sept. | Oct.  | Nov.  | Dec.  |
|---------------------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Chá bã (CB)                     | 1,936 | 1,964 | 1,973 | 1,882 | 1,846 | 1,850 | 1,891 | 1,922 | 1,950 | 1,960 | 1,981 | 1,975 |
| <i>StDev</i>                    | 47    | 54    | 72    | 56    | 34    | 66    | 57    | 41    | 36    | 25    | 29    | 33    |
| <i># Obs.</i>                   | 398   | 221   | 245   | 279   | 484   | 1,080 | 1,461 | 1,648 | 1,346 | 1,028 | 645   | 495   |
| Cape Elizabeth (CE)             | 1,975 | 1,949 | 1,944 | 1,898 | 1,891 | 1,893 | 1,915 | 1,925 | 1,944 | 1,958 | 1,986 | 1,987 |
| <i>StDev</i>                    | 54    | 70    | 64    | 57    | 71    | 54    | 47    | 41    | 49    | 41    | 34    | 34    |
| <i># Obs.</i>                   | 1,485 | 1,347 | 1,295 | 1,199 | 1,158 | 1,244 | 1,715 | 1,812 | 1,619 | 1,753 | 1,506 | 1,484 |
| Twanoh (TW)                     | 1,783 | 1,722 | 1,630 | 1,644 | 1,688 | 1,705 | 1,709 | 1,726 | 1,784 | 1,747 | 1,779 | 1,815 |
| <i>StDev</i>                    | 67    | 97    | 88    | 80    | 73    | 56    | 50    | 33    | 54    | 81    | 104   | 75    |
| <i># Obs.</i>                   | 371   | 392   | 506   | 725   | 875   | 877   | 743   | 961   | 1,650 | 1,145 | 613   | 363   |



|                 |       |       |       |       |       |       |       |       |       |       |       |       |
|-----------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Dabob Bay (DB)  | 1,991 | 1,929 | 1,828 | 1,728 |       | 1,716 | 1,677 | 1,720 | 1,788 | 1,867 | 1,931 | 1,904 |
| <i>StDev</i>    | 7     | 31    | 82    | 51    |       | 36    | 51    | 38    | 31    | 52    | 22    | 41    |
| <i># Obs.</i>   | 28    | 132   | 265   | 36    |       | 150   | 317   | 496   | 378   | 118   | 139   | 17    |
| Carr Inlet (CI) | 1,946 | 1,923 | 1,897 | 1,769 | 1,824 | 1,831 | 1,773 | 1,886 | 1,891 | 1,928 | 2,021 | 1,944 |
| <i>StDev</i>    | 20    | 18    | 31    | 68    | 48    | 36    | 97    | 68    | 72    | 89    | 16    | 18    |
| <i># Obs.</i>   | 51    | 42    | 112   | 130   | 140   | 56    | 228   | 372   | 360   | 268   | 74    | 15    |

814

815 **Table C5.** Average monthly  $f\text{CO}_2$  seasonal cycles for the five Washington State moorings with  
 816  $\text{CO}_2$  sensors or sufficient observations to estimate  $f\text{CO}_2$ . The average monthly standard deviation  
 817 (StDev) and total number of observations (*# Obs.*) contributing to the monthly average estimate  
 818 are included.

819

| $f\text{CO}_2$ ( $\mu\text{atm}$ ) | Jan.  | Feb.  | Mar.  | Apr.  | May   | June  | July  | Aug.  | Sept. | Oct.  | Nov.  | Dec.  |
|------------------------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Chá bá (CB)                        | 341   | 358   | 415   | 245   | 204   | 212   | 258   | 283   | 325   | 342   | 358   | 349   |
| <i>StDev</i>                       | 24    | 30    | 74    | 41    | 33    | 48    | 68    | 58    | 45    | 25    | 40    | 26    |
| <i># Obs.</i>                      | 398   | 221   | 245   | 279   | 484   | 1,080 | 1,461 | 1,648 | 1,346 | 1,028 | 645   | 495   |
| Cape Elizabeth (CE)                | 368   | 343   | 322   | 277   | 279   | 260   | 281   | 276   | 316   | 330   | 364   | 371   |
| <i>StDev</i>                       | 29    | 38    | 34    | 36    | 41    | 51    | 57    | 52    | 51    | 47    | 34    | 21    |
| <i># Obs.</i>                      | 1,485 | 1,347 | 1,295 | 1,199 | 1,158 | 1,244 | 1,715 | 1,812 | 1,619 | 1,753 | 1,506 | 1,484 |
| Twanoh (TW)                        | 619   | 427   | 231   | 233   | 346   | 383   | 342   | 344   | 392   | 379   | 487   | 655   |
| <i>StDev</i>                       | 144   | 201   | 103   | 88    | 58    | 75    | 34    | 30    | 64    | 145   | 263   | 217   |
| <i># Obs.</i>                      | 371   | 392   | 506   | 725   | 875   | 877   | 743   | 961   | 1,650 | 1,145 | 613   | 363   |
| Dabob Bay (DB)                     | 748   | 575   | 420   | 205   |       | 288   | 297   | 291   | 311   | 428   | 583   | 636   |
| <i>StDev</i>                       | 49    | 67    | 132   | 56    |       | 19    | 25    | 18    | 56    | 211   | 73    | 15    |
| <i># Obs.</i>                      | 28    | 132   | 265   | 36    |       | 150   | 317   | 496   | 378   | 118   | 139   | 17    |
| Carr Inlet (CI)                    | 731   | 679   | 602   | 261   | 351   | 377   | 264   | 450   | 420   | 502   | 843   | 659   |
| <i>StDev</i>                       | 64    | 67    | 137   | 133   | 84    | 84    | 167   | 177   | 184   | 241   | 69    | 57    |
| <i># Obs.</i>                      | 51    | 42    | 112   | 130   | 140   | 56    | 228   | 372   | 360   | 268   | 74    | 15    |

820

821 **Table C6.** Average monthly pH seasonal cycles for the six Washington State moorings with pH  
 822 sensors or sufficient observations to estimate pH. The average monthly standard deviation (StDev)  
 823 and total number of observations (*# Obs.*) contributing to the monthly average estimate are  
 824 included.

825

| pH                  | Jan.  | Feb.  | Mar.  | Apr.  | May   | June  | July  | Aug.  | Sept. | Oct.  | Nov.  | Dec.  |
|---------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Chá bá (CB)         | 8.07  | 8.08  | 8.02  | 8.22  | 8.27  | 8.26  | 8.21  | 8.17  | 8.11  | 8.09  | 8.06  | 8.07  |
| <i>StDev</i>        | 0.03  | 0.03  | 0.06  | 0.06  | 0.06  | 0.09  | 0.09  | 0.08  | 0.07  | 0.03  | 0.05  | 0.03  |
| <i># Obs.</i>       | 398   | 222   | 248   | 279   | 495   | 1,117 | 1,461 | 1,648 | 1,547 | 1,172 | 646   | 496   |
| Cape Elizabeth (CE) | 8.06  | 8.08  | 8.11  | 8.17  | 8.17  | 8.20  | 8.17  | 8.18  | 8.13  | 8.11  | 8.07  | 8.06  |
| <i>StDev</i>        | 0.03  | 0.04  | 0.04  | 0.05  | 0.05  | 0.07  | 0.07  | 0.07  | 0.06  | 0.06  | 0.04  | 0.02  |
| <i># Obs.</i>       | 1,485 | 1,347 | 1,295 | 1,199 | 1,158 | 1,244 | 1,715 | 1,812 | 1,619 | 1,753 | 1,506 | 1,484 |
| Twanoh (TW)         | 7.84  | 8.03  | 8.22  | 8.23  | 8.06  | 8.02  | 8.07  | 8.06  | 8.02  | 8.04  | 7.98  | 7.86  |
| <i>StDev</i>        | 0.14  | 0.22  | 0.16  | 0.13  | 0.06  | 0.06  | 0.04  | 0.03  | 0.06  | 0.13  | 0.22  | 0.11  |
| <i># Obs.</i>       | 371   | 392   | 506   | 725   | 875   | 877   | 743   | 961   | 1,650 | 1,145 | 613   | 363   |
| Dabob Bay (DB)      | 7.76  | 7.86  | 8.01  | 8.26  |       | 8.13  | 8.11  | 8.12  | 8.11  | 8.02  | 7.87  | 7.81  |
| <i>StDev</i>        | 0.03  | 0.04  | 0.14  | 0.08  |       | 0.02  | 0.03  | 0.02  | 0.05  | 0.13  | 0.06  | 0.02  |
| <i># Obs.</i>       | 28    | 132   | 265   | 36    |       | 150   | 317   | 496   | 378   | 118   | 139   | 17    |



|                     |      |       |       |       |       |       |       |       |       |       |       |       |
|---------------------|------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Carr Inlet (CI)     | 7.77 | 7.80  | 7.86  | 8.21  | 8.09  | 8.04  | 8.22  | 8.00  | 8.03  | 7.98  | 7.73  | 7.81  |
| <i>StDev</i>        | 0.03 | 0.04  | 0.09  | 0.16  | 0.10  | 0.09  | 0.20  | 0.15  | 0.16  | 0.21  | 0.03  | 0.03  |
| <i># Obs.</i>       | 51   | 42    | 112   | 130   | 140   | 56    | 228   | 372   | 360   | 268   | 74    | 15    |
| Bellingham Bay (BB) | 8.07 | 7.81  | 7.95  | 8.12  | 8.25  | 8.12  | 7.99  | 7.99  | 8.16  | 7.99  | 7.94  | 8.06  |
| <i>StDev</i>        | 0.03 | 0.08  | 0.09  | 0.20  | 0.10  | 0.12  | 0.19  | 0.17  | 0.07  | 0.11  | 0.09  | 0.05  |
| <i># Obs.</i>       | 844  | 2,114 | 4,377 | 4,168 | 4,367 | 4,227 | 4,318 | 3,963 | 4,226 | 4,385 | 4,229 | 4,365 |

826

827 **Table C7.** Average monthly  $\Omega_{Ar}$  seasonal cycles for the five Washington State moorings with  
828 sufficient observations to estimate  $\Omega_{Ar}$ . The average monthly standard deviation (*StDev*) and total  
829 number of observations (*# Obs.*) contributing to the monthly average estimate are included.

830

| $\Omega_{Ar}$       | Jan.  | Feb.  | Mar.  | Apr.  | May   | June  | July  | Aug.  | Sept. | Oct.  | Nov.  | Dec.  |
|---------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Chá bã (CB)         | 1.7   | 1.6   | 1.5   | 2.4   | 2.9   | 3.0   | 2.8   | 2.7   | 2.4   | 2.2   | 2.0   | 1.9   |
| <i>StDev</i>        | 0.2   | 0.1   | 0.2   | 0.3   | 0.3   | 0.5   | 0.5   | 0.4   | 0.3   | 0.1   | 0.2   | 0.1   |
| <i># Obs.</i>       | 398   | 221   | 245   | 279   | 484   | 1,080 | 1,461 | 1,648 | 1,346 | 1,028 | 645   | 495   |
| Cape Elizabeth (CE) | 1.8   | 1.8   | 1.9   | 2.1   | 2.3   | 2.6   | 2.6   | 2.7   | 2.5   | 2.3   | 2.0   | 1.8   |
| <i>StDev</i>        | 0.2   | 0.2   | 0.2   | 0.2   | 0.3   | 0.4   | 0.4   | 0.4   | 0.3   | 0.3   | 0.2   | 0.1   |
| <i># Obs.</i>       | 1,485 | 1,347 | 1,295 | 1,199 | 1,158 | 1,244 | 1,715 | 1,812 | 1,619 | 1,753 | 1,506 | 1,484 |
| Twanoh (TW)         | 0.9   | 1.4   | 1.9   | 2.1   | 1.6   | 1.6   | 2.0   | 2.0   | 1.7   | 1.5   | 1.3   | 1.0   |
| <i>StDev</i>        | 0.5   | 0.7   | 0.6   | 0.5   | 0.2   | 0.2   | 0.1   | 0.1   | 0.2   | 0.5   | 0.6   | 0.3   |
| <i># Obs.</i>       | 371   | 392   | 506   | 725   | 875   | 877   | 743   | 961   | 1,650 | 1,145 | 613   | 363   |
| Dabob Bay (DB)      | 0.8   | 1.0   | 1.3   | 2.1   |       | 2.0   | 2.1   | 2.2   | 2.2   | 1.7   | 1.1   | 0.8   |
| <i>StDev</i>        | 0.0   | 0.1   | 0.5   | 0.2   |       | 0.1   | 0.1   | 0.1   | 0.2   | 0.4   | 0.1   | 0.1   |
| <i># Obs.</i>       | 28    | 132   | 265   | 36    |       | 150   | 317   | 496   | 378   | 118   | 139   | 17    |
| Carr Inlet (CI)     | 0.9   | 0.9   | 1.0   | 2.3   | 1.9   | 1.8   | 3.0   | 2.0   | 2.1   | 1.9   | 1.0   | 1.0   |
| <i>StDev</i>        | 0.1   | 0.1   | 0.2   | 0.6   | 0.4   | 0.3   | 1.0   | 0.7   | 0.7   | 0.8   | 0.1   | 0.0   |
| <i># Obs.</i>       | 51    | 42    | 112   | 130   | 140   | 56    | 228   | 372   | 360   | 268   | 74    | 15    |

831

832 **Table C8.** Average monthly Revelle Factor seasonal cycles for the five Washington State  
833 moorings with sufficient observations to estimate the Revelle Factor. The average monthly  
834 standard deviation (*StDev*) and total number of observations (*# Obs.*) contributing to the monthly  
835 average estimate are included.

836

| Revelle Factor      | Jan.  | Feb.  | Mar.  | Apr.  | May   | June  | July  | Aug.  | Sept. | Oct.  | Nov.  | Dec.  |
|---------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Chá bã (CB)         | 13.4  | 13.9  | 14.4  | 11.2  | 10.1  | 9.9   | 10.5  | 10.7  | 11.4  | 11.8  | 12.5  | 13.0  |
| <i>StDev</i>        | 0.6   | 0.4   | 1.1   | 0.8   | 0.6   | 0.9   | 1.1   | 1.0   | 0.8   | 0.4   | 0.6   | 0.4   |
| <i># Obs.</i>       | 398   | 221   | 245   | 279   | 484   | 1,080 | 1,461 | 1,648 | 1,346 | 1,028 | 645   | 495   |
| Cape Elizabeth (CE) | 13.5  | 13.2  | 12.7  | 11.9  | 11.5  | 10.8  | 10.8  | 10.7  | 11.2  | 11.7  | 12.6  | 13.2  |
| <i>StDev</i>        | 0.6   | 0.6   | 0.5   | 0.7   | 0.7   | 1.0   | 0.9   | 0.9   | 0.8   | 0.8   | 0.6   | 0.4   |
| <i># Obs.</i>       | 1,485 | 1,347 | 1,295 | 1,199 | 1,158 | 1,244 | 1,715 | 1,812 | 1,619 | 1,753 | 1,506 | 1,484 |
| Twanoh (TW)         | 18.1  | 15.5  | 12.5  | 11.9  | 13.6  | 13.5  | 12.1  | 12.1  | 13.4  | 14.1  | 15.4  | 17.1  |
| <i>StDev</i>        | 1.8   | 3.0   | 2.3   | 1.8   | 0.9   | 1.0   | 0.5   | 0.4   | 1.0   | 2.0   | 2.9   | 1.2   |
| <i># Obs.</i>       | 371   | 392   | 506   | 725   | 875   | 877   | 743   | 961   | 1,650 | 1,145 | 613   | 363   |
| Dabob Bay (DB)      | 18.7  | 17.6  | 15.5  | 11.5  |       | 11.9  | 11.6  | 11.2  | 11.5  | 13.5  | 16.7  | 18.4  |
| <i>StDev</i>        | 0.2   | 0.5   | 2.1   | 1.0   |       | 0.3   | 0.4   | 0.3   | 0.8   | 1.7   | 0.9   | 0.6   |
| <i># Obs.</i>       | 28    | 132   | 265   | 36    |       | 150   | 317   | 496   | 378   | 118   | 139   | 17    |

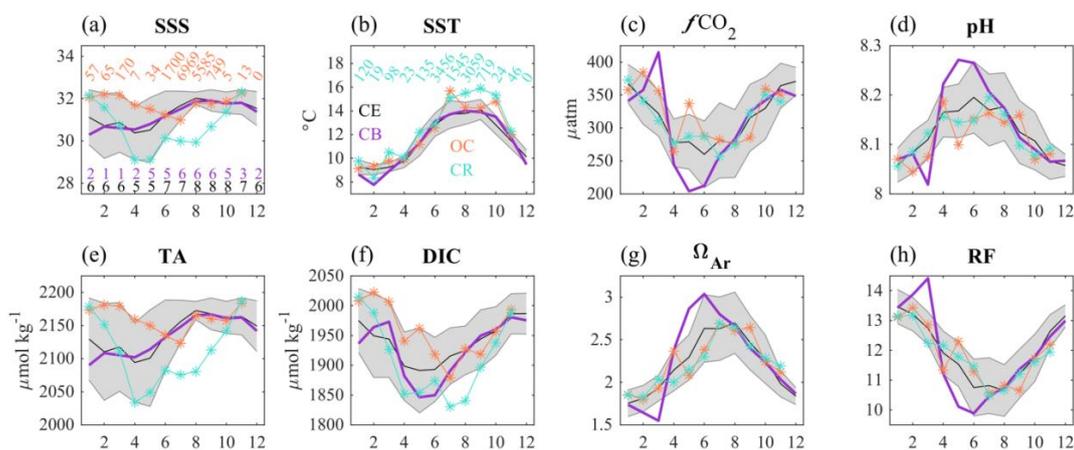


|                 |      |      |      |      |      |      |      |      |      |      |      |      |
|-----------------|------|------|------|------|------|------|------|------|------|------|------|------|
| Carr Inlet (CI) | 18.3 | 18.0 | 17.1 | 11.6 | 13.1 | 13.1 | 10.3 | 12.8 | 12.6 | 13.6 | 17.6 | 17.7 |
| <i>StDev</i>    | 0.3  | 0.5  | 1.3  | 2.3  | 1.6  | 1.5  | 2.4  | 2.2  | 2.4  | 2.9  | 0.3  | 0.3  |
| <i># Obs.</i>   | 51   | 42   | 112  | 130  | 140  | 56   | 228  | 372  | 360  | 268  | 74   | 15   |

837

838 **9. Appendix D**

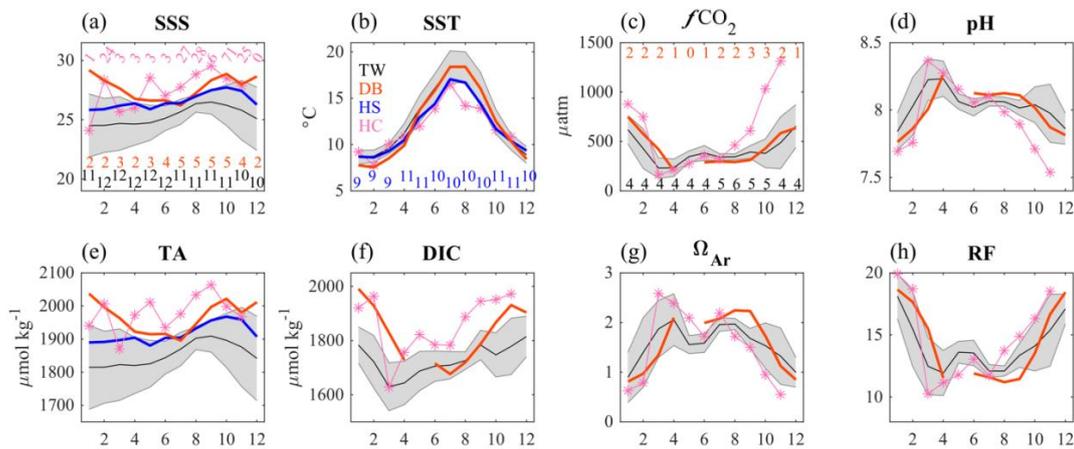
839



840

841 **Figure D1.** Seasonal cycles of (a) SSS, (b) SST, (c)  $f\text{CO}_2$ , (d)  $\text{pH}_T$ , (e) TA, (f) DIC, (g)  $\Omega_{Ar}$ , and  
 842 (h) RF at the Cape Elizabeth (CE) and Chá bã (CB) moorings, and for the Outer Coast (OC) and  
 843 Columbia River (CR) regions. Gray shading shows  $\pm 1\sigma$  for CE. Text near the bottom of a indicates  
 844 the number of years of moored SSS, SST, and  $f\text{CO}_2$  observations contributing to the monthly  
 845 seasonal cycle estimates. Slanted text in a and b indicates the total number of discrete observations  
 846 contributing to the monthly seasonal cycle estimates.

847

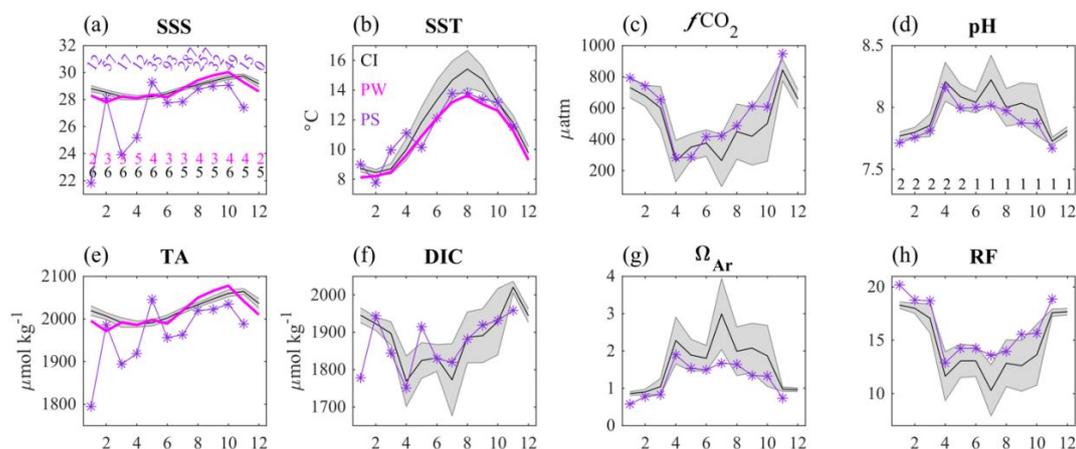


848

849 **Figure D2.** Seasonal cycles of (a) SSS, (b) SST, (c)  $f\text{CO}_2$ , (d)  $\text{pH}_T$ , (e) TA, (f) DIC, (g)  $\Omega_{Ar}$ , and  
 850 (h) RF at the Twanoh (TW), Dabob Bay (DB), and Hoodspport (HP) moorings and for the Hood



851 Canal (HC) region. Gray shading shows  $\pm 1\sigma$  for TW. Text near the bottom of **a** and **b** indicates  
 852 the number of years of moored SSS and SST observations contributing to the monthly seasonal  
 853 cycle estimates. Text in **c** indicates the same information but for the moored  $f\text{CO}_2$  observations.  
 854 Text is staggered in **a** and **b** for viewing. Slanted text in **a** indicates the total number of discrete  
 855 observations contributing to the monthly seasonal cycle estimates.  
 856



857

858 **Figure D3.** Seasonal cycles of **(a)** SSS, **(b)** SST, **(c)**  $f\text{CO}_2$ , **(d)**  $\text{pH}_T$ , **(e)** TA, **(f)** DIC, **(g)**  $\Omega_{\text{Ar}}$ , and  
 859 **(h)** RF at the Carr Inlet (CI) and Point Wells (PW) moorings and for the Puget Sound (PS) region.  
 860 Gray shading shows  $\pm 1\sigma$  for CI. Text near the bottom of **a** indicates the number of years of moored  
 861 SSS and SST observations contributing to the monthly seasonal cycle estimates. Text in **d** indicates  
 862 the same information but for the moored pH observations. Slanted text in **a** indicates the total  
 863 number of discrete observations contributing to the monthly seasonal cycle estimates.  
 864

864

## 865 10. Author contributions

866 A. Fassbender, S. Alin, R. Feely, and A. Sutton designed the study and contributed to the data  
 867 analysis and writing of the manuscript. J. Newton and W. Ruef provided much of the mooring and  
 868 cruise data and contributed to interpretation of the results and manuscript review. A. Devol  
 869 contributed to interpretation of the results and manuscript review. M. Keyzers wrote the sampling  
 870 plan and collected and provided data for ECY discrete samples. J. Bos, C. Krembs, and G. Pelletier  
 871 reviewed ECY data results, analyses, and the manuscript, and contributed to interpretation of the  
 872 results.

873

## 874 11. Acknowledgements

875 The Surface Ocean  $\text{CO}_2$  Atlas (SOCAT) is an international effort, endorsed by the International  
 876 Ocean Carbon Coordination Project (IOCCP), the Surface Ocean Lower Atmosphere Study  
 877 (SOLAS) and the Integrated Marine Biogeochemistry and Ecosystem Research program  
 878 (IMBER), to deliver a uniformly quality-controlled surface ocean  $\text{CO}_2$  database. The many  
 879 researchers and funding agencies responsible for the collection of data and quality control are  
 880 thanked for their contributions to SOCAT. The moored carbon observations were supported by  
 881 NOAA's Ocean Acidification Program and PMEL (NOAA FundRef 100000192). The University



882 of Washington acknowledges that the mooring work from Puget Sound and Chá bã assets were  
883 supported by numerous technicians, captains/crew, and scientists, including Al Devol, John  
884 Mickett, and Beth Curry. Cruises were often staffed by trained volunteer students through the  
885 PRISM, NANOOS, and WOAC programs. Funding was provided by the U.S. Integrated Ocean  
886 Observing System through NOAA to NANOOS; NOAA’s Ocean Acidification Program; the State  
887 of Washington to the Washington Ocean Acidification Center (WOAC); the Navy to the Hood  
888 Canal Dissolved Oxygen Program; the University of Washington to PRISM and for Bellingham  
889 Bay; and EPA. The Washington Dept. of Ecology (Christopher Krembs, Julia Bos, Mya Keyzers,  
890 Skip Albertson, Laura Hermanson, and Carol Maloy) conducts long-term monitoring of Puget  
891 Sound including Hood Canal, Strait of Juan de Fuca, and Washington’s coastal bays, collecting  
892 and providing quality-assured marine data since 1973. A.J.F. was supported by the Postdocs  
893 Applying Climate Expertise (PACE) Fellowship Program, partially funded by the WOAC and  
894 NOAA Climate Program Office and administered by the UCAR Visiting Scientist Programs. This  
895 is PMEL contribution number 4632.

896

897 **12. References**

- 898 Adelman, H. and Binder, L. W., Eds.: Washington State Blue Ribbon Panel on Ocean  
899 Acidification (2012): *Ocean Acidification: From Knowledge to Action, Washington State’s*  
900 *Strategic Response*, Washington Department of Ecology, Olympia, Washington., 2012.
- 901 Alford, M. H. and MacCready, P.: Flow and mixing in Juan de Fuca Canyon, Washington,  
902 *Geophys. Res. Lett.*, 41(5), 1608–1615, doi:10.1002/2013GL058967, 2014.
- 903 Alin, S. R., Brainard, R., Price, N., Newton, J. A., Cohen, A., Peterson, W., DeCarlo, E.,  
904 Shadwick, E., Noakes, S. and Bednaršek, N.: Characterizing the Natural System: Toward  
905 Sustained, Integrated Coastal Ocean Acidification Observing Networks to Facilitate Resource  
906 Management and Decision Support, *Oceanography*, 25(2), 92–107,  
907 doi:10.5670/oceanog.2015.34, 2015.
- 908 Andersson, A. J. and MacKenzie, F. T.: Revisiting four scientific debates in ocean acidification  
909 research, *Biogeosciences*, 9(3), 893–905, doi:10.5194/bg-9-893-2012, 2012.
- 910 Bakker, D. C. E., Pfeil, B., Landa, C. S., Metzl, N., O&apos;Brien, K. M., Olsen, A., Smith,  
911 K., Cosca, C., Harasawa, S., Jones, S. D., Nakaoka, S., Nojiri, Y., Schuster, U., Steinhoff, T.,  
912 Sweeney, C., Takahashi, T., Tilbrook, B., Wada, C., Wanninkhof, R., Alin, S. R., Balestrini, C.  
913 F., Barbero, L., Bates, N. R., Bianchi, A. A., Bonou, F., Boutin, J., Bozec, Y., Burger, E. F., Cai,  
914 W.-J., Castle, R. D., Chen, L., Chierici, M., Currie, K., Evans, W., Featherstone, C., Feely, R. A.,  
915 Fransson, A., Goyet, C., Greenwood, N., Gregor, L., Hankin, S., Hardman-Mountford, N. J.,  
916 Harlay, J., Hauck, J., Hoppema, M., Humphreys, M. P., Hunt, C. W., Huss, B., Ibáñez, J. S. P.,  
917 Johannessen, T., Keeling, R., Kitidis, V., Körtzinger, A., Kozyr, A., Krasakopoulou, E., Kuwata,  
918 A., Landschützer, P., Lauvset, S. K., Lefèvre, N., Lo Monaco, C., Manke, A., Mathis, J. T.,  
919 Merlivat, L., Millero, F. J., Monteiro, P. M. S., Munro, D. R., Murata, A., Newberger, T., Omar,  
920 A. M., Ono, T., Paterson, K., Pearce, D., Pierrot, D., Robbins, L. L., Saito, S., Salisbury, J. E.,  
921 Schlitzer, R., Schneider, B., Schweitzer, R., Sieger, R., Skjelvan, I., Sullivan, K. F., Sutherland,  
922 S. C., Sutton, A. J., Tadokoro, K., Telszewski, M., Tuma, M., van Heuven, S. M. A. C.,  
923 Vandemark, D. C., Ward, B., Watson, A. J. and Xu, S.: A multi-decade record of high-quality  
924  $f\text{CO}_2$  data in version 3 of the Surface Ocean  $\text{CO}_2$  Atlas (SOCAT), *Earth Syst. Sci. Data*, 8(2),  
925 383–413, doi:10.5194/essd-8-383-2016, 2016.



- 926 Barton, A., Hales, B., Waldbusser, G. G., Langdon, C. and Feely, R. A.: The Pacific oyster,  
927 *Crassostrea gigas*, shows negative correlation to naturally elevated carbon dioxide levels:  
928 Implications for near-term ocean acidification effects, *Limnol. Oceanogr.*, 57(3), 698–710,  
929 doi:10.4319/lo.2012.57.3.0698, 2012.
- 930 Barton, A., Waldbusser, G. G., Feely, R. A., Weisberg, S. B., Newton, J. A., Hales, B., Cudd, S.,  
931 Eudeline, B., Langdon, C., Jefferds, I., King, T., Suhrbier, A. and McLaughlin, K.: Impacts of  
932 Coastal Acidification on the Pacific Northwest Shellfish Industry and Adaptation Strategies  
933 Implemented in Response, *Oceanography*, 25(2), 146–159, doi:10.5670/oceanog.2015.38, 2015.
- 934 Bates, N., Astor, Y., Church, M. J., Currie, K., Dore, J., Gonaález-Dávila, M., Lorenzoni, L.,  
935 Muller-Karger, F., Olafsson, J. and Santana-Casiano, J. M.: A time-series view of changing  
936 ocean chemistry due to ocean uptake of anthropogenic CO<sub>2</sub> and ocean acidification,  
937 *Oceanography*, 27(1), 126–141, doi:10.5670/oceanog.2014.16, 2014.
- 938 Bauer, J. E., Cai, W.-J., Raymond, P. A., Bianchi, T. S., Hopkinson, C. S. and Regnier, P. A. G.:  
939 The changing carbon cycle of the coastal ocean, *Nature*, 504(7478), 61–70,  
940 doi:10.1038/nature12857, 2013.
- 941 Bednaršek, N., Feely, R. A., Reum, J. C. P., Peterson, B., Menkel, J., Alin, S. R. and Hales, B.:  
942 *Limacina helicina* shell dissolution as an indicator of declining habitat suitability owing to ocean  
943 acidification in the California Current Ecosystem, *Proc. R. Soc. B Biol. Sci.*, 281(1785),  
944 20140123–20140123, doi:10.1098/rspb.2014.0123, 2014.
- 945 Bednaršek, N., Klinger, T., Harvey, C. J., Weisberg, S. B., McCabe, R. M., Feely, R. A.,  
946 Newton, J. and Tolimieri, N.: New ocean, new needs: Application of pteropod shell dissolution  
947 as a biological indicator for marine resource management, *Ecol. Indic.*, 76, 240–244,  
948 doi:10.1016/j.ecolind.2017.01.025, 2017.
- 949 Boehm, A. B. A., Jacobson, M. Z. M., O'Donnell, M., Sutula, M., Wakefield, W. W., Weisberg,  
950 S. B. and Whiteman, E.: Ocean Acidification Science Needs for Natural Resource Managers of  
951 the North American West Coast, *Oceanography*, 25(2), 170–181, doi:10.5670/oceanog.2015.40,  
952 2015.
- 953 Borges, A. V.: Present Day Carbon Dioxide Fluxes in the Coastal Ocean and Possible Feedbacks  
954 Under Global Change, in *Oceans and the Atmospheric Carbon Content*, edited by P. Duarte and  
955 J. M. Santana-Casiano, pp. 47–77, Springer Netherlands, Dordrecht., 2011.
- 956 Bos, J., Krembs, C. and Albertson, S.: Quality Assurance for Long-Term Marine Water Column  
957 pH Data. 16-03-042. [online] Available from:  
958 <https://fortress.wa.gov/ecy/publications/SummaryPages/1603042.html>, 2016.
- 959 Bresnahan, P. J., Martz, T. R., Takeshita, Y., Johnson, K. S. and LaShomb, M.: Best practices for  
960 autonomous measurement of seawater pH with the Honeywell Durafet, *Methods Oceanogr.*,  
961 9(October), 44–60, doi:10.1016/j.mio.2014.08.003, 2014.
- 962 Brewer, P. G.: Direct Observation of the Oceanic CO<sub>2</sub> Increase, *Geophys. Res. Lett.*, 5(12), 0–3,  
963 1978.
- 964 Broecker, W. S., Takahashi, T., Simpson, H. J. and Peng, T.-H.: Fate of fossil fuel carbon  
965 dioxide and the global carbon budget., *Science*, 206(4417), 409–18,  
966 doi:10.1126/science.206.4417.409, 1979.



- 967 Byrne, R. H., Mecking, S., Feely, R. A. and Liu, X.: Direct observations of basin-wide  
968 acidification of the North Pacific Ocean, *Geophys. Res. Lett.*, 37(2), 1–5,  
969 doi:10.1029/2009GL040999, 2010.
- 970 Cai, W.-J., Hu, X., Huang, W.-J., Murrell, M. C., Lehrter, J. C., Lohrenz, S. E., Chou, W.-C.,  
971 Zhai, W., Hollibaugh, J. T., Wang, Y., Zhao, P., Guo, X., Gundersen, K., Dai, M. and Gong, G.-  
972 C.: Acidification of subsurface coastal waters enhanced by eutrophication, *Nat. Geosci.*, 4(11),  
973 766–770, doi:10.1038/ngeo1297, 2011.
- 974 Caldeira, K. and Wickett, M. E.: Anthropogenic Carbon and Ocean pH, *Nature*, 425(September),  
975 365–365, doi:10.1038/425365a, 2003.
- 976 Dickson, A. G.: An exact definition of total alkalinity and a procedure for the estimation of  
977 alkalinity and total inorganic carbon from titration data, *Deep Sea Res. Part A. Oceanogr. Res.*  
978 *Pap.*, 28(6), 609–623, doi:10.1016/0198-0149(81)90121-7, 1981.
- 979 Dickson, A. G.: Standard potential of the reaction :  $\text{AgCl(s)} + 1/2\text{H}_2(\text{g}) = \text{Ag(s)} + \text{HCl(aq)}$ , and  
980 the standard acidity constant of the ion  $\text{HSO}_4^-$  in synthetic sea water from 273.15 to 318.15 K, *J.*  
981 *Chem. Thermodyn.*, 22, 113–127, 1990.
- 982 Dickson, A. G. and Riley, J.: The effect of analytical error on the evaluation of the components  
983 of the aquatic carbon-dioxide system, *Mar. Chem.*, 6, 77–85, doi:10.1016/0304-4203(78)90008-  
984 7, 1978.
- 985 Dickson, A. G., Sabine, C. L. and Christian, J. R., Eds.: Guide to best practices for ocean CO<sub>2</sub>  
986 measurements, PICES Special Publication 3, 191 pp. [online] Available from:  
987 [http://cdiac.ornl.gov/oceans/Handbook\\_2007.html](http://cdiac.ornl.gov/oceans/Handbook_2007.html), 2007.
- 988 Doney, S. C., Fabry, V. J., Feely, R. A. and Kleypas, J. A.: Ocean acidification: the other CO<sub>2</sub>  
989 problem., *Ann. Rev. Mar. Sci.*, 1(1), 169–192, doi:10.1146/annurev.marine.010908.163834,  
990 2009.
- 991 Dunne, J. P., Devol, A. H. and Emerson, S.: The Oceanic Remote Chemical/Optical Analyzer  
992 (ORCA)—An Autonomous Moored Profiler, *J. Atmos. Ocean. Technol.*, 19(10), 1709–1721,  
993 doi:10.1175/1520-0426(2002)019<1709:TORCOA>2.0.CO;2, 2002.
- 994 Ekstrom, J. A., Suatoni, L., Cooley, S. R., Pendleton, L. H., Waldbusser, G. G., Cinner, J. E.,  
995 Ritter, J., Langdon, C., van Hooijdonk, R., Gledhill, D. K., Wellman, K., Beck, M. W., Brander,  
996 L. M., Rittschof, D., Doherty, C., Edwards, P. E. T. and Portela, R.: Vulnerability and adaptation  
997 of US shellfisheries to ocean acidification, *Nat. Clim. Chang.*, 5(3), 207–214,  
998 doi:10.1038/nclimate2508, 2015.
- 999 Evans, W., Hales, B. and Stratton, P. G.: Seasonal cycle of surface ocean pCO<sub>2</sub> on the Oregon  
1000 shelf, *J. Geophys. Res.*, 116, doi:10.1029/2010JC006625, 2011.
- 1001 Fassbender, A. J., Sabine, C. L., Lawrence-Slavas, N., De Carlo, E. H. H., Meinig, C. and  
1002 Maenner-Jones, S.: Robust Sensor for Extended Autonomous Measurements of Surface Ocean  
1003 Dissolved Inorganic Carbon, *Environ. Sci. Technol.*, 49(6), 3628–3635, doi:10.1021/es5047183,  
1004 2015.
- 1005 Fassbender, A. J., Sabine, C. L. and Feifel, K. M.: Consideration of coastal carbonate chemistry  
1006 in understanding biological calcification, *Geophys. Res. Lett.*, 43(9), 4467–4476,  
1007 doi:10.1002/2016GL068860, 2016.



- 1008 Fassbender, A. J., Alin, S. R., Feely, R. A., Sutton, A. J., Newton, J. A. J. A. and Byrne, R. H. :  
1009 Estimating Total Alkalinity in the Washington State Coastal Zone: Complexities and Surprising  
1010 Utility for Ocean Acidification Research, *Estuaries and Coasts*, 40(2), 404–418,  
1011 doi:10.1007/s12237-016-0168-z, 2017a.
- 1012 Fassbender, A. J., Sabine, C. L. and Palevsky, H. I.: Nonuniform ocean acidification and  
1013 attenuation of the ocean carbon sink, *Geophys. Res. Lett.*, 44(16), 8404–8413,  
1014 doi:10.1002/2017GL074389, 2017b.
- 1015 Feely, R. A., Sabine, C. L., Lee, K., Berelson, W. M., Kleypas, J. A., Fabry, V. J. and Millero, F.  
1016 J.: Impact of anthropogenic CO<sub>2</sub> on the CaCO<sub>3</sub> system in the oceans., *Science* (80-. ),  
1017 305(5682), 362–6, doi:10.1126/science.1097329, 2004.
- 1018 Feely, R. A., Sabine, C. L., Hernandez-Ayon, J. M., Ianson, D. and Hales, B.: Evidence for  
1019 Upwelling of Corrosive “Acidified” Water onto the Continental Shelf, *Science* (80-. ),  
1020 320(5882), 1490–1492, doi:10.1126/science.1155676, 2008.
- 1021 Feely, R. A., Doney, S. C. and Cooley, S. R.: Ocean Acidification: Present Conditions and  
1022 Future Changes in a High-CO<sub>2</sub> World, *Oceanography*, 22(4), 36–47,  
1023 doi:10.5670/oceanog.2009.95, 2009.
- 1024 Feely, R. A., Alin, S. R., Newton, J. A., Sabine, C. L., Warner, M. J., Devol, A., Krembs, C. and  
1025 Maloy, C.: The combined effects of ocean acidification, mixing, and respiration on pH and  
1026 carbonate saturation in an urbanized estuary, *Estuar. Coast. Shelf Sci.*, 88(4), 442–449,  
1027 doi:10.1016/j.ecss.2010.05.004, 2010.
- 1028 Feely, R. A., Alin, S. R., Carter, B., Bednaršek, N., Hales, B., Chan, F., Hill, T. M., Gaylord, B.,  
1029 Sanford, E., Byrne, R. H., Sabine, C. L., Greeley, D. and Juraneck, L.: Chemical and biological  
1030 impacts of ocean acidification along the west coast of North America, *Estuar. Coast. Shelf Sci.*,  
1031 183, 260–270, doi:10.1016/j.ecss.2016.08.043, 2016.
- 1032 Gray, S. E. C., DeGrandpre, M. D., Moore, T. S., Martz, T. R., Friederich, G. E. and Johnson, K.  
1033 S.: Applications of in situ pH measurements for inorganic carbon calculations, *Mar. Chem.*, 125,  
1034 82–90, doi:10.1016/j.marchem.2011.02.005, 2011.
- 1035 Harris, K.: Aragonite saturation state dynamics in a coastal upwelling zone, *Geophys. Res. ...*,  
1036 40(April), 2720–2725, doi:10.1002/grl.50460, 2013.
- 1037 Hauri, C., Gruber, N., Vogt, M., Doney, S. C., Feely, R. A., Lachkar, Z., Leinweber, a.,  
1038 McDonnell, a. M. P. and Munnich, M.: Spatiotemporal variability and long-term trends of ocean  
1039 acidification in the California Current System, *Biogeosciences*, 10(1), 193–216, doi:10.5194/bg-  
1040 10-193-2013, 2013.
- 1041 van Heuven, S. M. A. C., Pierrot, D., Rae, J. W. B., Lewis, E. and Wallace, D. W. : MATLAB  
1042 Program Developed for CO<sub>2</sub> System Calculations. ORNL/CDIAC-105b., ORNL/CDIAC-105b.  
1043 Carbon Dioxide Inf. Anal. Center, Oak Ridge Natl. Lab. U.S. Dep. Energy, Oak Ridge,  
1044 Tennessee, doi:10.3334/CDIAC/otg.CO2SYS\_MATLAB\_v1.1, 2011.
- 1045 Hofmann, G. E., Smith, J. E., Johnson, K. S., Send, U., Levin, L. A., Micheli, F., Paytan, A.,  
1046 Price, N. N., Peterson, B., Takeshita, Y., Matson, P. G., Crook, E. D., Kroeker, K. J., Gambi, M.  
1047 C., Rivest, E. B., Frieder, C. A., Yu, P. C. and Martz, T. R.: High-Frequency Dynamics of Ocean  
1048 pH: A Multi-Ecosystem Comparison, edited by W.-C. Chin, *PLoS One*, 6(12), e28983,  
1049 doi:10.1371/journal.pone.0028983, 2011.



- 1050 Johnson, K. M., Dickson, A. G., Eiseid, G., Goyet, C., Guenther, P., Key, R. M., Millero, F. J.,  
1051 Purkerson, D., Sabine, C. L., Schottle, R. G., Wallace, D. W. R., Wilke, R. J. and Winn, C. D.:  
1052 Coulometric total carbon dioxide analysis for marine studies: assessment of the quality of total  
1053 inorganic carbon measurements made during the US Indian Ocean CO<sub>2</sub> Survey 1994–1996, *Mar.*  
1054 *Chem.*, 63(1–2), 21–37, doi:10.1016/S0304-4203(98)00048-6, 1998.
- 1055 Keyzers, M.: Quality Assurance Project Plan: Puget Sound Total Alkalinity and Dissolved  
1056 Inorganic Carbon Pilot Project, Olympia, Washington. [online] Available from:  
1057 <https://fortress.wa.gov/ecy/publications/documents/1403116.pdf>, 2014.
- 1058 Keyzers, M.: Puget Sound Total Alkalinity and Dissolved Inorganic Carbon Pilot Project:  
1059 Feasibility Report, Olympia, Washington. [online] Available from:  
1060 <https://fortress.wa.gov/ecy/publications/documents/1603032.pdf>, 2016.
- 1061 Lewis, E. and Wallace, D. W. R.: MATLAB Program Developed for CO<sub>2</sub> System Calculations.  
1062 ORNL/CDIAC-105., Carbon Dioxide Inf. Anal. Center, Oak Ridge Natl. Lab. U.S. Dep. Energy,  
1063 Oak Ridge, Tennessee, 1998.
- 1064 Liu, X., Patsavas, M. C. and Byrne, R. H.: Purification and characterization of meta-cresol  
1065 purple for spectrophotometric seawater pH measurements., *Environ. Sci. Technol.*, 45(11),  
1066 4862–8, doi:10.1021/es200665d, 2011.
- 1067 Lueker, T. J., Dickson, A. G. and Keeling, C. D.: Ocean *p*CO<sub>2</sub> calculated from dissolved  
1068 inorganic carbon, alkalinity, and equations for K<sub>1</sub> and K<sub>2</sub>: validation based on laboratory  
1069 measurements of CO<sub>2</sub> in gas and seawater at equilibrium, *Mar. Chem.*, 70(1–3), 105–119,  
1070 doi:10.1016/S0304-4203(00)00022-0, 2000.
- 1071 Martin, W. D. and MacCready, P.: Influence of large-scale tidal asymmetry on subtidal dynamics  
1072 in the western Strait of Juan de Fuca, *J. Geophys. Res.*, 116(C2), C02009,  
1073 doi:10.1029/2010JC006363, 2011.
- 1074 Martz, T., Mclaughlin, K. and Weisberg, S. B.: California Current Acidification Network Best  
1075 Practices for autonomous measurement of seawater pH with the Honeywell Durafet pH sensor,  
1076 [online] Available from: [file:///C:/Users/fassbender/Downloads/CCAN Durafet Best Practices](file:///C:/Users/fassbender/Downloads/CCAN%20Durafet%20Best%20Practices%20Manual%20(1).pdf)  
1077 [Manual \(1\).pdf](file:///C:/Users/fassbender/Downloads/CCAN Durafet Best Practices Manual (1).pdf), 2015.
- 1078 McElhany, P. and Shallin Busch, D.: Appropriate *p*CO<sub>2</sub> treatments in ocean acidification  
1079 experiments, *Mar. Biol.*, 160(8), 1807–1812, doi:10.1007/s00227-012-2052-0, 2013.
- 1080 Millero, F. J.: The Marine Inorganic Carbon Cycle, *Chem. Rev.*, 107(2), 308–341,  
1081 doi:10.1021/cr0503557, 2007.
- 1082 Millero, F. J., Zhang, J.-Z., Lee, K. and Campbell, D. M.: Titration alkalinity of seawater, *Mar.*  
1083 *Chem.*, 44(2–4), 153–165, doi:10.1016/0304-4203(93)90200-8, 1993.
- 1084 Mucci, A.: The solubility of calcite and aragonite in seawater at various salinities, temperatures,  
1085 and one atmosphere total pressure., *Am. J. Sci.*, 283(7), 780–799, doi:10.2475/ajs.283.7.780,  
1086 1983.
- 1087 Newton, J. A. and Devol, A. H.: Quality Assurance Project Plan: Long-term, High Resolution  
1088 Marine Water Quality Monitoring in Puget Sound using Profiling Buoys. 2012-2013 Activities.  
1089 [online] Available from: [http://orcabase.ocean.washington.edu/ORCA\\_QAPP.pdf](http://orcabase.ocean.washington.edu/ORCA_QAPP.pdf), 2012.
- 1090 Newton, J. A., Siegel, E. and Albertson, S. L.: Oceanographic Changes in Puget Sound and the



- 1091 Strait of Juan de Fuca during the 2000–01 Drought, *Can. Water Resour. J.*, 28(4), 715–728,  
1092 doi:10.4296/cwrj2804715, 2003.
- 1093 Orr, J. C., Epitalon, J.-M. and Gattuso, J.-P.: Comparison of ten packages that compute ocean  
1094 carbonate chemistry, *Biogeosciences*, 12(5), 1483–1510, doi:10.5194/bg-12-1483-2015, 2015.
- 1095 Pelletier, G., Bianucci, L., Long, W., Khangaonkar, T., Mohamedali, T., Ahmed, A. and  
1096 Figueroa-Kaminsky, C.: Salish Sea Model: Ocean Acidification Module and the Response to  
1097 Regional Anthropogenic Nutrient Sources. [online] Available from:  
1098 <https://fortress.wa.gov/ecy/publications/SummaryPages/1703009.html>, 2017.
- 1099 Pelletier, G., Roberts, M., Keyzers, M. and Alin, S. R.: Seasonal variation in aragonite saturation  
1100 in surface waters of Puget Sound – a pilot study, *Elem Sci Anth.*, 6(1),  
1101 doi:<http://doi.org/10.1525/elementa.270>, 2018.
- 1102 Pfeil, B., Olsen, A., Bakker, D. C. E., Hankin, S., Koyuk, H., Kozyr, A., Malczyk, J., Manke, A.,  
1103 Metzl, N., Sabine, C. L., Akl, J., Alin, S. R., Bates, N. R., Bellerby, R. G. J., Borges, A. V.,  
1104 Boutin, J., Brown, P. J., Cai, W.-J., Chavez, F. P., Chen, A., Cosca, C. E., Fassbender, A. J.,  
1105 Feely, R. A., González-Dávila, M., Goyet, C., Hales, B., Hardman-Mountford, N., Heinze, C.,  
1106 Hood, M., Hoppema, M., Hunt, C. W., Hydes, D. J., Ishii, M., Johannessen, T., Jones, S. D.,  
1107 Key, R. M., Körtzinger, A., Landschützer, P., Lauvset, S. K., Lefèvre, N., Lenton, A., Lourantou,  
1108 A., Merlivat, L., Midorikawa, T., Mintrop, L., Miyazaki, C., Murata, A., Nakadate, A., Nakano,  
1109 Y., Nakaoka, S., Nojiri, Y., Omar, a. M., Padin, X. a., Park, G. H., Paterson, K., Perez, F. F.,  
1110 Pierrot, D., Poisson, A., Ríos, a. F., Santana-Casiano, J. M., Salisbury, J. E., Sarma, V. V. S. S.,  
1111 Schlitzer, R., Schneider, B., Schuster, U., Sieger, R., Skjelvan, I., Steinhoff, T., Suzuki, T.,  
1112 Takahashi, T., Tedesco, K., Telszewski, M., Thomas, H., Tilbrook, B., Tjiputra, J., Vandemark,  
1113 D. C., Veness, T., Wanninkhof, R., Watson, A. J., Weiss, R., Wong, C. S. and Yoshikawa-Inoue,  
1114 H.: A uniform, quality controlled Surface Ocean CO<sub>2</sub> Atlas (SOCAT), *Earth Syst. Sci. Data*,  
1115 5(1), 125–143, doi:10.5194/essd-5-125-2013, 2013.
- 1116 Raymond, P. A. and Cole, J. J.: Increase in the Export of Alkalinity from North America’s  
1117 Largest River, *Science* (80-. ), 301, 2003.
- 1118 Raymond, P. A., Oh, N.-H., Turner, R. E. and Broussard, W.: Anthropogenically enhanced  
1119 fluxes of water and carbon from the Mississippi River, *Nature*, 451(7177), 449–452,  
1120 doi:10.1038/nature06505, 2008.
- 1121 Regnier, P., Friedlingstein, P., Ciais, P., Mackenzie, F. T., Gruber, N., Janssens, I. a., Laruelle,  
1122 G. G., Lauerwald, R., Luyssaert, S., Andersson, A. J., Arndt, S., Arnosti, C., Borges, A. V., Dale,  
1123 A. W., Gallego-Sala, A., Goddérís, Y., Goossens, N., Hartmann, J., Heinze, C., Ilyina, T., Joos,  
1124 F., LaRowe, D. E., Leifeld, J., Meysman, F. J. R., Munhoven, G., Raymond, P. a., Spahni, R.,  
1125 Suntharalingam, P. and Thullner, M.: Anthropogenic perturbation of the carbon fluxes from land  
1126 to ocean, *Nat. Geosci.*, 6(8), 597–607, doi:10.1038/ngeo1830, 2013.
- 1127 Revelle, R. and Suess, H. E.: Carbon dioxide exchange between atmosphere and ocean and the  
1128 question of an increase of atmospheric CO<sub>2</sub> during the past decades, *Tellus*, 9(1), 18–27,  
1129 doi:10.1111/j.2153-3490.1957.tb01849.x, 1957.
- 1130 Sabine, C. L., Feely, R. A., Gruber, N., Key, R. M., Lee, K., Bullister, J. L., Wanninkhof, R.,  
1131 Wong, C. S., Wallace, D. W. ., Tilbrook, B., Millero, F. J., Peng, T.-H., Kozyr, A., Ono, T. and  
1132 Rios, A. F.: The Oceanic Sink for Anthropogenic CO<sub>2</sub>, *Science* (80-. ), 305(5682), 367–371,  
1133 doi:10.1126/science.1097403, 2004.



- 1134 Sabine, C. L., Hankin, S., Koyuk, H., Bakker, D. C. E., Pfeil, B., Olsen, A., Metzl, N., Kozyr, A.,  
1135 Fassbender, A. J., Manke, A., Malczyk, J., Akl, J., Alin, S. R., Bellerby, R. G. J., Borges, A. V.,  
1136 Boutin, J., Brown, P. J., Cai, W.-J., Chavez, F. P., Chen, A., Cosca, C. E., Feely, R. A.,  
1137 González-Dávila, M., Goyet, C., Hardman-Mountford, N., Heinze, C., Hoppema, M., Hunt, C.  
1138 W., Hydes, D. J., Ishii, M., Johannessen, T., Key, R. M., Körtzinger, A., Landschützer, P.,  
1139 Lauvset, S. K., Lefèvre, N., Lenton, A., Lourantou, A., Merlivat, L., Midorikawa, T., Mintrop,  
1140 L., Miyazaki, C., Murata, A., Nakadate, A., Nakano, Y., Nakaoka, S., Nojiri, Y., Omar, a. M.,  
1141 Padin, X. a., Park, G. H., Paterson, K., Perez, F. F., Pierrot, D., Poisson, A., Ríos, a. F.,  
1142 Salisbury, J. E., Santana-Casiano, J. M., S. Sarma, V. V. S., Schlitzer, R., Schneider, B.,  
1143 Schuster, U., Sieger, R., Skjelvan, I., Steinhoff, T., Suzuki, T., Takahashi, T., Tedesco, K.,  
1144 Telszewski, M., Thomas, H., Tilbrook, B., Vandemark, D. C., Veness, T., Watson, A. J., Weiss,  
1145 R., Wong, C. S. and Yoshikawa-Inoue, H.: Surface Ocean CO<sub>2</sub> Atlas (SOCAT) gridded data  
1146 products, *Earth Syst. Sci. Data*, 5(1), 145–153, doi:10.5194/essd-5-145-2013, 2013.
- 1147 Somero, G. N., Beers, J. M., Chan, F., Hill, T. M., Klinger, T. and Litvin, S. Y.: What Changes  
1148 in the Carbonate System, Oxygen, and Temperature Portend for the Northeastern Pacific Ocean:  
1149 A Physiological Perspective, *Bioscience*, 66(1), 14–26, doi:10.1093/biosci/biv162, 2016.
- 1150 Sundquist, E. T., Plummer, L. N. and Wigley, T. M. L.: Carbon dioxide in the ocean surface: The  
1151 homogeneous buffer factor, *Science* (80-. ), 204(4398), 1203–1205,  
1152 doi:10.1126/science.204.4398.1203, 1979.
- 1153 Sutton, A. J., Sabine, C. L., Maenner-Jones, S., Lawrence-Slavas, N., Meinig, C., Feely, R. A.  
1154 A., Mathis, J. T. T., Musielewicz, S., Bott, R., McLain, P. D., Fought, H. J. and Kozyr, A.: A  
1155 high-frequency atmospheric and seawater *p*CO<sub>2</sub> data set from 14 open-ocean sites using a  
1156 moored autonomous system, *Earth Syst. Sci. Data*, 6(2), 353–366, doi:10.5194/essd-6-353-2014,  
1157 2014.
- 1158 Sutton, A. J., Sabine, C. L., Feely, R. A., Cai, W., Cronin, M. F., McPhaden, M. J., Morell, J. M.,  
1159 Newton, J. A., Noh, J., Ólafsdóttir, S. R., Salisbury, J. E., Send, U., Vandemark, D. C. and  
1160 Weller, R. A.: Using present-day observations to detect when anthropogenic change forces  
1161 surface ocean carbonate chemistry outside preindustrial bounds, *Biogeosciences*, 13(17), 5065–  
1162 5083, doi:10.5194/bg-13-5065-2016, 2016.
- 1163 Takahashi, T., Broecker, W. S., Werner, S. R. and Bainbridge, A. E.: Carbonate chemistry of the  
1164 surface waters of the worlds ocean, in *Isotope marine Chemistry*, edited by K. S. Edward D.  
1165 Goldberg, Yoshio Horibe, pp. 291–326, Geochemistry Research Association., 1980.
- 1166 Takahashi, T., Goddard, J. G. and Chipman, D. W.: Seasonal variation of CO<sub>2</sub> and nutrients in  
1167 the high-latitude surface oceans: A comparative study, *Global Biogeochem. Cycles*, 7(4), 843–  
1168 878, 1993.
- 1169 Takahashi, T., Sutherland, S. C., Wanninkhof, R., Sweeney, C., Feely, R. A., Chipman, D. W.,  
1170 Hales, B., Friederich, G. E., Chavez, F. P., Sabine, C. L., Watson, A. J., Bakker, D. C. E.,  
1171 Schuster, U., Metzl, N., Yoshikawa-Inoue, H., Ishii, M., Midorikawa, T., Nojiri, Y., Körtzinger,  
1172 A., Steinhoff, T., Hoppema, M., Olafsson, J., Arnarson, T. S., Tilbrook, B., Johannessen, T.,  
1173 Olsen, A., Bellerby, R., Wong, C. S., Delille, B., Bates, N. R. and de Baar, H. J. W.:  
1174 Climatological mean and decadal change in surface ocean *p*CO<sub>2</sub>, and net sea-air CO<sub>2</sub> flux over  
1175 the global oceans, *Deep. Res. II*, 56(8–10), 554–577, doi:10.1016/j.dsr2.2008.12.009, 2009.
- 1176 Takahashi, T., Sutherland, S. C., Chipman, D. W., Goddard, J. G. and Ho, C.: Climatological



- 1177 distributions of pH,  $p\text{CO}_2$ , total  $\text{CO}_2$ , alkalinity, and  $\text{CaCO}_3$  saturation in the global surface  
1178 ocean, and temporal changes at selected locations, *Mar. Chem.*, 164, 95–125,  
1179 doi:10.1016/j.marchem.2014.06.004, 2014.
- 1180 Takeshita, Y., Frieder, C. A., Martz, T. R., Ballard, J. R., Feely, R. A., Kram, S., Nam, S.,  
1181 Navarro, M. O., Price, N. N. and Smith, J. E.: Including high-frequency variability in coastal  
1182 ocean acidification projections, *Biogeosciences*, 12(19), 5853–5870, doi:10.5194/bg-12-5853-  
1183 2015, 2015.
- 1184 UNESCO: Protocols for the Joint Global Ocean Flux Study (JGOFS) Core Measurements, IOC  
1185 Manual and Guides 29. [online] Available from:  
1186 <http://unesdoc.unesco.org/images/0009/000997/099739eo.pdf>, 1994.
- 1187 Uppström, L. R.: The boron-chlorinity ratio of deep seawater from the Pacific Ocean, *Deep. Res.*  
1188 Part I, 21, 161–162, 1974.
- 1189 Wahl, M., Saderne, V. and Sawall, Y.: How good are we at assessing the impact of ocean  
1190 acidification in coastal systems? Limitations, omissions and strengths of commonly used  
1191 experimental approaches with special emphasis on the neglected role of fluctuations, *Mar.*  
1192 *Freshw. Res.*, 67(1), 25–36, doi:10.1071/MF14154, 2016.
- 1193 Waldbusser, G. G. and Salisbury, J. E.: Ocean acidification in the coastal zone from an  
1194 organism's perspective: multiple system parameters, frequency domains, and habitats., *Ann.*  
1195 *Rev. Mar. Sci.*, 6, 221–47, doi:10.1146/annurev-marine-121211-172238, 2014.
- 1196 Weisberg, S. B., Bednaršek, N., Feely, R. A., Chan, F., Boehm, A. B., Sutula, M., Ruesink, J. L.,  
1197 Hales, B., Largier, J. L. and Newton, J. A.: Water quality criteria for an acidifying ocean:  
1198 Challenges and opportunities for improvement, *Ocean Coast. Manag.*, 126(June), 31–41,  
1199 doi:10.1016/j.ocecoaman.2016.03.010, 2016.
- 1200 Wong, P. P., Losada, I. J., Gattuso, J.-P., Hinkel, J., Khattabi, A., McInnes, K. L., Saito, Y. and  
1201 Sallenger, A.: Coastal ecosystems and low-lying areas, in *Climate Change 2014:*  
1202 *Impacts, Adaptation, and Vulnerability. Part A: Global and Sectoral Aspects. Contribution of*  
1203 *Working Group II to the Fifth Assessment Report of the Intergovernmental Panel on Climate*  
1204 *Change*, edited by C. B. Field, V. R. Barros, D. J. Dokken, K. J. Mach, M. D. Mastrandrea, T. E.  
1205 Bilir, M. Chatterjee, K. L. Ebi, Y. O. Estrada, R. C. Genova, B. Girma, E. S. Kissel, A. N. Levy,  
1206 S. MacCracken, P. R. Mastrandrea, and L.L. White, pp. 361–409, Cambridge University Press,  
1207 Cambridge, United Kingdom and New York, NY, USA., 2014.
- 1208