

1 Insights into carbonate environmental conditions in the Chukchi Sea
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21 **Abstract**

22 Healthy Arctic marine ecosystems are essential to the food security and sovereignty, **culture**,
23 and wellbeing of Indigenous Peoples in the Arctic. At the same time, Arctic marine ecosystems
24 are highly susceptible to impacts of climate change and ocean acidification. While increasing
25 ocean and air temperatures and melting sea ice act as direct stressors on the ecosystem, they also
26 indirectly enhance ocean acidification, accelerating the associated changes in the inorganic
27 carbon system. Yet, much is to be learned about the current state and variability of the inorganic
28 carbon system in remote, **high-latitude oceans**. Here, we present **time-series (2016-2020) of pH**
29 **and the partial pressure of carbon dioxide ($p\text{CO}_2$) from the northeast Chukchi Sea continental**
30 **shelf. The Chukchi Ecosystem Observatory includes a suite of subsurface year-round moorings**
31 **sited amid a biological hotspot that is characterized by high primary productivity and a rich**
32 **benthic food web that in turn supports coastal Inupiat, whales, ice seals, walrus (*Odobenus***
33 ***rosmarus*), and Arctic cod (*Boreogadus saida*). Our observations suggest that near-bottom**
34 **waters (33 m depth, 13 m above the seafloor) are a high carbon dioxide and low pH and**
35 **aragonite saturation state (Ω_{arag}) environment in summer and fall, when organic material from the**
36 **highly productive summer remineralizes. During this time, Ω_{arag} can be as low as 0.4. In winter,**
37 **when the site was covered by sea ice, pH was < 8 and Ω_{arag} remained undersaturated under the**
38 **sea ice. There were only two short seasonal periods with relatively higher pH and Ω_{arag} , which**
39 **we term ocean acidification relaxation events. In spring, high primary production from sea ice**
40 **algae and phytoplankton blooms led to spikes in pH (pH > 8) and aragonite oversaturation. In**
41 **late fall, strong wind driven mixing events that delivered low CO_2 surface water to the shelf also**
42 **led to events with elevated pH and Ω_{arag} . Given the recent observations of high rates of ocean**
43 **acidification, and sudden and dramatic shift of the physical, biogeochemical, and ecosystem**

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75 conditions in the Chukchi Sea, it is possible that the observed extreme conditions at the Chukchi
76 Ecosystem Observatory are deviating from carbonate conditions to which many species are
77 adapted.

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79 1. Introduction

80 The quickly changing Arctic Ocean has climatic, societal, and geopolitical implications for
81 the peoples of the Arctic and beyond (Huntington et al., 2022). Arctic Indigenous Peoples are at
82 the forefront of this change and their food security, food sovereignty, culture, and ways of life
83 depend on healthy Arctic marine ecosystems (ICC, 2015). The Arctic is warming at a rate that is
84 up to four times that of the rest of the globe (Serreze and Barry, 2011; Serreze and Francis, 2006;
85 Rantanen et al., 2022). This phenomenon, called Arctic Amplification, is observed in air and sea
86 temperatures, has accelerated in recent years, and is expected to continue in the future (Rantanen
87 et al., 2022; Shu et al., 2022). Warming exerts a toll on sea ice extent, ice thickness, and the
88 duration of seasonal sea ice cover: ice is forming later in fall and retreating earlier in spring,
89 thereby increasing the length of the open water period (Stroeve et al., 2011; Serreze et al., 2016;
90 Wood et al., 2015; Stroeve et al., 2014). The lowest Arctic wide minimum sea ice extents were
91 recorded during the last 16 years of the 44 year-long satellite time-series (National Snow and Ice
92 Data Center).

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93 At the same time, the Arctic Ocean is vulnerable to ocean acidification. Although oceanic
94 uptake of anthropogenic carbon dioxide (CO₂) increases oceanic CO₂ and decreases pH and
95 calcium carbonate (CaCO₃) saturation states of calcite (Ω_{calc}) and aragonite (Ω_{arag}) globally,
96 climate induced changes to riverine input, temperature, sea ice, and circulation are accelerating
97 the rate of ocean acidification in the Arctic Ocean like nowhere else in the world (Woosley and

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103 Millero, 2020; Qi [et al.](#), 2022a; Yamamoto-Kawai et al., 2009; Orr et al., 2022; Semiletov et al.,
 104 2016; Qi et al., 2017). Recent observational studies propose that freshening of the Arctic Ocean
 105 [due to](#) increased riverine input may play an even greater role in acidifying the Arctic Ocean than
 106 the uptake of anthropogenic CO₂ (Woosley and Millero, 2020; Semiletov et al., 2016). In
 107 addition, the cold Arctic waters have naturally low concentrations of carbonate ions (CO₃²⁻) and
 108 are therefore closer to aragonite undersaturation ($\Omega_{\text{arag}} < 1$) than more temperate waters (Orr,
 109 2011; Sarmiento and Gruber, 2006), which leads to the chemical dissolution of free aragonitic
 110 CaCO₃ structures ([Bednaršek et al.](#), 2021). Because of the naturally low concentrations of CO₃²⁻,
 111 such high latitude waters have a lower capacity to take up anthropogenic CO₂ and buffer these
 112 changes (Orr, 2011). As a result, concentrations of [hydrogen ions \(H⁺\)](#) increase and pH decreases
 113 faster in the Arctic than in the tropics, for example.

114 In the Pacific Arctic, the Chukchi shelf waters have warmed by 0.45 °C decade⁻¹ since 1990,
 115 triple the rate since the beginning of the data record in 1922 (Danielson [et al.](#), 2020). Direct
 116 observations of the inorganic carbon dynamics of the Chukchi Sea are mostly limited to June
 117 through November because of the region's remoteness and accessibility during sea ice covered
 118 months. Summertime profiles across the Chukchi Sea show steep vertical gradients in inorganic
 119 carbon chemistry (Bates, 2015; Bates [et al.](#), 2009; Pipko [et al.](#), 2002; Mathis and Questel, 2013).
 120 Surface waters [have a low partial pressure of carbon dioxide \(pCO₂\)](#) as a result of high primary
 121 production after sea ice retreat, leading to aragonite supersaturated conditions, with $\Omega_{\text{arag}} > 2$
 122 ([Bates, 2015; Bates et al., 2009](#)). In areas with sea ice melt or riverine freshwater influence, Ω_{arag}
 123 tends to be lower and at times undersaturated ([Bates et al., 2009; Yamamoto-Kawai et al., 2009](#)).
 124 At the same time, [pCO₂](#) values near the seafloor are around 1000 μatm as a result of [organic](#)
 125 [matter](#), leading to summertime aragonite undersaturation (Mathis and Questel, 2013; Pipko [et al.](#),

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163 2002; Bates, 2015). Between September and November, continuous measurements from within a
164 few meters of the surface suggest a mosaic of $p\text{CO}_2$ levels between ~ 200 to 600 μatm , likely due
165 to patchy wind-induced mixing entraining high- CO_2 waters from depth into the surface mixed
166 layer (Hauri et al., 2013). Yamamoto-Kawai et al. (2016) used mooring observations of S, T, and
167 apparent oxygen utilization to estimate dissolved inorganic carbon (DIC), total alkalinity (TA),
168 and Ω_{arag} in bottom waters at their mooring site in the Hope Valley in the southwestern Chukchi
169 Sea to give first insights into year round variability of the inorganic carbon system. They found
170 slightly less intense aragonite undersaturation in spring and winter compared to summer, with a
171 net undersaturation duration of 7.5-8.5 months per year.

172 The Chukchi Ecosystem Observatory (CEO) is situated in a benthic hotspot (Figure 1) where
173 high primary production supports rich and interconnected benthic and pelagic food webs
174 (Grebmeier et al., 2015; Moore and Stabeno, 2015). The benthos is dominated by calcifying
175 bivalves, polychaetes, amphipods, sipunculids, echinoderms and crustaceans (Grebmeier et al.,
176 2015; Blanchard et al., 2013). Benthic foraging bearded seals (*Erignathus barbatus*), walrus
177 (*Odobenus rosmarus divergens*), gray whale (*Eschrichtius robustus*), and seabirds feed on these
178 calcifiers during the open water season (Kuletz et al., 2015; Jay et al., 2012; Moore et al., 2022).
179 The CEO site, located on the southern flank of Hanna Shoal, is a region of reduced stratification
180 (relative to other sides of the shoal) that likely alternately feels the effects of differing flow
181 regimes located to the west and to the east (Fang et al., 2020). Consequently, the site exhibits
182 relatively weaker currents (Tian et al., 2021) and so is conducive to deposition of sinking organic
183 matter that in turn feeds the local benthos (Grebmeier et al., 2015). Prolonged open-water
184 seasons during periods of high solar irradiance, in combination with an influx of new nutrients
185 and wind mixing, are likely enhancing primary and secondary production as well as advection of

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200 zooplankton (Lewis [et al.](#), 2020; Arrigo and van Dijken, 2015; Wood [et al.](#), 2015). These
 201 physical processes in turn fuel keystone consumers such as Arctic cod (*Boreogadus saida*) and
 202 upper trophic level ringed seals (*Phoca hispida*), beluga (*Delphinapterus leucas*) and bowhead
 203 whales (*Balaena mysticetus*) as well as predatory polar bears (*Ursus arctos*) and **Indigenous**
 204 **People** who rely on the marine ecosystem for traditional and customary harvesting (Huntington
 205 [et al.](#), 2020).

206 Perturbation of the seawater carbonate system associated with ocean acidification and
 207 climate change can have significant physiological and ecological consequences for marine
 208 species and ecosystems ([Doney et al.](#), 2020). All parameters of the carbonate system (pH, $p\text{CO}_2$,
 209 Ω_{arag} , concentrations of HCO_3^- , CO_3^{2-} , etc.) have the potential to affect the physiology of marine
 210 organisms while a change in the saturation state (Ω) can lead to the dissolution of unprotected or
 211 “free” CaCO_3 structures. Recent work has highlighted the importance of local adaptation to the
 212 present environmental variability as a key factor driving species sensitivity to ocean acidification
 213 ([Vargas et al.](#), 2017, 2022). As carbonate chemistry conditions vary enormously between
 214 regions, marine organisms are naturally exposed to different selective pressures and can evolve
 215 different strategies to cope with low pH or Ω , or high $p\text{CO}_2$. For example, the deep-sea mussel
 216 *Bathymodiolus brevior* living around vents at 1600 m depths is capable of precipitating calcium
 217 carbonate at pH ranging between 5.36 and 7.30 and highly undersaturated waters (Tunnicliffe [et](#)
 218 [al.](#), 2009). The response to changes in the carbonate chemistry is also modulated by other
 219 environmental drivers such as temperature or food availability (e.g. Thomsen [et al.](#), 2013;
 220 [Breitberg et al.](#), 2015). **Consequently**, no absolute or single threshold is expected for ocean
 221 acidification (e.g., Bednaršek [et al.](#), 2021) and a pre-requisite to assessing the impact on any
 222 biota is the monitoring at a short temporal scale to characterize the present environmental niche.

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265 When it comes to future impacts, the more intense and faster the changes associated with ocean
266 acidification, the more ~~adverse associated~~ biological impacts ~~are~~ expected (Vargas et al. 2017,
267 2022). ~~As a result, it is anticipated that~~ Arctic marine waters that are experiencing widespread
268 and rapid ocean acidification ~~will potentially~~ undergo severe negative ecosystem impacts
269 (AMAP 2018).

270 Here, we present satellite sea ice coverage data and four years of nearly continuous salinity,
271 temperature, and $p\text{CO}_2$ data, accompanied by pH, nitrate (NO_3), dissolved oxygen (O_2), and
272 chlorophyll fluorescence data for some of the time (Table 1, Figures 2 and 3). ~~We developed an~~
273 ~~empirical equation for estimating pH from moored $p\text{CO}_2$, temperature, and salinity and evaluated~~
274 ~~it using discrete samples collected across the Chukchi Sea, Bering Sea, and Beaufort Sea. Our~~
275 ~~timeseries~~ allow us to ~~assess~~ the seasonal and interannual variability and controls of the
276 inorganic carbon system in the Chukchi Sea between 2016 and 2020 and characterize the
277 chemical conditions experienced by organisms. ~~We discuss our observations in terms of~~
278 ~~progressing acidification and implications to organisms in the Chukchi Sea region.~~

279 2. Materials and Methods

280 2.1 The Chukchi Ecosystem Observatory (CEO)

281 The Chukchi Sea is a shallow shelf sea with maximum depths < 50 m. It is largely a
282 unidirectional ~~inflow shelf~~ system with Pacific origin water entering the Chukchi Sea through the
283 Bering Strait and advecting north into the Arctic Ocean (Carmack and Wassmann, 2006). ~~The~~
284 ~~CEO (71°36' N, 161°30' W, Figure 1, Hauri et al., 2018) is located along the pathway of waters~~
285 ~~flowing through Bering Strait (Fang et al., 2020) and thence from the west of Hanna Shoal~~
286 ~~toward Barrow Canyon to the south, although the wind can also drive waters from the east over~~
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308 the observatory site (Fang et al., 2020). From both shipboard and moored acoustic Doppler
309 current profiler records, the south side of Hanna Shoal mean flow is characterized by a weak
310 southward-directed current (Tian et al., 2021).

311 The observatory consists of oceanographic moorings that sample year-round, equipped with a
312 variety of sensors that measure sea ice cover and thickness (Sandy et al., 2022), light, currents,
313 waves, salinity, temperature, concentrations of dissolved oxygen, nitrate, and particulate matter,
314 pH, $p\text{CO}_2$, chlorophyll fluorescence, zooplankton abundance and vertical migration (Lalande et
315 al., 2021, 2020), the presence of Arctic cod and zooplankton (Gonzalez et al., 2021), and the
316 vocalizations of marine mammals. During some years, the observatory included a third mooring,
317 an experimental “freeze-up detection mooring”, which transmitted real-time data of conductivity
318 and temperature throughout the water column until sea ice formation. The primary moorings
319 stretch from the seafloor at 46 m to about 33 m depth, designed to avoid collisions with ice keels.

320 Pressure sensors at the top of the moorings show less than ± 1 m of excursion of the moored
321 sensor package from its deployment mean depth in any given year, indicating that mooring blow-
322 over or diving is not the cause of any observed large variability. Description of the CEO and lists
323 of sensors deployed at the site can be found in Danielson et al. (2017) and Hauri et al., (2018).

324 For this study we focus on the inorganic carbon system and its controlling mechanisms.

326 2.2 $p\text{CO}_2$

327 We used a CONTROS HydroC CO_2 sensor (4H-Jena Engineering GmbH, Kiel, Germany) to
328 measure $p\text{CO}_2$. The Contros HydroC CO_2 sensor was outfitted with a pump (SBE 5M, Sea-Bird
329 Electronics) that flushes ambient seawater against a thin semi permeable membrane, which
330 serves as equilibrator for dissolved CO_2 between the ambient seawater and the headspace of the

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345 sensor. Technical details about the sensor and its performance are described in Fietzek [et al.](#)
346 (2014), who estimated sensor accuracy to be better than 1% with postprocessing.

347 A HydroC CO₂ sensor has been deployed at the CEO site since 2016. In all deployments,
348 except 2016, HydroC CO₂ sensors were post-calibrated. The lack of post-calibration in 2016 is
349 not expected to negatively affect data quality because a battery failure resulted in [data returns](#)
350 only [over the first 3 months](#) (August through November). [Following a zero interval where the](#)
351 [gas was pumped through a soda lime cartridge to create a zero-signal reference with respect to](#)
352 [CO₂ and subsequent flush interval to allow CO₂ concentrations to return to ambient conditions,](#)
353 [measurements were taken](#) in a burst fashion every 12 or 24 hours depending on deployment year
354 (Table 1). Average *p*CO₂ values are reported as the mean of the measure interval (Table 1) with
355 standard uncertainty ([Equation 1](#)) [defined following best practices](#) (Orr [et al.](#), 2018) [and where](#)
356 [the random](#) component [is the standard deviation of the mean](#), and the systematic components
357 [include](#) sensor accuracy and estimated error of the regression during calibration.

$$u = \sqrt{u_{\text{systematic}}^2 + u_{\text{random}}^2} \quad (1)$$

358 [More than 96% of the time, the relative uncertainty of the *p*CO₂ data met the weather data](#)
359 [quality goal, defined as 2.5% by the Global Ocean Acidification Observing Network \(GOA-ON,](#)
360 [Newton et al., 2015\).](#)

362 [HydroC CO₂ data were processed using Jupyter notebook scripts developed by 4H-Jena](#)
363 [Engineering GmbH using pre- and post-calibration coefficients interpolated with any change in](#)
364 [the \[zero-signal\]\(#\) reference over the deployment](#) (Fietzek [et al.](#), 2014). Further processing using in-
365 house MATLAB scripts included removal of outliers, calculation of the average *p*CO₂, and
366 calculation of uncertainty estimates for each [measure](#) interval.

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392 **2.3 pH**

393 A SeapHOx sensor (Satlantic SeaFET™ V1 pH sensor integrated with Sea-Bird Electronics
394 SBE 37-SMP-ODO) was used to concurrently measure pH, salinity, temperature, pressure, and
395 oxygen (Martz et al., 2010). A SeapHOx was deployed at CEO in 2016, 2017, and 2018. No
396 SeapHOx was deployed in 2019 or 2020 due to supply chain delays and communication issues at
397 sea. Unfortunately, measured pH (pH_{SeaFET}) from the 2016 and 2018 SeapHOx deployments
398 were unusable due to high levels of noise in both the internal and external electrodes. In short,
399 we only have usable pH data between August 2017 and August 2018.
400 pH_{SeaFET} data were excluded during a 14-day conditioning period following deployment and
401 were processed with post-calibration corrected temperature and salinity from the SBE37
402 following Bresnahan et al. (2014) using voltage from the external electrode (V_{ext}), and $pH_{V_{ext}}$
403 (pH calculated from the external electrode of the SeaFET) from an extended period of low
404 variability (18 February 2018). Despite the availability of discrete data from one calibration cast
405 (Cross et al., 2020b; Table 2), $pH_{V_{ext}}$ was used as the single calibration point (Bresnahan et al.,
406 2014) for a variety of reasons: 1) high variability of pH_{SeaFET} (0.0581 pH units) straddling a 12
407 hour window around the discrete sample collection time, 2) high temporal and spatial variability
408 often seen in the Chukchi Sea, and 3) the discrete pH sample was within the published SeaFET
409 accuracy of 0.05 (Table 2, Figure S1). pH_{SeaFET} values are reported as the mean of the measure
410 interval (Table 1) and standard uncertainty is calculated with Equation 1 with the standard
411 deviation of the average (random), and the SeaFET accuracy (systematic). Data handling and
412 processing were done using in-house MATLAB scripts. pH is reported in total scale and at *in*
413 *situ* temperature for the entirety of this paper.

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464 **2.4 Nitrate**

465 NO₃ measurements were from a Submersible Ultraviolet Nitrate Analyzer (SUNA) V2 by
466 Sea-Bird Scientific. The SUNA is an *in situ* ultraviolet spectrophotometer designed to measure
467 the concentration of nitrate ions in water. SUNA V2 data were processed using a publicly
468 available toolbox (Hennon *et al.*, 2022; Irving, 2021) with QA/QC steps that included thermal
469 and salinity corrections (Sakamoto *et al.*, 2009), assessment of spectra and outlier removal based
470 on spectral counts (Mordy *et al.*, 2020), and concentration adjustments (absolute offset and linear
471 drift) based on pre-deployment and post-recovery reference measurements of zero concentration
472 (DI) water and a nitrate standard and, when available, nutrient samples taken from Niskin bottles
473 near the mooring site (e.g. Daniel *et al.*, 2020).

474
475 **2.5 CTD and Oxygen**

476 Two CTDs were deployed on the CEO mooring near the HydroC CO₂ depth. The main
477 pumped Sea-Bird SeaCAT (SBE16) has been deployed on the CEO mooring around 33 m depth
478 since 2014. A pumped SBE43 oxygen sensor was deployed with the SBE16 during the 2015-
479 2016, 2017-2018, and 2019-2020 deployments but only data returns from the 2017-2018
480 deployment is discussed briefly in this manuscript (Figure S2).
481 The other pumped CTD was a Sea-Bird MicroCAT (SBE37-SMP-ODO), which was
482 integrated with an optical dissolved oxygen sensor (SBE63; Figure S2), and the SeaFET pH
483 sensor within the SeapHOx instrument. The SeapHOx was deployed in fall 2016, 2017, and
484 2018. The SBE37-SMP-ODO did not record any CTD or oxygen data during the 2016
485 deployment and only recorded CTD and oxygen data between August and November 3 in 2018
486 due to battery failure.

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504 Processing of these data included temperature and conductivity correction using pre- and
505 post-calibration data following Sea-Bird Application Note 31 and oxygen correction using pre-
506 and post-calibration data following Sea-Bird Module 28. Oxygen was converted from ml/l to
507 μmol/kg following Bittig et al. (2018). Density and practical salinity were calculated using the
508 TEOS-10 GSW Oceanographic Toolbox (McDougall and Baker, 2011).

509 Differences between the two oxygen sensors (SBE43 and SBE63) of approximately 145 to
510 265 μmol/kg were observed over the 2017-2018 deployment, and both moored sensors had
511 varying offsets compared to nearby casts (Figure S2). Therefore, only relative oxygen values
512 from the freshly calibrated SBE63 are discussed in this paper.

513 The freeze-up detection mooring (Figure 6) consisted of four Sea-Bird SBE 37 inductive
514 modem CTD sensors that transmitted in real time hourly temperature, salinity, and pressure data
515 via the surface float from four subsurface depths (8, 20, 30, and 40 m; Hauri et al., 2018).

517 2.6 Development of empirical relationship to estimate pH

518 Empirical relationships for estimating water column pH have been developed for regions
519 spanning southern, tropical, temperate and Arctic biomes, using a variety of commonly measured
520 parameters (e.g., pH(S, T, NO₃, O₂, Si), Carter et al., 2018; pH(O₂, T, S), Li et al., 2016; pH(θ, O₂),
521 Watanabe et al., 2020; pH(NO₃, T, S, P) and pH(O₂, T, S, P), Williams et al., 2016; pH(O₂, T),
522 Alin et al., 2012; pH(O₂, T) and pH(NO₃, T), Juranek et al., 2009). Given the tight coupling
523 between the concentration of H⁺ and concentration of CO₂ solution, an empirical relationship for
524 estimating surface pH from pCO₂ was developed by the National Academies of Sciences,
525 Engineering and Medicine (2017) appendix F. Licker et al., (2019) used this empirical
526 relationship to calculate the global average surface ocean pH and found it represented the

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694 relationship for surface water temperatures spanning 5°C to 45°C. Here, we take a similar
 695 approach but extend it to water column pH in our cold region using temperature (T) and salinity
 696 (S) as additional proxy parameters (Equation 2).

$$pH^{est} = \alpha_0 + \alpha_1 \log(pCO_2) + \alpha_2 T + \alpha_3 S \quad (2)$$

698 Where pH^{est} is the estimated value of water column pH, pCO_2 is from the HydroC, and T and S
 699 are from the SBE16, and all α ($\alpha_0 = 10.4660$, $\alpha_1 = -0.4088$, $\alpha_2 = 0.0013$, $\alpha_3 = -0.0001$) terms are
 700 model-estimated coefficients determined using MATLAB's multiple linear regression algorithm
 701 *regress.m* (Chatterjee and Hadi, 1986). After interpolating pH_{SeaFET} (Figure 4, red dots) to the
 702 pCO_2 timestamp, the algorithm was trained over an arbitrarily chosen 180-day period
 703 (15/9/2017-14/3/2018, Figure 4, dashed box). An uncertainty of 0.0525 for pH^{est} (Figure 3 and
 704 Figure S1, gray shading) was determined with Equation 1, where the RMSE (the uncertainty in
 705 the estimation) over the entire pH_{SeaFET} timeseries is the random component and the published
 706 accuracy of the SeaFET is the systematic component (since the algorithm was trained with
 707 pH_{SeaFET}). The algorithm cross-validation and evaluation are discussed in section 3.1. Unless
 708 explicitly defined otherwise, observations of pH refer to pH^{est} for the remainder of this paper.

710 2.7 Carbonate system calculations

711 Moored data were collected at different sample intervals (Table 1) and were linearly
 712 interpolated to the HydroC CO_2 timestamp to enable further calculations. TA, DIC, and Ω_{arag}
 713 (Figure 11 a & b and Figure 3d) were calculated based on measured pCO_2 , S, T, and pressure (P)
 714 and algorithm-based pH (pH^{est}). Due to a lack of data, nutrient concentrations (Si, PO_4 , NH_4 ,
 715 H_2S) were assumed to be negligible in the CO2SYS calculations (e.g. deGrandpre et al., 2019;
 716 Vergara-Jara et al., 2019; Islam et al., 2017). pH^{est} was used in lieu of pH_{SeaFET} to allow for

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862 calculations over the whole $p\text{CO}_2$ record and due to erroneously large variability of DIC and TA
 863 when $\text{pH}_{\text{SeaFET}}$ was used as an input parameter (Raimondi [et al.](#), 2019; Cullison-Gray [et al.](#),
 864 2011). The pH - $p\text{CO}_2$ input pair leads to **large, calculated** errors in DIC and TA (Raimondi [et al.](#),
 865 2019; Cullison-Gray [et al.](#), 2011) due to strong covariance between the two parameters (both
 866 temperature and pressure dependent). Cullison-Gray [et al.](#) (2011) attributed unreasonably large
 867 short-term variability in calculated TA and DIC to temporal or spatial measurement mismatches
 868 between input pH and $p\text{CO}_2$ parameters and found that appropriate filtering alleviated noise
 869 spikes. By using pH^{est} , which by the nature of its definition is well correlated to $p\text{CO}_2$, we are
 870 eliminating some of these spurious noise spikes. We show Ω_{arag} calculated from $\text{pH}_{\text{SeaFET}}-p\text{CO}_2$
 871 (Figure [3d, red line](#)) because it is less sensitive to calculated errors as it accounts for a small
 872 portion of the total CO_2 in seawater (Cullison-Gray [et al.](#), 2011).

873 All inorganic carbon parameters were calculated using CO2SYSv3 (Sharp [et al.](#), 2023; Lewis
 874 and Wallace, 1998) with dissociation constants for carbonic acid of Lueker [et al.](#) (2000),
 875 bisulfate of Dickson (1990), hydrofluoric acid of Perez and Fraga (1987), and the boron-to-
 876 chlorinity ratio of Lee [et al.](#) (2010). Sulpis [et al.](#) (2020) found that the carbonic acid dissociation
 877 constants of Lueker [et al.](#) (2000) may underestimate $p\text{CO}_2$ in cold regions (below $\sim 8^\circ\text{C}$), and
 878 therefore overestimate pH and CO_3^{2-} . However, we choose to use Lueker [et al.](#) (2000) because
 879 they are recommended (Dickson [et al.](#), 2007; Woosley, 2021), continue to be the standard (Jiang
 880 [et al.](#), 2021; Lauvset [et al.](#), 2021), and are commonly used at high latitudes (Duke [et al.](#), 2021;
 881 Raimondi [et al.](#), 2019; Woosley [et al.](#), 2017). Furthermore, the difference between DIC
 882 **calculated from pH^{est} and $p\text{CO}_2$** and discrete samples interpolated to moored instrument depth
 883 ranged from 266 to $-195 \mu\text{mol}/\text{kg}$ using the k_1 k_2 of Sulpis [et al.](#) (2020), compared to -38 to -7
 884 $\mu\text{mol}/\text{kg}$ using Lueker [et al.](#) (2000).

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978 **2.8 Sea ice concentration**

979 Sea ice concentration at the observatory site was taken from the National Snow and Ice Data

980 Center (NSIDC; DiGirolamo *et al.*, 2022). Latitude and longitude coordinates were converted to

981 NSIDC's EASE grid coordinate system (Brodzik and Knowles, 2002) and the 25-km gridded

982 data were bilinearly interpolated to calculate sea ice concentration at ~~the CEO site~~. Low sea ice is

983 defined by < 15 % sea ice coverage per grid cell.

984

985 **2.9 Estimation of model-based ocean acidification trend**

986 Model results were obtained from historical simulations of five different global Earth System

987 Models: 1) GFDL-CM4 (Silvers *et al.*, 2018), 2) GFDL-ESM4 (Horowitz *et al.*, 2018), 3) IPSL-

988 CM6A-LR-INCA (Boucher *et al.*, 2020), 4) CNRM-ESM2-1 (Seferian, 2019), and 5) Max Plank

989 Earth System Model 1.2 (MPI-ESM1-2-LR, Wieners *et al.*, 2019) that are part of the Coupled

990 Model Intercomparison Project Phase 6 (CMIP6). Each simulation was used to calculate the

991 annual trend of aragonite saturation state and pH at the closest depth and grid cell to the CEO

992 mooring.

993

994 **3. Results**

995 ~~In the following, we will evaluate the pH algorithm (section 3.1), analyze the large~~

996 ~~variability patterns (sections 3.2 and 3.3), and then take a closer look at the data from 2020 since~~

997 ~~the seasonal cycle was different in 2020 than in previous years (section 3.4).~~

998

999 **3.1 pH algorithm**

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Salinity influence on inorganic carbon system*
 Inorganic carbon chemistry at the CEO site can be influenced by freshwater from sea ice melt and meteoric sources (precipitation and rivers). The DIC and TA signatures within these different freshwater sources can vary significantly. TA and DIC concentrations of 450 $\mu\text{mol kg}^{-1}$ and 400 $\mu\text{mol kg}^{-1}$, respectively, have been measured in Arctic sea ice (Rysgaard *et al.*, 2007). The CEO site is influenced by upstream riverine sources in the Gulf of Alaska and Bering and Chukchi seas, and also at times by the Mackenzie River outflow from the eastern Beaufort Sea and the large Russian Arctic rivers located to the west (... [97])

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1090 The algorithm estimated pH data from the CEO site reasonably well and within the weather
 1091 uncertainty goal as defined by Newton et al. (2015) most of the time. As a first step, pH^{est}
 1092 consistency was assessed through cross-validation (Figure 5) using the test dataset (outside the
 1093 training period, $r^2 = 0.9666$, RMSE = 0.166) and across the whole timeseries ($r^2 = 0.9598$, RMSE
 1094 = 0.0161, $p < 0.0001$, Figure 5). Observed high frequency spikes in pH_{SeaFET} (Figure 4, red dots;
 1095 Figure 5d, red line) were not captured by the HydroC pCO₂ sensor (sampling frequency of 12 h)
 1096 and as a result, are not reproduced in the pH^{est} timeseries. Throughout the pH_{SeaFET} timeseries,
 1097 pH^{est} overestimates pH_{SeaFET} by a mean of 0.0008 and median of 0.0039. Since pH^{est} generally
 1098 overestimates pH_{SeaFET} (mean difference of 0.0008), we assume that Ω_{arag} is also somewhat
 1099 overestimated throughout this manuscript. Discrete water samples were used as reference values
 1100 to evaluate the algorithm at the CEO site (Table 2) and were found to be within the pH^{est}
 1101 uncertainty (Figure S1).

1102 An independent verification of our algorithm was done using discrete data collected from the
 1103 Bering Sea to the Arctic Ocean on four research cruises in 2020, 2019, 2018, and 2017 (Figure
 1104 6d; Monacci et al., 2022; Cross et al., 2021; 2020a; 2020b), henceforth called the DBO dataset.
 1105 Samples collected from deeper than 500 m below the surface or flagged as questionable or bad
 1106 were excluded from this analysis. pH and pCO₂ were calculated from 1275 discrete samples
 1107 analyzed for TA, DIC, silicate, phosphate, and ammonium (except when silicate, phosphate, and
 1108 ammonium were assumed to be negligible for the 327 samples from SKQ202014S; Monacci et
 1109 al., 2022) using CO2SYSv3 (Sharp et al., 2023; section 2.7 for details) and are referred to as
 1110 pH^{disc_{calc}} and pCO₂^{disc_{calc}}, respectively. pH^{disc_{est}} was based on discrete water samples and
 1111 calculated using Equation 2 and was fit to pH^{disc_{calc}} using a linear regression ($r^2 = 0.9975$, RMSE
 1112 = 0.0078, p-value < 0.0001; Figure 6 a – c). Mean and median differences between pH^{disc_{calc}} and

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1135 $\text{pH}_{\text{est}}^{\text{disc}}$ were zero and 0.0022, respectively, with largest anomalies observed at lower salinities
 1136 (Figure 6c). Absolute differences between $\text{pH}_{\text{est}}^{\text{disc}}$ and $\text{pH}_{\text{calc}}^{\text{disc}}$ over the salinity range ~~observed~~
 1137 at the CEO site (30.87 to 33.93) fall within the weather data quality goal (Newton *et al.*, 2015)
 1138 98.7% of the time with maximum absolute differences < 0.03 . The uncertainty of 0.0154 for
 1139 $\text{pH}_{\text{est}}^{\text{disc}}$ was determined using Equation 1, where the mean combined standard uncertainty (u_c)
 1140 for $\text{pH}_{\text{calc}}^{\text{disc}}$ (0.0133; Orr *et al.*, 2018) was the systematic component, and the regression RMSE
 1141 was the random component.

1142 Empirical relationships for estimating water column pH that rely on dissolved oxygen often
 1143 ignore surface waters to limit biases due to decoupling the stoichiometry of the $\text{O}_2:\text{CO}_2$
 1144 relationship due to air-sea gas exchange (e.g. Juranek *et al.*, 2011; Alin *et al.*, 2012; Li *et al.*,
 1145 2016). We see evidence of this bias in our algorithm at low salinity (Figure 6c) and low $p\text{CO}_2$
 1146 (not shown) when compared with the DBO dataset samples collected across the Arctic and from
 1147 the surface to 500 m, with $\text{pH}_{\text{est}}^{\text{disc}}$ overestimating $\text{pH}_{\text{calc}}^{\text{disc}}$ by a maximum of 0.049. If depth is
 1148 restricted to between 30 and 500 m when evaluating the algorithm with the DBO dataset,
 1149 algorithm performance improves ($r^2 = 0.9990$, $\text{RMSE} = 0.0055$, $p\text{-value} < 0.0001$; not shown)
 1150 and the maximum $\text{pH}_{\text{est}}^{\text{disc}}$ overestimates $\text{pH}_{\text{calc}}^{\text{disc}}$ by 0.022.

1152 3.2 Relaxation events

1153 The sub-surface waters at the CEO site comprise a high $p\text{CO}_2$, low pH, and low Ω_{arag}
 1154 environment, with mean values of $p\text{CO}_2^{\text{mean}} = 538 \pm 7 \mu\text{atm}$, $\text{pH}^{\text{mean}} = 7.91 \pm 0.05$, $\Omega_{\text{arag}}^{\text{mean}} =$
 1155 0.94 ± 0.23 across the full data record (Figure 3 b, d). In the following we will focus on spikes
 1156 of high pH and Ω_{arag} and low $p\text{CO}_2$ that occur in spring (May-June) and fall (September-
 1157 December); we define these spikes as relaxation events (see discussion for justification of term).

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1171 *Spring:* Springtime relaxation events at 33 m depth that exhibit relatively higher pH and
 1172 Ω_{arag} and lower $p\text{CO}_2$ compared to the overall mean, are likely consequences of photosynthetic
 1173 activity during sea ice break-up (Figures 2 and 3). In June of 2019 and 2020, near bottom pH and
 1174 Ω_{arag} spiked to > 8.17 and > 1.5 , respectively, while $p\text{CO}_2$ dropped to $< 286 \mu\text{atm}$. Ω_{arag} remained
 1175 oversaturated and pH was greater than 8.0 for nearly all of June in 2018. In 2019, the relaxation
 1176 event was less sustained, with only four short (2-6 day-long) events of relatively higher pH and
 1177 $\Omega_{\text{arag}} > 1$ in June. In both years, chlorophyll fluorescence spiked and either O_2 increased (in
 1178 2018) or NO_3 decreased (in 2019), which are signs of photosynthetic activity and primary
 1179 production.

1180 *Fall:* The relaxation events in fall were characterized by large and sudden drops in $p\text{CO}_2$,
 1181 abrupt increases in pH and Ω_{arag} , and considerable interannual variability in their timing. Unlike
 1182 the relaxation events observed in spring, we attribute these fall relaxation events to wind-induced
 1183 physical mixing. To examine the controlling mechanisms causing these abrupt relaxation events
 1184 in fall, we will start with using water column salinity and temperature data from a freeze-up
 1185 detection buoy (Hauri et al., 2018) that was deployed in summer 2017 approximately 1 km away
 1186 from the biogeochemical mooring. The freeze-up detection mooring provided temperature and
 1187 salinity measurements every 7 meters throughout the water column from the time of its
 1188 deployment in mid-August until freeze-up. Data from the freeze-up detection mooring suggest
 1189 that warmer and fresher water from the upper water column gets periodically entrained down to
 1190 the location of the biogeochemical sensor package at 33 m depth, leading to enhanced variability
 1191 of density in August and September (Figure 7). Fluctuations of the pycnocline associated with
 1192 the passage of internal waves could also elevate signal variances. During this time $p\text{CO}_2$ often
 1193 decreased to or below atmospheric levels and pH sporadically reached values > 8 . At the end of

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1256 September, a strong mixing event (with coincident strong surface winds) homogenized the water
1257 column from the surface down to the location of the sensor package and caused a sudden
1258 temperature increase from 0.4 °C to 3.9 °C (Figure 7c and 8a). At the same time, $p\text{CO}_2$ (Figure
1259 7b and 8) decreased from 590 to 308 μatm . This suggests that warm and low CO_2 surface water
1260 mixed with CO_2 -rich subsurface water and led to a sustained relaxation period that subsequently
1261 lasted until mid-November. Another mixing event further eroded the water column stratification
1262 and replaced subsurface water with colder and fresher water (ice melt) from the surface at the
1263 end of October. This second large mixing event did not lead to large changes in $p\text{CO}_2$, pH, and
1264 Ω_{arag} .

1265 Salinity and temperature records from the biogeochemical mooring at 33 m depth also
1266 suggest fall season mixing events in all other years, when increases in temperature coincide with
1267 decreases in $p\text{CO}_2$ (Figure 8). For example, two mixing events shaped the carbonate chemistry
1268 evolution in fall 2018. $p\text{CO}_2$ decreased from 915 μatm to around 565 μatm and Ω_{arag} increased to
1269 0.9 as temperature increased and salinity decreased in early September (Figures 2 and 8). $p\text{CO}_2$
1270 then increased to 1160 μatm in late October, before decreasing to 385 μatm at the beginning of
1271 November, causing a spike in Ω_{arag} to 1.34. At the same time, salinity decreased by 1 unit,
1272 suggesting a strong mixing event. Throughout November 2018, $p\text{CO}_2$ oscillated between 344 and
1273 757 μatm and salinity between 31.01 and 32.97, hinting at additional mixing.

1274 Similarly, an early mixing event in 2019 decreased $p\text{CO}_2$ to 352 μatm at the beginning of
1275 September. Short-term variability in $p\text{CO}_2$ with maximum levels of up to 855 μatm and
1276 minimum values below 300 μatm , variable temperature and salinity, and sporadic aragonite
1277 oversaturation events point to mixing through mid-September. At the end of October, a large
1278 mixing event homogenized the water column, accompanied by a decline of salinity by >1 unit,

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1290 increase of temperature to 4 °C, and decrease of $p\text{CO}_2$ from 565 μatm to below 400 μatm . In a
1291 similar fashion to 2018, this fall mixing event was followed by a month-long period of large
1292 variability of $p\text{CO}_2$, salinity, pH, and Ω_{arag} , leading to short and sporadic aragonite oversaturation
1293 events in November, and sustained oversaturation in December.

1295 3.3 Sustained periods of low pH and Ω_{arag} , and high $p\text{CO}_2$

1296 Waters at 33 m depth at the CEO site were most acidified during the sea ice free periods

1297 until mixing events entrained surface waters to the sensor depth (section 3.2). pH and Ω_{arag}

1298 started to gradually decrease from their maximum levels ($\Omega_{\text{arag_max}} = 1.65$, $\text{pH}_{\text{max}} = 8.19$) at the

1299 beginning of June in 2018 to their annual low at the beginning of November ($\Omega_{\text{arag_min}} = 0.47$,

1300 $\text{pH}_{\text{min}} = 7.58$, Figure 3.d and c). In November, the waters were also undersaturated with regards

1301 to calcite (not shown) and $p\text{CO}_2$ peaked at 1159 μatm (Figure 3.b). Dissolved oxygen decreased

1302 by about 400 $\mu\text{mol kg}^{-1}$ between July and October, when the sensor stopped working properly.

1303 The decrease of dissolved oxygen suggests remineralization of organic material. The decrease of

1304 pH, Ω_{arag} , O_2 and increase of $p\text{CO}_2$ was briefly interrupted by a strong mixing event in

1305 September, which entrained warmer, fresher, and CO_2 -poorer water down to 33 m depth (section

1306 3.2, Figure 8). The 2019 observations paint a similar picture of remineralization during the

1307 summer months, as the $p\text{CO}_2$ increase and pH and Ω_{arag} decreases were accompanied by an NO_3^-

1308 increase.

1309 $p\text{CO}_2$ steadily increased and pH and Ω_{arag} decreased during the sea ice covered periods

1310 (Figures 8). pH was < 8 and Ω_{arag} remained undersaturated under the sea ice. At the same time,

1311 NO_3^- slowly increased and O_2 decreased, which points to slow organic matter remineralization

1312 (Figure 9). Short-term variability in $p\text{CO}_2$, especially in January of all three observed years, was

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1458 also reflected in salinity, O_2 and NO_3 (Figure 9) and could be attributed to advection, as the CEO
1459 site is adjacent to contrasting regimes of flow and hydrographic properties (Fang et al., 2020).

1461 3.4 Spring and summer of 2020 were different

1462 The seasonal cycle in 2020 strongly contrasted with the previous observed years. pCO_2
1463 gradually increased by roughly 200 μatm throughout the sea ice covered months to 650 μatm
1464 when sea ice started to retreat at the beginning of July. By the end of July, pCO_2 doubled and
1465 increased to 1389 μatm , which is the highest pCO_2 level recorded in this timeseries. The peak of
1466 pCO_2 was accompanied by an increase in salinity of 0.5 while temperature did not change,
1467 suggesting the influence of advection. At the beginning of August, pCO_2 dropped to 536 μatm
1468 and then oscillated around 600 μatm through much of August before returning to around 900
1469 μatm for the next month. Similarly, pH decreased to 7.5 at the end of July and then oscillated
1470 around 7.85, while O_{atag} dropped to 0.37, and oscillated around 0.85. The steep drop and
1471 oscillation of pCO_2 was reflected in NO_3 , suggesting that primary production and
1472 remineralization played a role. When pCO_2 and NO_3 decreased at the beginning of August,
1473 temperature simultaneously increased by 0.7 °C and salinity decreased by 0.12, suggesting that
1474 entrainment of shallower water masses may have played a role too. Comprehensive analyses of
1475 the factors that resulted in the 2020 differing conditions are beyond the scope of this paper, but
1476 deserve attention in a future effort.

1478 4. Discussion

1479 CEO data provide new insights into the synoptic, seasonal and interannual variability of
1480 the inorganic carbon system in a time when ocean acidification and climate change have already

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1530 started to transform this area. The observations suggest that the CEO site is a high-CO₂ and low-
1531 pH and low- Ω_{arag} environment most of the time, except during sea ice break-up when the effects
1532 of photosynthetic activity remove CO₂ from the system, and later in fall, when strong storm
1533 events entrain low $p\text{CO}_2$ surface waters to the seafloor. Lowest pH and CaCO₃ saturation states
1534 and highest $p\text{CO}_2$ occur in summer through late fall when organic matter remineralization
1535 dominates the carbonate system balance. During this time, Ω_{arag} can fall below 0.5 and even Ω_{calc}
1536 becomes sporadically undersaturated ($\Omega_{\text{calc}} < 1$).

1538 4.1 pH algorithm

1539 Deploying oceanographic equipment in remote Arctic locations is challenging. The data
1540 return from the SeaphOX sensors was disappointingly minimal, despite annual servicing and
1541 calibration by the manufacturer. Our new pH algorithm is therefore even more important as it
1542 fills pH data gaps in the CEO timeseries and can be applied with confidence from the Bering to
1543 the Beaufort seas (Figure 6). While another successful year of moored pH data return at the CEO
1544 site is needed to fully evaluate our algorithm throughout the year, comparison with single
1545 discrete water samples nearby the CEO site and the DBO dataset (section 3.1, Table 2, Figures 6
1546 and S1) suggest that our algorithm-derived pH meets the weather quality uncertainty goal of \pm
1547 0.02 (Newton et al., 2015) much of the time.

1548 The combination of our new algorithm with recent progress in monitoring $p\text{CO}_2$ with
1549 Seagliders (Hayes et al., 2022) will further increase our ability to study the inorganic carbon
1550 dynamics at times and locations when shipboard or mooring based measurements may not be
1551 practical. Additional assessment is needed to determine to what degree the algorithm needs
1552 adjustments beyond the region evaluated in this work.

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- Deleted: The algorithm generally overestimates pH by 0.0008, which means that the Ω_{arag} is also somewhat overestimated throughout the manuscript. Empirical relationships for estimating water column pH that rely on dissolved oxygen often ignore surface waters to limit biases due to decoupling the stoichiometry of the O₂:CO₂ relationship due to air-sea gas exchange (e.g. Juranek et al., 2011; Alin et al., 2012; Li et al., 2016). We see evidence of this bias in our algorithm at low salinity (Figure 5c) (... [116])
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4.2 Uncertainty

Inherent spatial and temporal variability of the inorganic carbon parameters in the Chukchi Sea make the use of discrete water samples for evaluating sensor-based measurements difficult. Historic continuous surface measurements from the area suggest that surface $p\text{CO}_2$ can be as low $< 250 \mu\text{atm}$ in early fall (Hauri et al., 2013), at a time of year when subsurface $p\text{CO}_2$ reaches its max of $> 800 \mu\text{atm}$ at the CEO site. This suggests a steep $p\text{CO}_2$ gradient of $> 17 \mu\text{atm}$ per meter. High-resolution pH data from the 2017/2018 deployment suggests high temporal variability as well, further complicating the collection of discrete water samples to adequately evaluate the sensors. The HydroC's zeroing function, in addition to our pre- and post-calibration routines that factor into the post-processing of the data, gives us confidence in the accuracy of the $p\text{CO}_2$ data, and further confidence in pH derived from $p\text{CO}_2$.

The pH^{est} uncertainty of 0.0525 is likely a conservative estimate based on our validation of pH^{est} (section 3.1, Table 2). Consequently, propagated uncertainties in the calculated parameters are high. As discussed in section 2.7, the pH- $p\text{CO}_2$ input pair exacerbates these larger uncertainties. Mean $\text{TA}(\text{pH}^{\text{est}}, p\text{CO}_2)$, $\text{DIC}(\text{pH}^{\text{est}}, p\text{CO}_2)$, and $\Omega_{\text{arag}}(\text{pH}^{\text{est}}, p\text{CO}_2) \pm u_c$ (Orr et al., 2018) are $2173 \pm 281 \mu\text{mol kg}^{-1}$, $2111 \pm 263 \mu\text{mol kg}^{-1}$ and 0.94 ± 0.23 , respectively, when input uncertainties are the standard uncertainty (Equation 1). When the input uncertainty for pH^{est} is only the RMSE of 0.0161 (section 3.1), uncertainties decrease to $\pm 98 \mu\text{mol kg}^{-1}$, $\pm 93 \mu\text{mol kg}^{-1}$, and ± 0.09 , respectively. When input uncertainties are only the random component of the input parameters (i.e. standard deviation for $\text{pH}_{\text{SeaFET}}$ and $p\text{CO}_2$ and instrument precision for T and S), $\text{TA}(\text{pH}_{\text{SeaFET}}, p\text{CO}_2)$, $\text{DIC}(\text{pH}_{\text{SeaFET}}, p\text{CO}_2)$, and $\Omega_{\text{arag}}(\text{pH}_{\text{SeaFET}}, p\text{CO}_2) \pm u_c$ drops to $\pm 38 \mu\text{mol kg}^{-1}$, $\pm 37 \mu\text{mol kg}^{-1}$, and ± 0.06 , respectively. Given the above uncertainties and that we

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1678 do not see significant biofouling at the CEO site, we believe that short term variability can be
 1679 discussed with confidence with this dataset. In other words, wiggles in the data represent real
 1680 events, despite the high uncertainty in the precise value of the calculated parameters.

1681

1682 **4.3 Subsurface biogeochemical drivers of pH, Ω_{arag} , and pCO_2**

1683 Inorganic carbon chemistry can be influenced by advection and vertical entrainment of
 1684 different water masses, temperature, salinity, biogeochemistry, and conservative mixing with TA
 1685 and DIC freshwater endmembers. Here, we followed Rheuban et al. (2019) and separated the
 1686 drivers of the observed large pH, Ω_{arag} , and pCO_2 variability to provide additional insights into
 1687 our timeseries (Figure 10) using CO2SYS by altering input parameters temperature, salinity, TA,
 1688 and DIC. Anomalies relative to the reference values $pH(T_0, S_0, DIC_0, TA_0)$, $\Omega_{arag}(T_0, S_0, DIC_0,$
 1689 $TA_0)$, and $pCO_2(T_0, S_0, DIC_0, TA_0)$, were calculated using a linear Taylor series decomposition,
 1690 adding up the thermodynamic effects of temperature and salinity, and the perturbations due to
 1691 biogeochemistry, and conservative mixing with freshwater DIC and TA endmembers, (Rheuban
 1692 et al., 2019). Reference values $T_0, S_0, DIC_0,$ and $TA_0,$ are the mean of the CEO timeseries.
 1693 Freshwater from sea ice melt and meteoric sources (precipitation and rivers) may influence the
 1694 CEO site. TA and DIC concentrations of $450 \mu\text{mol kg}^{-1}$ and $400 \mu\text{mol kg}^{-1}$, respectively, have
 1695 been measured in Arctic sea ice (Rysgaard et al., 2007). Riverine input along the Gulf of Alaska
 1696 tends to have lower TA ($366 \mu\text{mol kg}^{-1}$) and DIC ($397 \mu\text{mol kg}^{-1}$) concentrations (Stackpoole et
 1697 al., 2016, 2017) than rivers draining into the Bering, Chukchi, and Beaufort Seas (TA = 1860
 1698 $\mu\text{mol kg}^{-1}$, DIC = 2010 $\mu\text{mol kg}^{-1}$, Holmes et al., 2021) all of which can influence the CEO site
 1699 to some extent (Asahara et al., 2012; Jung et al., 2021). In this Taylor decomposition we used sea
 1700 ice TA and DIC endmembers (Rysgaard et al., 2007) but want to emphasize that using Arctic

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1914 river endmembers did not meaningfully change the results (not shown). Figure 10 shows the

1915 effects of biogeochemical processes, temperature, salinity, and conservative mixing with TA and

1916 DIC freshwater endmembers on pH, Ω_{arag} , and $p\text{CO}_2$. The effects of salinity (red) and

1917 conservative mixing with TA and DIC freshwater endmembers (green) are negligible for pH,

1918 Ω_{arag} , and $p\text{CO}_2$. Temperature varied between $-1.7\text{ }^\circ\text{C}$ during the sea ice covered months and up

1919 to $4\text{ }^\circ\text{C}$ in late fall, when wind events mixed the whole water column and entrained warm and

1920 low $p\text{CO}_2$ surface waters to the instrument depth at 33 m (see section 3.2 for a more in-depth

1921 discussion of these mixing events). During this time, the increase in temperature counteracted the

1922 effect of biogeochemistry slightly and increased $p\text{CO}_2$ and decreased pH (Figure 10 a,c).

1923 Temperature did not affect Ω_{arag} .

1924 Biogeochemistry (photosynthesis, respiration, calcification, dissolution) is the most

1925 important driver of the inorganic carbon dynamics at 33 m depth at the CEO site. The springtime

1926 relaxation events in 2018 and 2019 with relatively higher pH and Ω_{arag} , and lower $p\text{CO}_2$, were

1927 mainly driven by biogeochemistry (Figure 10). During these events O_2 increased and NO_3

1928 decreased, suggesting photosynthetic activity (Figure 2d,e and 3a). Near bottom photosynthetic

1929 activity by phytoplankton or sea ice algae has been observed at different locations across the

1930 Chukchi Sea (Arrigo et al., 2017; Ouyang et al., 2022; Stabeno et al., 2020; Koch et al., 2020).

1931 Sediment trap data from a CEO deployment prior to the start of this $p\text{CO}_2$ and pH time-series

1932 suggest that export of the exclusively sympagic sea ice algae *Nitzschia frigida* peaked in May

1933 and June, during snow and ice melt events (Lalande et al., 2020), further supporting the

1934 hypothesis that sea ice algae contributed to the CO_2 draw down. Interestingly, TA also increased

1935 significantly during these events in 2018 and 2019, which cannot be solely attributed to organic

1936 matter production. Specifically, TA increased by $23\text{ }\mu\text{mol kg}^{-1}$ in 2019 (Figure 11a). However,

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Biogeochemistry is the most important driver of the inorganic carbon dynamics at the 33 m depth at the CEO site
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2019 with an observed NO_3^- increase of 7.6 umol kg^{-1} , we would expect an increase of TA by 7.6 umol
 2020 kg^{-1} . This is assuming that NO_3^- is the primary source of nitrogen during organic matter
 2021 formation, and that assimilation of 1 umol of NO_3^- leads to an increase of TA of 1 umol (Wolf-
 2022 Gladrow et al., 2007). The TA increase of 23 umol kg^{-1} is therefore larger than expected from
 2023 organic matter formation alone and is likely due to CaCO_3 mineral dissolution. While direct
 2024 evidence is missing, the strong TA increase suggests that CaCO_3 mineral dissolution during sea
 2025 ice break up also plays an important role at the CEO site. As observed in other Arctic areas, it is
 2026 possible that ikaite crystals that were trapped in the ice matrix dissolved in the water column
 2027 when sea ice melted (Rysgaard et al., 2012, 2007).

2028

2029 **4.4 Progression of ocean acidification in the Chukchi Sea**

2030 The Arctic Ocean acidification rate will continue to exceed the rate of CO_2 change in the
 2031 atmosphere because of the impacts of freshening and other more localized, seasonal or short-
 2032 term consequences of climate change (Woosley and Millero, 2020; Terhaar et al., 2021; Orr et
 2033 al., 2022; Qi et al., 2017). Seventeen years of ship-based data from sub surface Chukchi Summer
 2034 water suggests a mean pH change of -0.0047 ± 0.0026 and mean Ω_{arag} change of -0.017 ± 0.009
 2035 (Qi et al., 2022b). As a comparison, an average across historic simulations from five CMIP6
 2036 models (see methods) estimates a change in pH of $-0.0077 \text{ year}^{-1}$ and Ω_{arag} of $-0.0063 \text{ year}^{-1}$ at 33
 2037 m of the CEO site between 2002 – 2014. The historic CMIP6 simulations end in 2014 and
 2038 therefore miss the last years of extreme sea ice loss. Both observations and global model-based
 2039 trend estimates must be used with caution. The observations were collected during the sea ice
 2040 free period (Qi et al., 2022b), and therefore do not depict an annually representative trend.
 2041 Global models do not resolve important local physical, chemical, and biological meso-scale

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The observe... d... aCO₃ mineral dissolution during sea ice break up at the... also plays an important role at the CEO site. As observed in other Arctic areas, it is possible that ikaite crystals that were trapped in the ice matrix dissolved in the water column when sea ice melted are likely driven by ikaite crystal dissolution in spring and aragonite and calcite dissolution in summer and fall. In spring, TA increased while both aragonite and calcite were supersaturated. Thus, the chemical dissolution of aragonite and calcite is not possible. The observed spike in TA happened in conjunction with sea ice break up. We therefore hypothesize that ikaite crystals that were trapped in the ice matrix during brine rejection...

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2274 processes and therefore mask out the variability of the inorganic carbon system and effects of
2275 climate change.
2276 Organisms living at the CEO site may have always been exposed to large seasonal
2277 variability and low pH and Ω_{arag} (high $p\text{CO}_2$), but the combined and cumulative effects of
2278 climate change and ocean acidification have rapidly made these conditions more extreme and
2279 longer lasting. Ocean acidification serves as a gradual environmental press by increasing the
2280 system's mean and extreme $p\text{CO}_2$ and decreasing mean and extreme pH and Ω_{arag} . Climate
2281 induced changes to other important controls of the inorganic carbon system, such as sea ice,
2282 riverine input, temperature, and circulation can act as sudden pulses and further modulate the
2283 inorganic carbon system to a less predictable degree and cause extreme events (Woosley and
2284 Millero, 2020; Orr et al., 2022; Hauri et al., 2021; Oi et al., 2017). Huntington et al. (2020)
2285 describe a sudden and dramatic shift of the physical, biogeochemical and ecosystem conditions
2286 in the Chukchi and Northern Bering seas in 2017. For example, satellite data for the CEO site
2287 illustrate that the longest open water seasons on record occurred between 2017 and 2020. Before
2288 2017, the open water season was on average 81 (± 40) days long (i.e., below 15 %
2289 concentration), of which 60 (± 44) days were ice free, whereas between 2017 and 2020, the low
2290 sea ice period was 157 (± 30) days long, of which 152 (± 24) days were ice free (Figure 12). Sea
2291 ice decline and increased nutrient influx has also promoted increased phytoplankton primary
2292 production in the area (Lewis et al., 2020; Arrigo and van Dijken, 2015; Payne et al., 2021).
2293 Since our inorganic carbon timeseries started after the "dramatic shift" that was observed in the
2294 Chukchi Sea in 2017 (Huntington et al., 2020) and given the uncertainty in model output in this
2295 region, we can only speculate about how the changes in sea ice, temperature and biological
2296 production may have affected seasonal variability and extremes of the inorganic carbon

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2313 chemistry at the CEO site. However, since the summertime low pH and Ω_{arag} and high $p\text{CO}_2$ are
2314 tightly coupled to the length of the ice-free period and intensity of organic matter production, it
2315 is possible that the observed summertime period of extreme conditions may have been
2316 previously unexperienced at this site. We therefore think it is justified to call the spikes of pH
2317 and Ω_{arag} “ocean acidification relaxation events”, since the long-lasting summertime period of
2318 extremely low pH and Ω_{arag} may be a new pattern.

2319

2320 **4.5. Relevance for ecosystem**

2321 Marine organisms are exposed to a wide range of naturally fluctuating environmental
2322 conditions such as temperature, salinity, carbonate chemistry and food concentrations that
2323 together constitute their ecological niche. As evolution works toward adaptation, the tolerance
2324 range of species and ecosystems to such parameters varies between locations and is often closely
2325 related to niche status (Vargas et al., 2022). Stress can be defined as a condition evoked in an
2326 organism by one or more environmental and biological factors that bring the organism near or
2327 over the limits of its ecological niche (after Van Straalen, 2003). The consequence of the
2328 exposure to a stressor will depend on organismal sensitivity, stress intensity (how much it
2329 deviates from present conditions) and stress duration. In a synthesis of the global literature on the
2330 biological impacts of ocean acidification, Vargas et al. (2017, 2022) showed that the extreme of
2331 the present range of variability of carbonate chemistry is a good predictor of species sensitivity.
2332 In other words, larger deviations from present extreme high $p\text{CO}_2$ or extreme low pH, would be
2333 expected to exert more negative biological impacts. Organismal stress and niche boundaries have
2334 implications for the definition and understanding of controls and future ocean acidification
2335 conditions in experiments aimed at evaluating future biological impacts.

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2339 Our data provide insights on conditions that affect and determine local species’
 2340 ecological niches, and a necessary key is to evaluate or re-evaluate their sensitivity to present and
 2341 future carbonate chemistry conditions, particularly for the sessile benthic calcifiers that constitute
 2342 prey for mobile and upper trophic level taxa. For example, an experimental study on three
 2343 common Arctic bivalve species (*Macoma calcarean*, *Astarte montagui* and *Astarte borealis*)
 2344 collected in the CEO concluded that these species were generally resilient to decreasing pH
 2345 (Goethel et al., 2017). However, only two pH were compared (a “control” (pH of 8.1) and an
 2346 “acidified” treatment (pH of 7.8) and our results show that organisms are already experiencing
 2347 more extreme conditions today than have been experimentally manipulated. While these data
 2348 provide insights on these species’ plasticity to present pH conditions, they cannot be used to infer
 2349 sensitivity to future ocean acidification or extremes of current conditions. Based on the local
 2350 adaptation hypothesis (Vargas et al., 2017, 2022), stress and associated negative effect on species
 2351 fitness can be expected when pH deviates from the extreme of the present range of variability
 2352 (pH<7.5) as shown in other regions (e.g. echinoderms: Dorey et al., (2013); crustaceans: Thor
 2353 and Dupont, (2015); bivalves: Ventura et al., (2016)).

2354 At the CEO, our results show sustained periods of remarkably low pH (e.g., 7.5; summer
 2355 to fall, winter). Higher pH values are observed in spring and late fall. While we are lacking the
 2356 local biological data to sufficiently evaluate past and future ecosystem changes, a high rate of
 2357 ocean acidification as observed in the Chukchi Sea (Qi et al., 2022b, a), associated with potential
 2358 temperature-induced shifts in the carbonate chemistry cycle (e.g. Orr et al., 2022), have the
 2359 potential to impact species and ecosystems. Exposure to low pH increases organismal energy
 2360 requirements for maintenance (e.g. acid-base regulation: Stumpp et al., 2012, compensatory
 2361 calcification: Ventura et al., 2016). Organisms can cope with increased energy costs using a

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2402 variety of strategies, ranging from individual physiological to behavioral responses, depending
2403 on trophic level, mobility, and other ecological factors. For example, they can use available
2404 stored energy to compensate for increased costs or they can decrease their metabolism to limit
2405 costs (AMAP 2018). At the CEO, the low pH period observed during the summer and fall is
2406 associated with elevated temperature and an elevated food supply for herbivores (Lalande et al.,
2407 2020). The high availability of food may then foster compensation for the higher energetic costs
2408 associated with exposure to low pH. However, a longer period of low pH as suggested by our
2409 data could lead to a mismatch between the low pH and food availability, with cascading negative
2410 consequences for the ecosystem (Kroeker et al., 2021). In winter, the low pH conditions are
2411 associated with low temperature, no light, and low food level concentrations. These conditions
2412 are likely to keep metabolisms low and limit the negative effects of exposure to low pH
2413 (Gianguzza et al., 2014). As food availability is limited by the absence of light, this strategy may
2414 be compromised by an increase in temperature that could also lead to increased metabolism.
2415 Additional work is needed to understand impacts of acidification conditions and variability on
2416 the marine biota of the Chukchi Sea, including field and laboratory experiments that evaluate
2417 biological response under realistic scenarios. The characterization of the environmental
2418 conditions at the CEO, including the variability in time, can be used to design single and multiple
2419 stressor experiments (carbonate chemistry, temperature, salinity, food, oxygen; Boyd et al.,
2420 2018).

2421 Indigenous communities are at the forefront of the changing Arctic, including changes in
2422 accessibility, availability, and condition of traditional marine foods (Buschman and Sudlovenick,
2423 2022; Hauser et al., 2021). Several marine species are critical to the food and cultural security of
2424 coastal Inupiat who have thrived in Arctic Alaska for millenia. While it is not possible to resolve

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2432 [the consequences of the seasonal and interannual variations in carbonate chemistry documented](#)
2433 [in this manuscript without a proper sensitivity evaluation, the seasonally low pH conditions have](#)
2434 [the potential to impact organisms like bivalves in a foraging hotspot for walrus \(Jay et al., 2012;](#)
2435 [Kuletz et al., 2015\). Walrus, as well as their bivalve stomach contents, are important nutritional,](#)
2436 [spiritual, and cultural components, raising concerns for food security in the context of ecosystem](#)
2437 [shifts associated with the variability and multiplicity of climate impacts within the region \(ICC,](#)
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2440 5. Concluding Thoughts

2441 The Chukchi Sea is undergoing a rapid environmental transformation with potentially
2442 far-reaching consequences across the ecosystem. While we are lacking a long-term time-series,
2443 we used this dataset to [investigate](#) the drivers of extreme pH, Ω_{arag} , and $p\text{CO}_2$ and document
2444 conditions that could affect the ecological niches of organisms, including a fast rate of ocean
2445 acidification, elongated sea ice free periods, increased primary productivity and elevated
2446 temperature. While a combination of experimental and monitoring approaches is needed for an
2447 understanding of the ecological consequences of these changes, our results also highlight [the](#)
2448 urgency to mitigate CO₂ emissions and simultaneously support Indigenous-led conservation
2449 measures to safeguard an ecosystem in transition. Indigenous People in the Arctic have
2450 established strategies to monitor, adapt to, and conserve the ecosystems upon which they depend.
2451 Ethical and equitable engagement of Indigenous Knowledge and the communities at the forefront
2452 of climate impacts can help guide research and conservation action by centering local priorities
2453 and traditional practices, thereby supporting self-determination and sovereignty (Buschman and
2454 Sudlovenick, 2022).

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2462 **Data availability**

2463 The **inorganic carbon** data used in this manuscript are publicly available (Hauri and
2464 Irving, 2023a; Hauri and Irving, 2023b).

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2466 **Author contributions**

2467 CH and BI managed and serviced the HydroC CO₂ and SeapHOx sensors, analyzed and
2468 published the data, and wrote the manuscript. **SD and Peter Shipton carried out the CEO mooring
2469 deployments and recoveries and managed and serviced the CTD and NO₃ sensors.** RP, DH, SD,
2470 and SLD contributed to the manuscript.

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2472 **Competing interests**

2473 The authors have no competing interests.

2474

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2476 The Chukchi Ecosystem Observatory is located on the traditional and contemporary
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2480 this not only because we are grateful to the Indigenous communities who have been in deep
2481 connection with the land and water for time immemorial, but also in recognition of the historical
2482 and ongoing legacy of colonialism. We are committed to improving our scientific approaches
2483 and working towards co-production for a better future for everyone.

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3176 **Tables**

3177 **Table 1.** [Chukchi Ecosystem Observatory location and instrument sampling frequency.](#) **Sensor**
3178 **type and parameter measured (italicized) shown in top row. Values in parenthesis indicate the**
3179 **number of measurements averaged over the measurement interval window.**

Deployment	Latitud <i>ε</i>	Longitude	SUNA <i>NO₃</i>	HvdroC <i>CO₂</i> <i>pCO₂</i>	SBE16 <i>CTD+</i>	SBE37 <i>CTD</i>	SeaFET <i>pH</i>	SBE63 <i>O₂</i>
2016-2017	71.5996	-161.5184	1 h	12 h (300/5 min)*	1 h	-	-	-
2017-2018	71.5997	-161.5189	1 h	12 h (5/5 min)	2 h	2 h	2 h (30/5 min)	2 h

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2018-2019	71.5999	-161.5281	1 h	24 h (5/5 min)	1 h	2 h*	-	2 h*
2019-2020	71.5997	-161.5275	1 h	12 h (5/5 min)	2 h	-	-	-
* indicates the sensor did not return data over the whole year due to battery failure								
CTD+ indicates ancillary data was available with the SBE16 file (e.g., chlorophyll fluorescence)								

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Table 2. Evaluation of pH_{SeaFET} and pH^{est} using reference pH from nearby discrete samples (pH^{disc}_{calc}). Uncertainty, u_c , is the propagated combined standard uncertainty from *errors.m* (Orr et al., 2018). pH_{SeaFET} and pH^{est} were interpolated to the discrete timestamp. [Figure S1](#) for visualization of reference values.

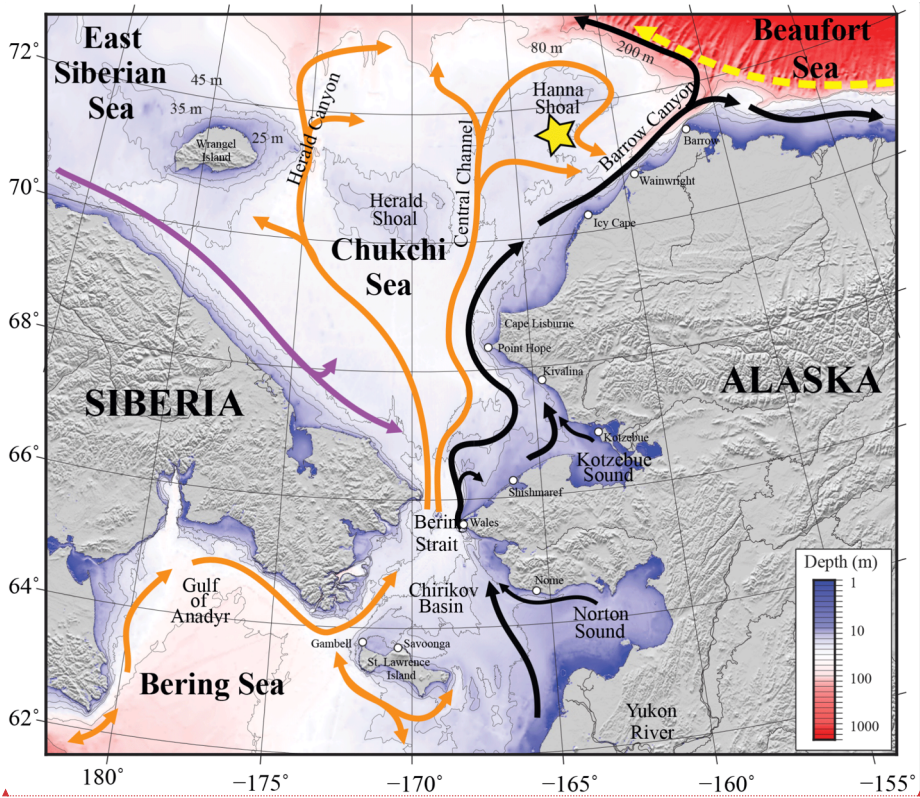
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Date	Cruise	Cast No.	Distance (km)	$pH^{disc}_{calc} \pm u_c$	Anomaly ($pH^{est} - pH^{disc}_{calc}$)	Anomaly ($pH_{SeaFET} - pH^{disc}_{calc}$)	Source
2017-09-10	HLY1702	127	0.52	8.0123±0.0166	-0.0450*	-0.0354	Cross et al., 2020a
2019-08-11	HLY1901	39	3.75	7.6423±0.012	0.0079*	-	Cross et al., 2021
2019-08-19	OS1901	33	0.27	7.7367±0.0145	-0.0200	-	unpublished

* indicates pH^{disc}_{calc} was interpolated to mooring depth

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3327 **Figure 1. Map of the study area.** Bathymetry of the Chukchi, northern Bering, East Siberian
 3328 and eastern Beaufort seas is shown in color. The Chukchi Ecosystem Observatory (CEO)
 3329 location near Hanna Shoal is marked with a yellow star. General circulation patterns are shown
 3330 with arrows: black – Alaskan Coastal Water and Alaskan Coastal Current, dividing into the
 3331 Shelf-break Jet (right) and Chukchi Slope Current (left, Corlett and Pickart, (2017)); orange –
 3332 Anadyr, Bering, and Chukchi Seawater; purple – Siberian Coastal Current; yellow – Beaufort
 3333 Gyre boundary current. Figure is from Hauri *et al.*, (2018).

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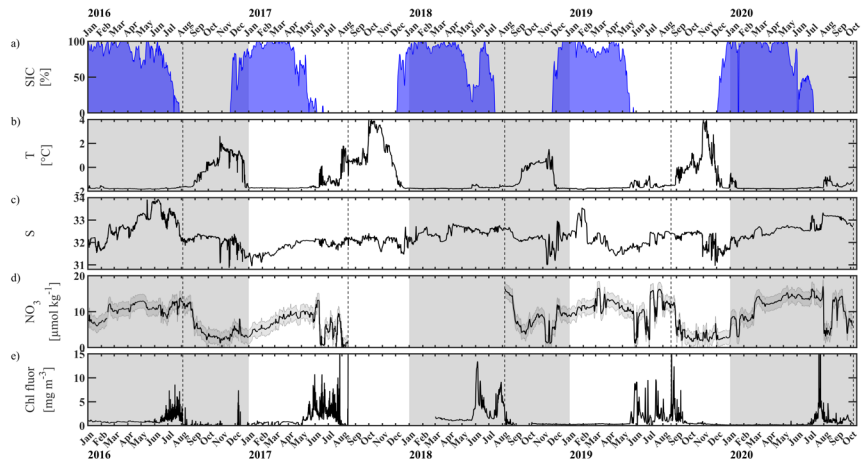
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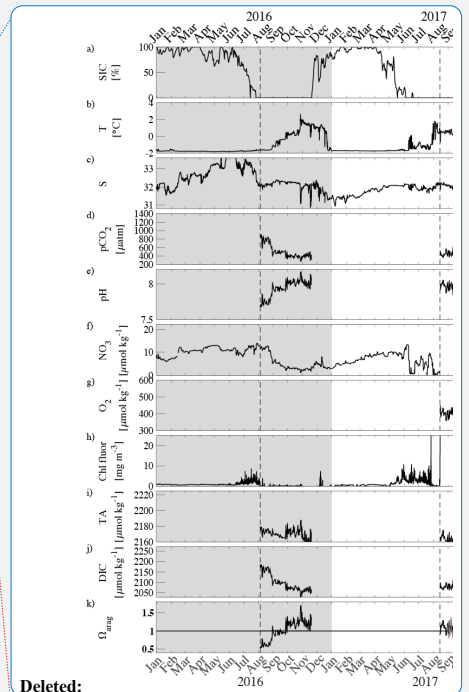
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3341 **Figure 2. Chukchi Ecosystem Observatory timeseries from 2016 through 2020.** a) sea ice
 3342 concentration (blue shading to highlight coverage. %; DiGirolamo et al., 2022), b) temperature
 3343 ($^{\circ}\text{C}$), c) salinity, d) NO_3 with uncertainty envelope ($\mu\text{mol kg}^{-1}$), and e) chlorophyll fluorescence
 3344 (mg m^{-3}). Years are indicated by alternating gray and white background shading. The vertical
 3345 dotted gray lines indicate the mooring turn around timing.



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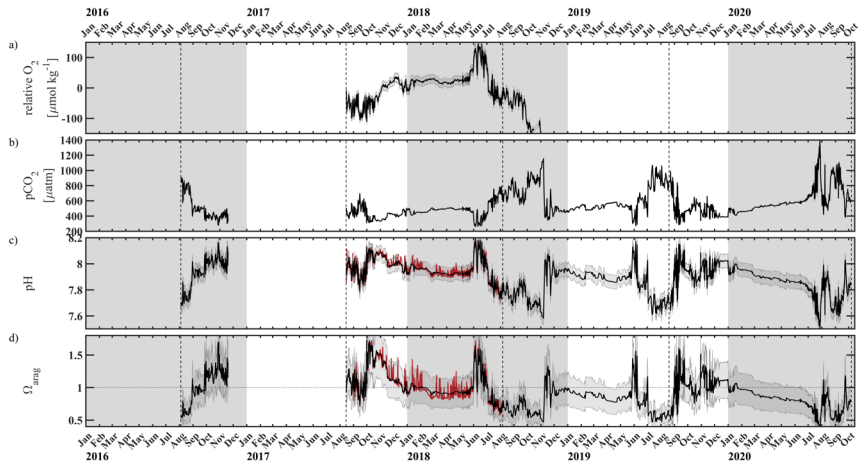
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Deleted: Chukchi Ecosystem Observatory timeseries from 2016 through 2020. Shown are a) sea ice concentration (%; DiGirolamo et al., 2022), b) temperature ($^{\circ}\text{C}$), c) salinity, d) pCO_2 (μatm ; Hauri and Irving, 2023a), e) pH (estimated in black, measured in gray; Hauri and Irving 2023b), f) NO_3 ($\mu\text{mol kg}^{-1}$), g) dissolved oxygen ($\mu\text{mol kg}^{-1}$), h) chlorophyll fluorescence (mg m^{-3}), i) total alkalinity ($\mu\text{mol kg}^{-1}$), j) dissolved inorganic carbon ($\mu\text{mol kg}^{-1}$), and k) aragonite saturation state (Ω_{arag}). Years are indicated by alternating grey and white background shading



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3361 [Figure 3. Chukchi Ecosystem Observatory timeseries from 2016 through 2020, part 2, a\)](#)

3362 [relative dissolved oxygen with uncertainty envelope \(relative to the mean; \$\mu\text{mol kg}^{-1}\$ \), b\) \$p\text{CO}_2\$](#)

3363 [with uncertainty envelope \(\$\mu\text{atm}\$; Hauri and Irving, 2023a\), c\) pH with uncertainty envelope](#)

3364 [\(\$\text{pH}^{\text{est}}\$ in black, \$\text{pH}_{\text{SeaFET}}\$ in red; Hauri and Irving 2023b\), and d\) aragonite saturation state with](#)

3365 [uncertainty envelope \(\$\Omega_{\text{arag}}\(p\text{CO}_2, \text{pH}^{\text{est}}\)\$ in black; \$\Omega_{\text{arag}}\(p\text{CO}_2, \text{pH}_{\text{SeaFET}}\)\$ in red\). Years are](#)

3366 [indicated by alternating gray and white backgrounds. The vertical dotted gray lines indicate the](#)

3367 [mooring turn around timing.](#)

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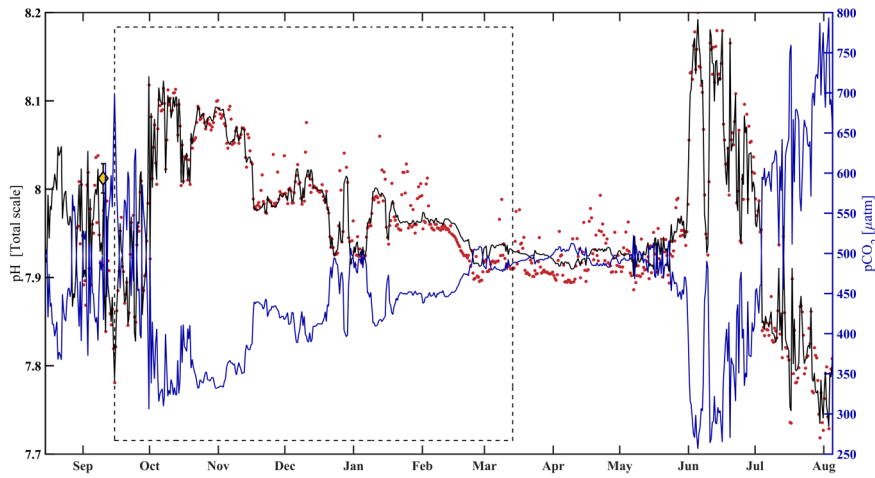
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3373 **Figure 4. HydroC pCO_2 and pH highlighting mirrored trend from mid-August 2017 to**

3374 **beginning of August 2018. Measured pH (pH_{ScaFET} , red dots) is interpolated onto the HydroC**

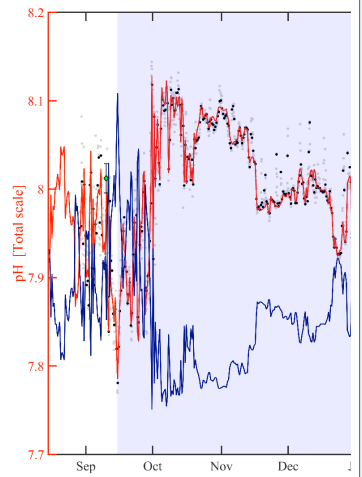
3375 **pCO_2 timestamp (blue), and pH^{est} is shown as the solid black line. The dashed box shows the**

3376 **period over which pH^{est} was trained. The yellow faced diamond with error bars show reference**

3377 **$pH^{disc}_{calc} \pm u_c$ (Table 2; Cross et al., 2020a; Orr et al., 2018).**

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Figure 3. HydroC pCO_2 (solid blue line) and SeaphOx pH (black and gray circles) highlighting mirrored trend from mid-August 2017 to beginning of August 2018.

Measured pH is shown at its original resolution (2 hr, gray circles) and interpolated onto the HydroC timestamp (12 hr, black circles), and estimated pH is shown as the solid red line. The blue highlighted section shows the period over which estimated pH was trained... The green

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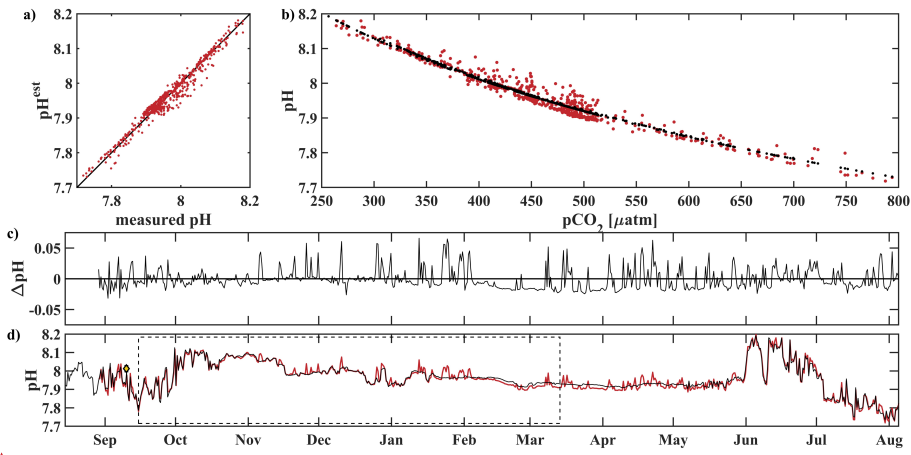
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3408 **Figure 5. Performance of the pH algorithm.** (a) pH_{SeaFET} vs pH^{est} with black line highlighting

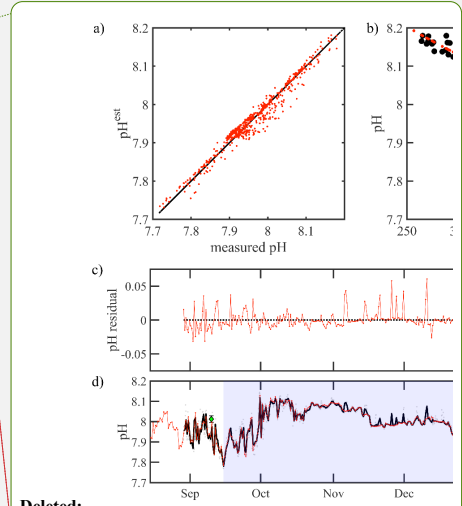
3409 1:1 ratio, (b) pCO_2 vs pH_{SeaFET} (red) and pCO_2 vs pH^{est} (black), (c) residual pH (pH_{SeaFET} -

3410 pH^{est}), and (d) pH_{SeaFET} (red) and pH^{est} (black) vs. time, with dashed box highlighting the period

3411 over which pH^{est} was trained (15 September - 14 March 2017), and the yellow faced diamond

3412 with error bars showing reference $pH^{disc}_{calc} \pm u_c$ (Table 2; Cross et al., 2020).

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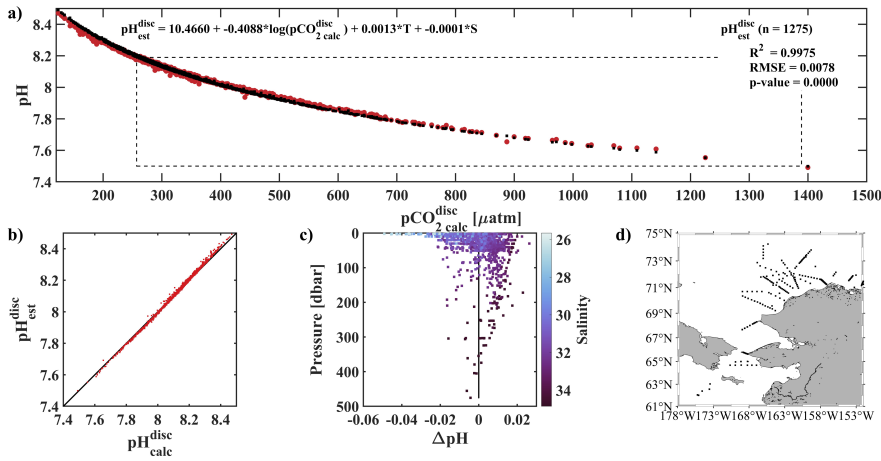
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Deleted: Evaluation of estimated pH. (a) measured pH vs estimated pH (pH^{est}), (b) measured pCO_2 vs pH (black) and pCO_2 vs pH^{est} (red), (c) residual pH (measured pH - pH^{est}) and (d) measured pH (black) and pH^{est} (red) vs. time. The blue highlighted section in (d) shows the period over which pH^{est} was trained (15 September - 14 March 2017) with $pH^{disc}_{calc} \pm u_c$ for reference (green faced diamond with error bar showing combined standard uncertainty; Cross et al., 2020a; Orr et al., 2018).

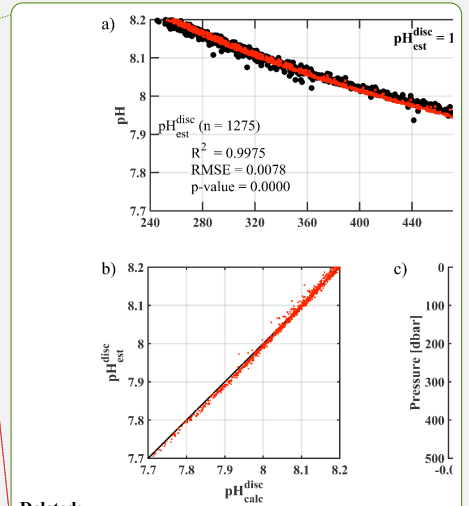
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3442 **Figure 6. Evaluation of the pH algorithm.** pH^{disc}_{est} evaluation with pH^{disc}_{calc} from discrete
 3443 samples collected during 4 cruises in the fall or early winter (August - November) of 2017-2020
 3444 and pH^{disc}_{est} from our linear regression model (Equation 2). (a) $pCO_2^{disc}_{calc}$ (TA, DIC) vs pH (red
 3445 pH^{disc}_{calc} and black pH^{disc}_{est}) with dashed black box showing the range of pH and pCO_2 observed
 3446 at the CEO at 33 m depth. (b) pH^{disc}_{calc} vs pH^{disc}_{est} with black 1:1 ratio. (c) residual pH ($pH^{disc}_{calc} -$
 3447 pH^{disc}_{est}) vs depth with color shading by salinity and black vertical line at 0, and (d) map showing
 3448 the locations of the 1275 discrete water samples used for evaluation (Monacci et al., 2022; Cross
 3449 et al., 2021; 2020a; 2020b).

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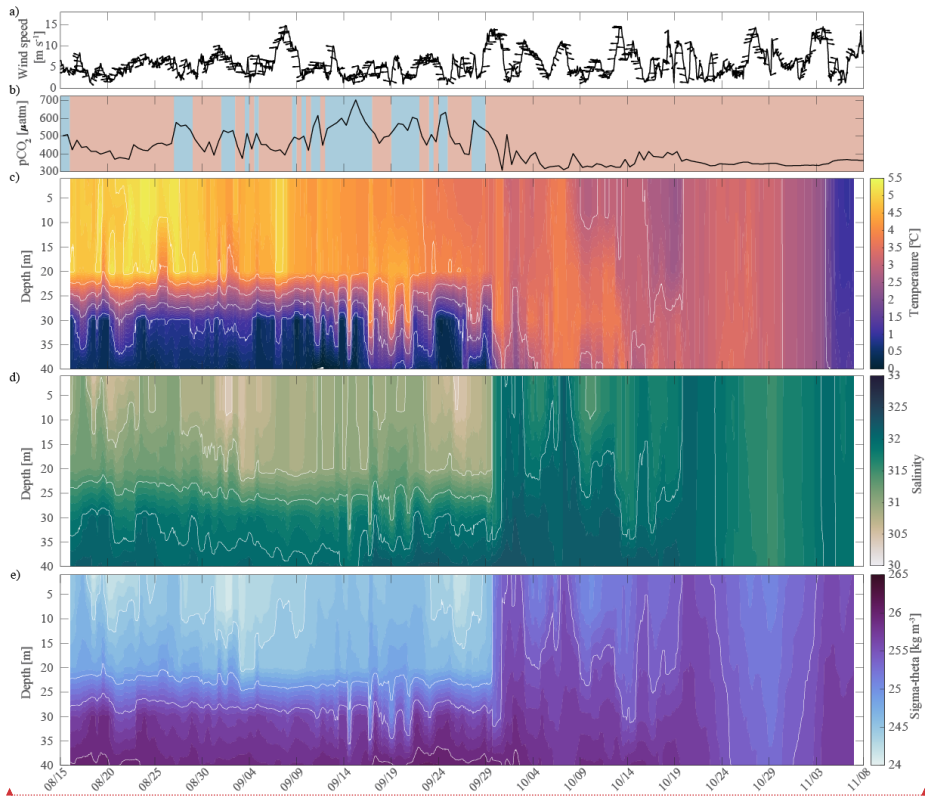
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Deleted: pH algorithm evaluation with pH from discrete samples collected in fall 2017-2020 and pH estimated using our linear regression model. (a) calculated pCO_2 (TA, DIC) vs pH (black pH^{disc}_{calc} and red pH^{disc}_{est}), (b) pH^{disc}_{calc} vs. pH^{disc}_{est} , (c) residual pH ($pH^{disc}_{calc} - pH^{disc}_{est}$) vs depth with color shading by salinity, and (d) map showing locations of 1275 discrete water samples used for evaluation (Monacci et al., 2022; Cross et al., 2021; 2020a; 2020b).

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3487 **Figure 7. Water column structure from late summer 2017 to freeze up.** Profiles of a) wind

3488 speed and direction (arrows pointing downwind) from the NOAA-operated Wiley Post-Will

3489 Rogers Memorial Airport, b) $p\text{CO}_2$ (μatm) with blue background indicating the water was

3490 undersaturated regarding aragonite ($\Omega_{\text{arag}} < 1$) and red shading indicating aragonite

3491 oversaturation ($\Omega_{\text{arag}} \geq 1$), c) temperature ($^{\circ}\text{C}$), d) salinity, and e) sigma-theta (kg m^{-3}).

3492 Temperature (c) and salinity (d) were measured at 8, 20, 30, and 40 m by the Chukchi Ecosystem

3493 Observatory freeze-up detection mooring deployed in fall 2017. Density was calculated with the

3494 TEOS-10 GSW Oceanographic Toolbox (McDougall and Baker, 2011).

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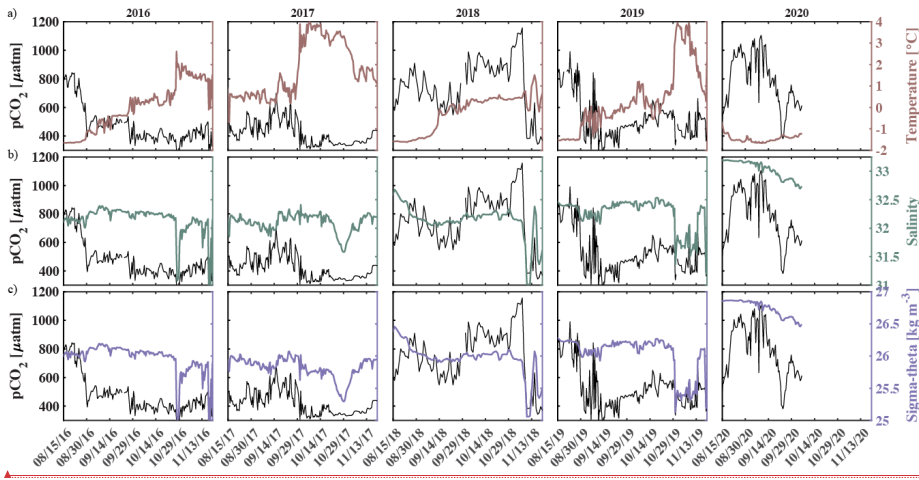


Figure 8. Impact of water column mixing on $p\text{CO}_2$. Timeseries of $p\text{CO}_2$ (black, left axis) and a) temperature (maroon, right axis), b) salinity (green, right axis), and c) density (purple, right axis) for 15 August to 1 December in 2016 -2020 measured at ~33m septh at the Chukchi Sea Ecosystem Observatory.

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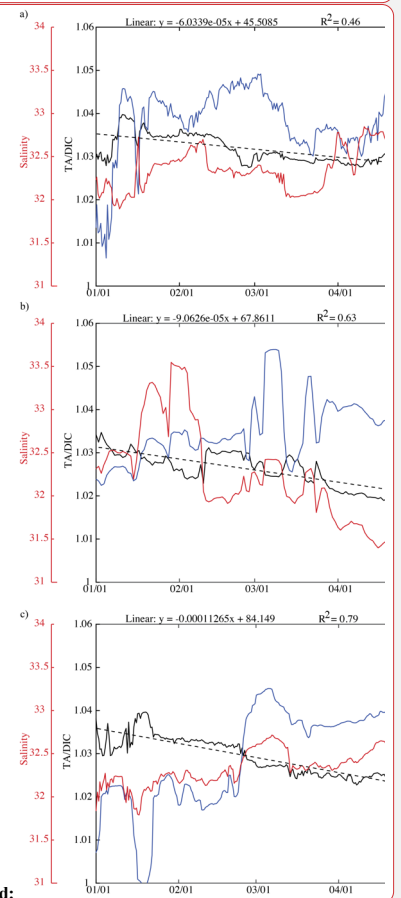
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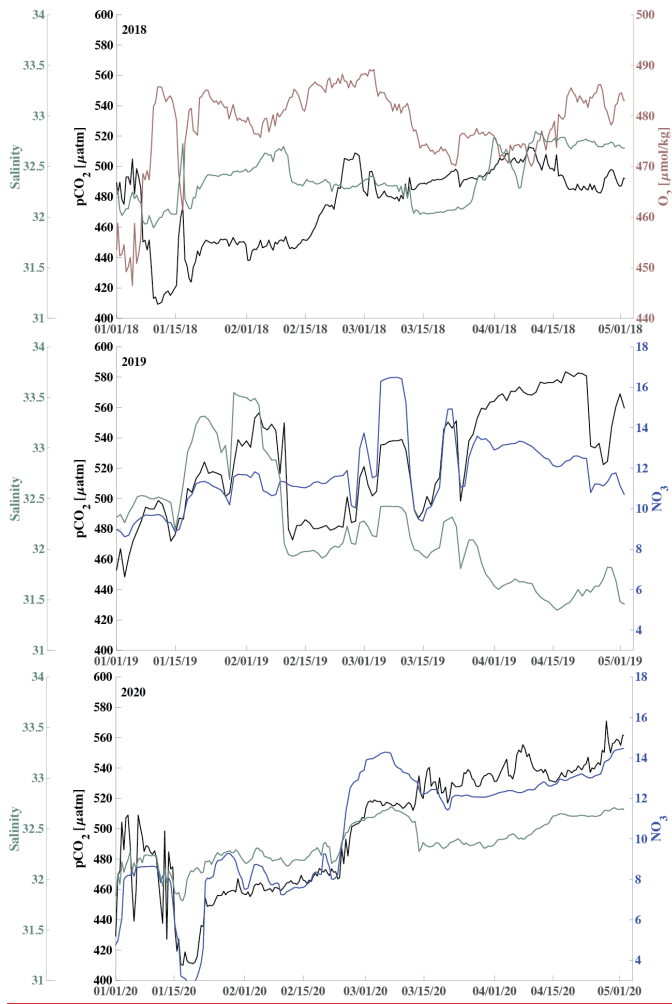
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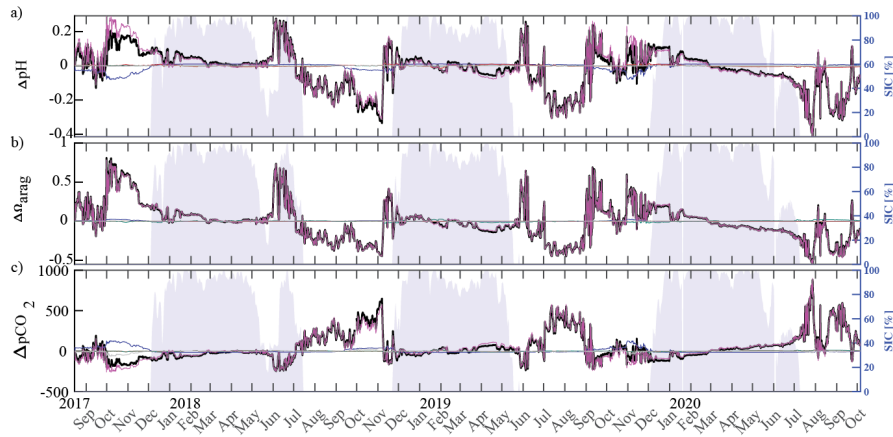
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3521 **Figure 9. Respiration under the sea ice. Timeseries of $p\text{CO}_2$ (black) and salinity (green, left**
 3522 **axis), and oxygen (O_2 , $\mu\text{mol kg}^{-1}$, maroon, top) and nitrate (NO_3 , $\mu\text{mol kg}^{-1}$, blue, middle and**
 3523 **bottom) concentration (right axis during January through April for 2018 (top), 2019 (middle) and**
 3524 **2020 (bottom).**

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- Deleted: Timeseries of TA/DIC ratio and salinity (left axis), and nitrate (NO_3 , $\mu\text{mol kg}^{-1}$) concentration (right axis) during January through April for 2018 (top), 2019 (middle) and 2020 (bottom).
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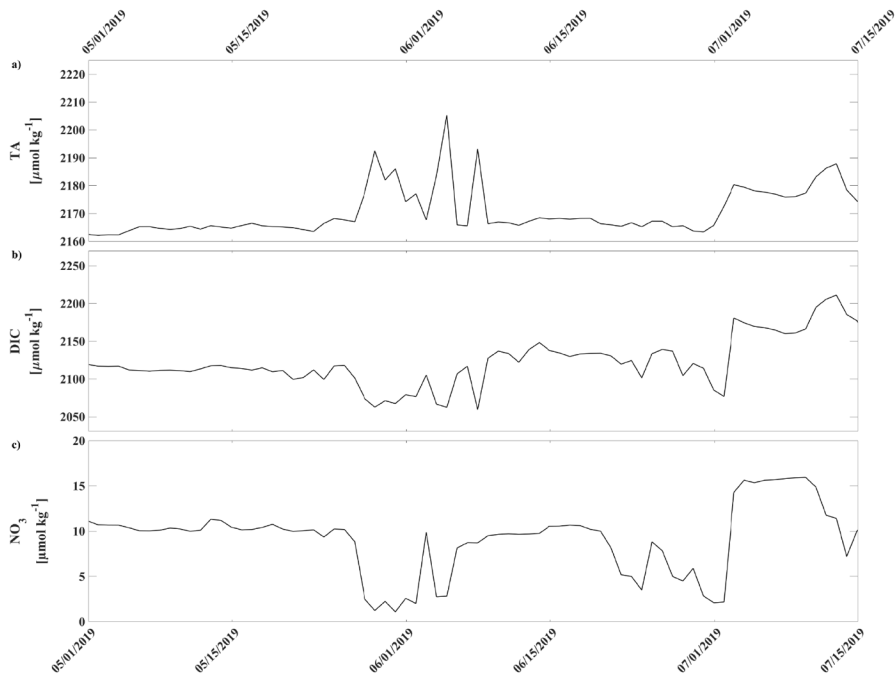
3533 **Figure 10. Drivers of the inorganic carbon system.** Component timeseries of the linear Taylor
3534 decomposition of a) pH, b) Ω_{arag} , and c) $p\text{CO}_2$. The perturbation effects due to salinity (red),
3535 temperature (blue), biogeochemistry (pink), and freshwater mixing (green), and an estimated
3536 residual term (grey) were computed following Rheuban et al. (2019). Sea ice concentration (blue
3537 shading, %; DiGirolamo et al., 2022) is shown on the right axes.

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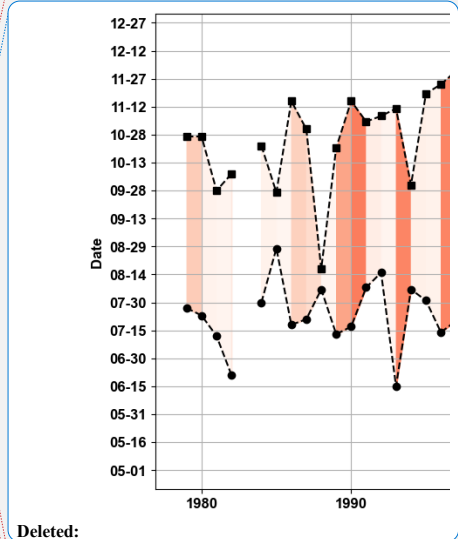
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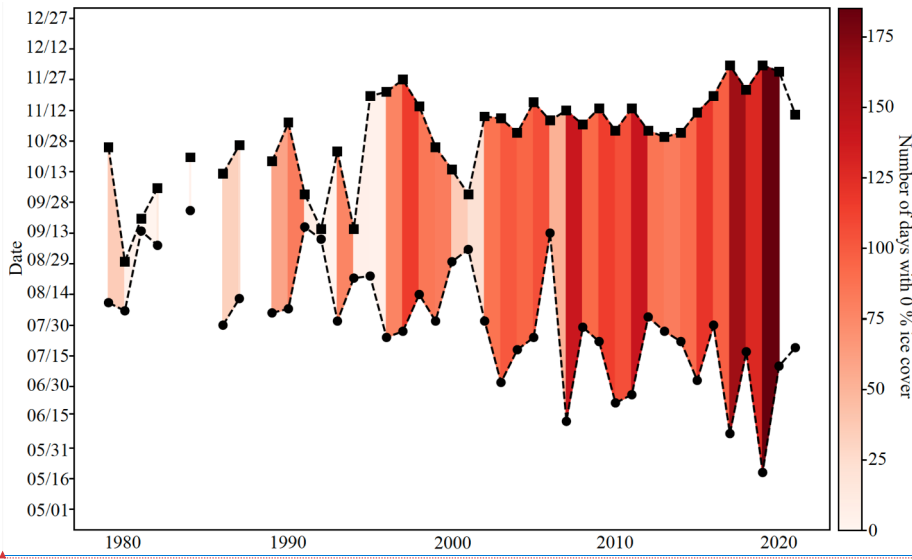
3545
 3546 **Figure 11. Spring 2019 relaxation event.** Timeseries of a) total alkalinity (TA, $\mu\text{mol kg}^{-1}$), b)
 3547 dissolved inorganic carbon (DIC, $\mu\text{mol kg}^{-1}$), and c) nitrate (NO_3 , $\mu\text{mol kg}^{-1}$) from May 1st, 2019
 3548 through July 15th, 2019.

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3553 **Figure 12. Low sea ice period at the Chukchi Sea Observatory.** Timeseries of start (circle)

3554 and end (square) of low sea ice (<15% per grid cell) period from 1982-2021. Shades of red

3555 illustrate number of days with 0% sea ice cover. The satellite sea ice cover at the observatory

3556 site was taken from the NSIDC (DiGirolamo [et al.](#), 2022).

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