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Supplemental Online Material for

Ocean and Coastal Acidification off New England and Nova Scotia

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Supplemental Methods

Primary Controls on NECAN Seasonal Dynamics

Time-series analyses were evaluated at each of the locations denoted in Figure 4 using actual discrete time-series data where available (western Gulf of Maine, Long Island Sound) or by extracting estimates using the gridded data product produced by Signorini et al. (2013).

Signorini-Based Estimates

Figure 4 denotes the estimated individual effects of CO_2 solubility (SOL), air-sea CO_2 flux (AS), mixing (MIX) and biologic activity (BIO) on Ω_a at six New England/Nova Scotia locations. . These estimates were derived from the calculations made using modeled 12-month climatologies of sea surface temperature (SST), sea surface salinity (SSS), total alkalinity (TA), $pCO_{2,air\text{-}sea}$ CO_2 flux (FLUX), and surface mixed-layer depth (MLD). BIO was determined by combining December TA, $pCO_{2,sea}$, SST, and SSS to derive dissolved inorganic carbon ($DIC_{TA\text{-}pCO2}$) using the CO2SYS program (Lewis and Wallace, 1998; K_1 and K_2 of Millero, 2010; KSO_4 of Dickson, 1990; TB [total boron] of Uppstrom, 1974), then deriving $pCO_{2,sea,TA\text{-}DIC}$ and $\Omega_{arag,TA\text{-}DIC}$ at monthly SST and SSS using December TA and $DIC_{TA\text{-}pCO2}$. AS was determined by adding (for release of CO_2 to the atmosphere) or removing (for uptake of CO_2 from the atmosphere) the DIC (dissolved inorganic carbon) represented by each monthly FLUX out of or into the MLD ($FLUX_{DIC}$) to the DIC calculated for the previous month, then deriving $pCO_{2,sea}$ and Ω_{arag} from monthly TA and $DIC+FLUX_{DIC}$. AS MLD depth was held to at least 5 m, which is was arbitrarily chosen as a logical minimum depth over which CO_2 is added or removed. We note that during the strongly stratified summer season, 5 m can be 1–3 m less than the pycnocline. MIX was determined using the Gulf of Maine TA-SSS and DIC-SSS using regressions taken

from Table 1 of Wang et al. (2013) using a TA:DIC ratio of 1:07 TA = 37.3*SSS + 998; DIC = 34.6*SSS + 933. Monthly TA and DIC were calculated according to these regressions (TA_{Wang} and DIC_{Wang}, respectively), with changes from month n-1 to month n calculated as $\Delta TA_{Wang\text{-}n} = TA_{Wang\text{-}n} - TA_{Wang\text{-}n\text{-}1}$ and $\Delta DIC_{Wang\text{-}n} = DIC_{Wang\text{-}n} - DIC_{Wang\text{-}n\text{-}1}$. Then MIX pCO_2 and Ω_a were derived from monthly SSS together with TA and DIC at month n: $TA_n=TA_{n=0} + \Delta TA_{Wang\text{-}n}$ and $DIC_n= DIC_{n=0} + \Delta DIC_{Wang\text{-}n}$. BIO was calculated as the residual between the climatologies of pCO_2 and Ω_a and the SOL, AS and MIX terms: $BIO_{pCO2}=pCO_2 - [(SOL - pCO_2) + (AS - pCO_2) + (MIX - pCO_2)]$.

Western Gulf of Maine Data

The calculated individual effects on $pCO_{2,sw}$ and Ω_a of changes in CO_2 solubility (SOL), air-sea CO_2 flux (AS), and mixing (MIX) at UNH Buoy D were calculated as above. SSS, SST, pCO_2, and FLUX source data were monthly climatologies derived from 2006–2014 buoy observations. MLD source data was a monthly climatology derived from 2004–2014 shipboard salinity and temperature profiles (n = 151) in the region of UNH Buoy D, again limited to 5 m or deeper. TA was derived from salinity according to a locally derived regression: TA = (SSS*52.24) + 476.3 (unpublished data from author Joe Salisbury).

Long Island Sound Data

The calculated individual effects on pCO_2 and Ω_{arag} of changes in CO_2 solubility (SOL), air-sea CO_2 flux (AS), and mixing (MIX) in Long Island Sound (LIS) were calculated in the *Signorini-based estimate*. SSS, SST, and TA source data were monthly climatologies. A monthly climatology of pH (NBS scale), paired with TA, SSS, and SST was used with CO2SYS as above to generate a monthly pCO_2 climatology. FLUX was calculated from pCO_2, atmospheric $pCO2$

at Mauna Loa (398 µatm), and monthly winds, according to the k660 parameterization of Ho et al. (2006). MLD for LIS was from the same model employed in the *Signorini-based estimate* at location 41.25°N –71.25°W, and again limited to 5 m or deeper.

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