Supplemental Online Material for

Ocean and Coastal Acidification off
New England and Nova Scotia

R.C. Chambers, C.J. Gobler, C.W. Hunt, A.L. King, N.N. Price, S.R. Signorini, E. Stancioff, C. Stymiest,

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 CO₂ solubility (SOL), air-sea CO₂ flux (AS), mixing (MIX) and biologic activity (BIO) on $\Omega_a$ 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), $pCO_{2,air-sea}$, CO₂ 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$_{TA-pCO2}$) using the CO2SYS program (Lewis and Wallace, 1998; $K_1$ and $K_2$ of Millero, 2010; KSO₄ of Dickson, 1990; TB [total boron] of Uppstrom, 1974), then deriving $pCO_{2,sea-TA-DIC}$ and $\Omega_{arag,TA-DIC}$ at monthly SST and SSS using December TA and DIC$_{TA-pCO2}$. AS was determined by adding (for release of CO₂ to the atmosphere) or removing (for uptake of CO₂ 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 $\Omega_{arag}$ 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 CO₂ 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$_{\text{Wang}}$ and DIC$_{\text{Wang}}$, respectively), with changes from month n-1 to month n calculated as $\Delta$TA$_{\text{Wang}}$-n = TA$_{\text{Wang}}$-n - TA$_{\text{Wang}}$-n-1 and $\Delta$DIC$_{\text{Wang}}$-n = DIC$_{\text{Wang}}$-n - DIC$_{\text{Wang}}$-n-1. Then MIX $p$CO$_2$ and $\Omega_a$ were derived from monthly SSS together with TA and DIC at month n: TA$_n$=TA$_n$=0 + $\Delta$TA$_{\text{Wang}}$-n and DIC$_n$= DIC$_{n=0}$ + $\Delta$DIC$_{\text{Wang}}$-n. BIO was calculated as the residual between the climatologies of $p$CO$_2$ and $\Omega_a$ and the SOL, AS and MIX terms: BIO$_{p$CO$_2}=p$CO$_2$ - [(SOL - $p$CO$_2$) + (AS - $p$CO$_2$) + (MIX - $p$CO$_2$)].

**Western Gulf of Maine Data**

The calculated individual effects on $p$CO$_2$$_{sw}$ and $\Omega_a$ 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, $p$CO$_2$, 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 $p$CO$_2$ and $\Omega_{arag}$ 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 $p$CO$_2$ climatology. FLUX was calculated from $p$CO$_2$, atmospheric $p$CO$_2$
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.

References
Dickson, A.G. 1990. Standard potential of the reaction: \( \text{AgCl(s)} + \frac{1}{2}\text{H}_2\text{(g)} = \text{Ag(s)} + \text{HCl(aq)} \), and the standard acidity constant of the ion \( \text{HSO}_4^- \) in synthetic sea water from 273.15 to 318.15 K. *Journal of Chemical Thermodynamics* 22:113–127, http://dx.doi.org/10.1016/0021-9614(90)90074-Z.


