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Supporting Online Material for

A Time-Series View of Changing Surface Ocean Chemistry Due to Ocean Uptake of Anthropogenic CO_2 and Ocean Acidification

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Some of the highest rates of change in seawater CO_2-carbonate chemistry have been observed in the North Pacific Ocean (e.g., Wakita et al., 2005; Sabine et al., 2008; Supplementary Table S1), as well as some of the lowest rates of change (Watanabe et al., 2011; Midorikawa et al., 2012). At the Kyodo North Pacific Time-series (KNOT), trends determined over the longest time scale (> 10 to 17 years) tend to converge on values of about +1.0 to +1.3 µmol kg^{-1} yr^{-1} (e.g., Wakita et al., 2010) similar to the nDIC trends at the seven ocean CO_2 time series. As discussed by Tanhua et al. (2013) and in the text, the shorter duration time-series data tend to have highly variable trends, reflecting the difficulties of assessing long-term trends in regions where there is considerable short-term spatio-temporal variability and insufficient reoccupation of sites to evaluate change over time. Interannual variability, associated with climatic variations such as the North Atlantic Oscillation, the El Niño-Southern Oscillation, and the Pacific Decadal Oscillation, also requires that observations be made over a sufficient duration to account for their influences.

Assessment of trends from annual reoccupation of ocean sites can also be complicated if the time of sampling is not the same each year, especially in ocean regions where there is large seasonal variability (refer to section in main text, How to Establish Trends in Seawater CO_2-Carbonate Chemistry Using Irregularly Sampled Seasonal Data). For example, at selected stations along line P in the North Pacific, the DIC trends during the period ~ 1992–2010 at open ocean sites P26 and P12 (P12 is closer to the Canadian coast at 131°W), based on 16 to 21 sampling points (Miller et al., 2010) are different at +1.65 ± 2.02 µmol kg^{-1} yr^{-1}, and +0.86 ± 2.78 µmol kg^{-1} yr^{-1}, respectively (note that the trends are not statistically different due to r^2 value of the trends < 0.05, reflecting a larger standard deviation of the trend due to ocean variability; Table S1). But, importantly, the standard errors of the trends were large (i.e., +2.08 to +2.78 µmol kg^{-1}) when compared to the ocean CO_2 time series (~ +0.07 to +0.42 µmol kg^{-1}; Table S1). Because sampling along Line P is not uniform in time each year, long-term trends are more highly influenced by larger seasonal variability (for DIC and other seawater CO_2-carbonate

Table S1. Comparison of trends in seawater dissolved inorganic carbon (DIC), normalized DIC (nDIC), and pCO_2 at (A) six fixed location time series, and (B) annual occupations of a time-series site or episodically repeated ocean sections typically separated in time by > 5 years.

Time-Series Site	Location	Period	DIC (µmol kg^{-1} yr^{-1})	nDIC (µmol kg^{-1} yr^{-1})	pCO_2 (µatm yr^{-1})	Reference
A. TIME-SERIES OCEAN CARBON CYCLE TRENDS						
Iceland Sea	68°N, 12.66°W	1985–2008	1.22 ± 0.27	0.93 ± 0.24	1.29 ± 0.36	Olafsson et al. (2010)
Irminger Sea	64.3°N, 28°W	1985–2010	1.62 ± 0.35	1.49 ± 0.35	2.37 ± 0.49	Olafsson et al. (2010)
BATS	32°N, 64°N	1983–2012	1.37 ± 0.07	1.12 ± 0.04	1.69 ± 0.11	Bates et al. (2012)
ESTOC	29.04°N, 15.5°W	1996–2012	1.09 ± 0.10	1.08 ± 0.08	1.92 ± 0.24	González-Dávila et al. (2010)
HOT	22°45'N, 158°W	1988–2012	1.78 ± 0.12	1.05 ± 0.05	1.72 ± 0.09	Dore et al. (2009)
CARIACO	10°30'N, 64°40'W	1996–2012	0.06 ± 0.42	1.43 ± 0.55	1.77 ± 0.43	Astor et al. (2013)
Munida	45.7°S, 171.5°E	1998–2012	0.88 ± 0.30	0.78 ± 0.30	1.28 ± 0.33	Currie et al. (2011)
B. ANNUAL OCCUPATIONS (¶) AND EPISODIC REPEATED OCEAN SECTIONS (§)						
¶ Norwegian Sea		2001–2006	1.3 +0.7	n/a	2.6 ±1.2	Skjelvan et al. (2008)
¶ Japan; 137°E; Kuroshio		1994–2008	1.23 + 0.40	n/a	1.54 ± 0.33	Ishii et al. (2011)
¶ Line P; North Pacific		1973–2005	n/a	n/a	1.36 ± 0.16	Wong et al. (2010)
§ N. Pacific	137°E; 3°N-33°N	1983–2007	0.96 +0.26	n/a	1.58 ± 0.12	Midorikawa et al. (2012)
§ KNOT	44°N, 155°E	1992–2002	1.0 to 1.3	n/a	0.5 to 2.5	Sabine et al. (2004)
§ KNOT	44°N, 155°E	1992–2001	1.3 to 2.3	n/a	1.9 ± 0.7	Wakita et al. (2005)
§ KNOT	44°N, 155°E	1999–2006	0.86 +0.12	n/a	n/a	Watanabe et al. (2011)
§ KNOT and K2	44°N, 155°E	1992–2009	1.3 +0.13	n/a	n/a	Wakita et al. (2010)
§ North Pacific	150°W, 30°N	1991–2006	1.5 to 2.0	n/a	1.0	Sabine et al. (2008)
§ Equatorial Pacific		1985–2004	n/a	n/a	1.5 ± 0.4	Ishii et al. (2009)
§ 149°E		1993–2005	0.9 to 1.1	n/a	n/a	Murata et al. (2009)
§ Rockall	52–56°N, 11–21°W	1991–2010	~1.0 + 0.3	n/a	n/a	McGrath et al. (2012)
§ Subarctic Pacific		1992–2008	1.3 to 1.5	n/a	0.7 ± 0.5	Wakita et al. (2010)
§ Southern Ocean Prime Meridian		1972–2008	0.12 (WSBW)	n/a	n/a	van Heuven et al. (2011)

parameters) than the secular changes due to uptake of anthropogenic CO_2 or natural ocean basin variability over time scales of a few decades. The trends in DIC increase (as well as other seawater CO_2-carbonate chemistry parameters) tend to increase along Line P and other annual or episodic reoccupations that have low statistical significance. Nonetheless, they have important and meaningful value when viewed in context of similar changes observed in the cohort of ocean CO_2 time-series.

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