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Supplement of

Ocean acidification in the subpolar North Atlantic: rates and mechanisms controlling pH changes

Maribel I. García-Ibáñez et al.

Correspondence to: Maribel I. García-Ibáñez (maribelgarcia@iim.csic.es)

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Table S1. Temporal evolution (1991–2015) of the values (average \pm standard deviation) of in situ temperature (T_{is} , in $^{\circ}\text{C}$), salinity, oxygen (O_2 , in $\mu\text{mol}\cdot\text{kg}^{-1}$), pH in total scale at in situ conditions of temperature and pressure (pH_{Tis}), total alkalinity (A_T , in $\mu\text{mol}\cdot\text{kg}^{-1}$), total dissolved inorganic carbon (DIC, in $\mu\text{mol}\cdot\text{kg}^{-1}$) and anthropogenic CO_2 estimated with the φC_T^0 method (C_{ant} , in $\mu\text{mol}\cdot\text{kg}^{-1}$) for the layers considered in the Irminger basin (S1a) and the Iceland basin (S1b). The mean pressure (Press, in dbar) of the layer is also given. Values listed here were obtained by vertically and horizontally integrating each property within each layer, except for the pH_{Tis} that was calculated using integrated values of DIC and A_T and referred to the mean Press of the layer over the studied time period to avoid the effects of the sampling strategy. Consult Fig. 1 of the main text for layer acronyms.

Year	Press	T_{is}	Salinity	O_2	pH_{Tis}	A_T	DIC	C_{ant}
SPMW ($\sigma_0 < 27.68 \text{ kg}\cdot\text{m}^{-3}$)								
1991	272	5.49 ± 0.67	34.988 ± 0.055	279 ± 10	8.056 ± 0.016	2308 ± 3	2145 ± 6	34.2 ± 4.0
1994	242	5.51 ± 1.22	34.916 ± 0.314	276 ± 16	8.061 ± 0.025	2305 ± 14	2140 ± 15	33.2 ± 4.6
1997	277	5.62 ± 1.28	34.957 ± 0.126	273 ± 13	8.054 ± 0.020	2306 ± 6	2144 ± 9	32.8 ± 6.3
2002	290	5.77 ± 1.21	34.986 ± 0.160	273 ± 17	8.039 ± 0.027	2307 ± 10	2150 ± 12	39.6 ± 5.8
2004	309	5.74 ± 1.17	34.976 ± 0.201	274 ± 18	8.034 ± 0.027	2307 ± 12	2152 ± 14	42.4 ± 6.3
2006	316	5.95 ± 1.17	35.006 ± 0.110	273 ± 16	8.033 ± 0.023	2306 ± 6	2150 ± 10	41.8 ± 6.1
2008	234	5.81 ± 1.78	35.005 ± 0.372	280 ± 20	8.031 ± 0.030	2309 ± 18	2154 ± 15	47.7 ± 6.3
2010	267	5.88 ± 1.29	35.003 ± 0.304	274 ± 19	8.027 ± 0.035	2310 ± 14	2156 ± 18	46.4 ± 6.8
2012	201	5.43 ± 1.45	34.955 ± 0.315	279 ± 18	8.022 ± 0.033	2307 ± 16	2158 ± 17	48.6 ± 5.9
2014	243	5.36 ± 1.31	34.950 ± 0.313	280 ± 21	8.024 ± 0.028	2307 ± 12	2158 ± 13	48.5 ± 5.2
2015	252	5.43 ± 0.41	35.019 ± 0.035	294 ± 8	8.018 ± 0.015	2310 ± 4	2162 ± 3	57.3 ± 3.6
uLSW ($27.68 \leq \sigma_0 < 27.76 \text{ kg}\cdot\text{m}^{-3}$)								
1991	631	3.72 ± 0.39	34.898 ± 0.030	292 ± 10	8.047 ± 0.013	2302 ± 2	2147 ± 4	30.3 ± 3.9
1994	601	3.73 ± 0.39	34.891 ± 0.034	281 ± 9	8.033 ± 0.017	2302 ± 2	2152 ± 3	29.4 ± 4.7
1997	734	3.72 ± 0.40	34.886 ± 0.036	283 ± 11	8.034 ± 0.014	2302 ± 2	2152 ± 5	28.5 ± 5.4
2002	818	3.90 ± 0.35	34.903 ± 0.031	280 ± 10	8.021 ± 0.014	2303 ± 2	2156 ± 3	32.0 ± 4.0
2004	910	3.86 ± 0.40	34.898 ± 0.037	278 ± 10	8.013 ± 0.009	2303 ± 3	2160 ± 4	33.7 ± 3.2
2006	868	3.97 ± 0.35	34.913 ± 0.030	280 ± 8	8.015 ± 0.009	2301 ± 2	2157 ± 3	33.6 ± 3.4
2008	808	4.02 ± 0.32	34.915 ± 0.027	286 ± 9	8.015 ± 0.012	2304 ± 2	2160 ± 3	39.9 ± 4.8
2010	856	4.03 ± 0.35	34.919 ± 0.033	281 ± 9	8.009 ± 0.009	2304 ± 2	2162 ± 3	38.5 ± 3.8
2012	769	4.07 ± 0.32	34.925 ± 0.027	285 ± 9	8.007 ± 0.012	2304 ± 2	2162 ± 2	42.3 ± 4.8
2014	791	3.98 ± 0.27	34.909 ± 0.026	286 ± 10	8.006 ± 0.011	2304 ± 2	2163 ± 2	42.3 ± 5.5
2015	756	3.77 ± 0.28	34.904 ± 0.020	300 ± 10	8.010 ± 0.018	2304 ± 3	2162 ± 4	49.0 ± 5.6
cLSW ($27.76 \leq \sigma_0 < 27.81 \text{ kg}\cdot\text{m}^{-3}$)								
1991	1388	3.28 ± 0.26	34.885 ± 0.036	291 ± 8	8.023 ± 0.011	2301 ± 3	2148 ± 3	26.3 ± 4.4
1994	1298	3.10 ± 0.29	34.871 ± 0.034	294 ± 9	8.019 ± 0.019	2301 ± 2	2150 ± 3	26.5 ± 6.0
1997	1442	3.16 ± 0.30	34.876 ± 0.033	291 ± 9	8.018 ± 0.013	2301 ± 2	2151 ± 3	27.6 ± 4.4
2002	1441	3.38 ± 0.32	34.907 ± 0.033	281 ± 6	8.006 ± 0.008	2302 ± 2	2155 ± 2	26.1 ± 1.7
2004	1458	3.44 ± 0.27	34.914 ± 0.028	278 ± 6	7.999 ± 0.008	2304 ± 3	2159 ± 3	27.8 ± 1.5
2006	1439	3.56 ± 0.23	34.931 ± 0.028	278 ± 4	7.999 ± 0.007	2302 ± 2	2157 ± 2	27.1 ± 1.4
2008	1453	3.56 ± 0.19	34.929 ± 0.016	279 ± 4	7.997 ± 0.005	2306 ± 2	2160 ± 3	30.2 ± 1.9
2010	1444	3.61 ± 0.27	34.935 ± 0.028	277 ± 4	7.995 ± 0.007	2306 ± 2	2162 ± 2	30.0 ± 1.8
2012	1445	3.64 ± 0.17	34.940 ± 0.020	274 ± 3	7.989 ± 0.006	2306 ± 1	2163 ± 1	30.2 ± 1.8
2014	1404	3.67 ± 0.19	34.943 ± 0.017	274 ± 4	7.989 ± 0.008	2306 ± 1	2163 ± 2	29.8 ± 2.3
2015	1594	3.59 ± 0.12	34.929 ± 0.013	280 ± 1	7.988 ± 0.012	2305 ± 3	2164 ± 3	33.0 ± 2.2
ISOW ($27.81 \leq \sigma_0 < 27.88 \text{ kg}\cdot\text{m}^{-3}$)								
1991	2239	3.06 ± 0.23	34.937 ± 0.016	279 ± 4	8.005 ± 0.017	2306 ± 2	2150 ± 3	14.9 ± 3.3
1994	2210	2.94 ± 0.33	34.917 ± 0.020	280 ± 6	8.003 ± 0.011	2304 ± 3	2150 ± 4	19.0 ± 7.0
1997	2243	2.92 ± 0.23	34.910 ± 0.014	284 ± 5	7.998 ± 0.013	2305 ± 2	2153 ± 2	19.9 ± 2.6
2002	2177	2.95 ± 0.30	34.918 ± 0.021	281 ± 5	7.992 ± 0.012	2304 ± 2	2154 ± 2	19.6 ± 2.4
2004	2107	2.93 ± 0.39	34.916 ± 0.030	280 ± 7	7.985 ± 0.017	2306 ± 4	2159 ± 3	22.7 ± 2.4
2006	2117	3.04 ± 0.28	34.930 ± 0.015	281 ± 6	7.985 ± 0.014	2304 ± 2	2156 ± 2	22.2 ± 2.6
2008	2101	3.06 ± 0.24	34.932 ± 0.012	282 ± 4	7.983 ± 0.011	2306 ± 2	2159 ± 2	25.1 ± 2.2
2010	2108	3.04 ± 0.26	34.930 ± 0.016	281 ± 5	7.982 ± 0.013	2307 ± 2	2161 ± 2	24.9 ± 2.2
2012	2093	3.08 ± 0.26	34.935 ± 0.014	277 ± 4	7.976 ± 0.012	2307 ± 2	2162 ± 2	24.6 ± 2.2
2014	2034	3.09 ± 0.27	34.937 ± 0.017	277 ± 6	7.976 ± 0.012	2307 ± 2	2162 ± 2	24.2 ± 3.6
2015	2156	3.09 ± 0.13	34.935 ± 0.003	279 ± 1	7.976 ± 0.021	2307 ± 5	2162 ± 5	25.5 ± 6.8
DSOW ($\sigma_0 \geq 27.88 \text{ kg}\cdot\text{m}^{-3}$)								
1991	2857	2.00 ± 0.35	34.897 ± 0.011	294 ± 6	7.994 ± 0.015	2302 ± 2	2148 ± 4	14.6 ± 2.8
1994	2645	1.80 ± 0.34	34.873 ± 0.008	299 ± 5	7.990 ± 0.012	2300 ± 1	2148 ± 2	17.8 ± 3.8
1997	2853	1.98 ± 0.37	34.890 ± 0.009	296 ± 4	7.984 ± 0.007	2302 ± 2	2152 ± 2	19.2 ± 2.7
2002	2764	1.91 ± 0.35	34.886 ± 0.014	294 ± 6	7.982 ± 0.018	2301 ± 2	2152 ± 2	17.7 ± 1.6
2004	2773	1.71 ± 0.39	34.868 ± 0.023	299 ± 8	7.976 ± 0.016	2301 ± 3	2155 ± 3	22.0 ± 1.1
2006	2794	2.19 ± 0.30	34.907 ± 0.004	292 ± 6	7.973 ± 0.010	2303 ± 1	2155 ± 2	21.3 ± 1.6
2008	2750	2.21 ± 0.36	34.913 ± 0.005	294 ± 6	7.968 ± 0.011	2305 ± 2	2160 ± 2	25.1 ± 1.8
2010	2733	2.08 ± 0.36	34.896 ± 0.011	297 ± 7	7.970 ± 0.017	2304 ± 2	2158 ± 2	25.8 ± 1.6
2012	2718	2.17 ± 0.36	34.909 ± 0.008	292 ± 7	7.961 ± 0.012	2305 ± 2	2162 ± 2	25.9 ± 2.1
2014	2798	2.04 ± 0.42	34.905 ± 0.015	294 ± 8	7.961 ± 0.014	2304 ± 2	2162 ± 1	26.7 ± 2.5
2015	2739	1.94 ± 0.37	34.916 ± 0.011	290 ± 7	7.959 ± 0.014	2306 ± 3	2166 ± 2	25.1 ± 4.2

Table S1b. The same as S1a for the Iceland basin.

Year	Press	T_{is}	Salinity	O_2	pH_{Tis}	A_T	DIC	C_{ant}
SPMW ($\sigma_0 < 27.68 \text{ kg}\cdot\text{m}^{-3}$)								
1991	425	6.28 ± 1.19	34.988 ± 0.084	255 ± 12	8.024 ± 0.023	2308 ± 4	2151 ± 11	32.9 ± 7.1
1994	423	6.37 ± 1.17	34.986 ± 0.075	253 ± 14	8.023 ± 0.031	2309 ± 4	2151 ± 16	34.0 ± 7.6
1997	458	7.14 ± 1.44	35.081 ± 0.091	246 ± 13	8.030 ± 0.024	2314 ± 4	2147 ± 13	30.5 ± 11.1
2002	449	7.06 ± 1.25	35.076 ± 0.082	250 ± 20	8.019 ± 0.034	2312 ± 5	2151 ± 15	36.6 ± 10.6
2004	407	6.97 ± 1.53	35.060 ± 0.110	244 ± 18	8.006 ± 0.034	2311 ± 6	2156 ± 17	37.3 ± 10.3
2006	411	7.14 ± 1.43	35.051 ± 0.085	243 ± 20	8.003 ± 0.037	2310 ± 5	2155 ± 18	38.5 ± 10.0
2008	423	6.82 ± 1.35	35.063 ± 0.100	253 ± 14	8.006 ± 0.026	2314 ± 5	2160 ± 12	43.3 ± 10.0
2010	410	6.70 ± 1.30	35.029 ± 0.112	249 ± 17	7.999 ± 0.027	2311 ± 6	2160 ± 14	42.1 ± 9.6
2012	426	6.70 ± 1.09	35.028 ± 0.080	250 ± 18	7.995 ± 0.025	2309 ± 5	2160 ± 12	43.0 ± 10.5
2014	406	6.64 ± 1.15	35.043 ± 0.077	254 ± 18	7.998 ± 0.029	2313 ± 4	2163 ± 13	45.9 ± 10.7
uLSW ($27.68 \leq \sigma_0 < 27.76 \text{ kg}\cdot\text{m}^{-3}$)								
1991	957	4.19 ± 0.37	34.932 ± 0.033	264 ± 7	8.010 ± 0.006	2305 ± 2	2158 ± 3	24.0 ± 1.7
1994	947	4.19 ± 0.30	34.933 ± 0.021	259 ± 8	8.005 ± 0.008	2305 ± 2	2160 ± 3	24.5 ± 1.9
1997	1012	4.27 ± 0.26	34.943 ± 0.017	258 ± 7	8.009 ± 0.005	2306 ± 1	2159 ± 3	21.1 ± 1.5
2002	1002	4.28 ± 0.35	34.944 ± 0.032	260 ± 6	7.998 ± 0.007	2304 ± 2	2161 ± 2	26.0 ± 1.6
2004	967	4.15 ± 0.32	34.927 ± 0.028	263 ± 8	7.995 ± 0.005	2303 ± 3	2162 ± 3	28.1 ± 1.6
2006	960	4.21 ± 0.38	34.936 ± 0.033	265 ± 9	7.992 ± 0.007	2303 ± 4	2163 ± 4	30.2 ± 2.4
2008	1001	4.16 ± 0.36	34.928 ± 0.036	269 ± 8	7.994 ± 0.005	2306 ± 2	2165 ± 3	33.5 ± 2.0
2010	1005	4.17 ± 0.34	34.929 ± 0.034	270 ± 7	7.992 ± 0.005	2305 ± 3	2165 ± 3	34.8 ± 1.8
2012	1031	4.22 ± 0.31	34.934 ± 0.025	266 ± 7	7.986 ± 0.004	2303 ± 2	2165 ± 3	33.4 ± 1.3
2014	1009	4.22 ± 0.29	34.934 ± 0.023	268 ± 6	7.986 ± 0.004	2306 ± 2	2168 ± 3	36.3 ± 2.0
cLSW ($27.76 \leq \sigma_0 < 27.81 \text{ kg}\cdot\text{m}^{-3}$)								
1991	1665	3.45 ± 0.17	34.923 ± 0.027	277 ± 3	8.003 ± 0.012	2304 ± 3	2154 ± 2	20.9 ± 3.1
1994	1657	3.41 ± 0.16	34.916 ± 0.028	276 ± 3	7.995 ± 0.013	2304 ± 2	2157 ± 3	21.1 ± 3.1
1997	1710	3.39 ± 0.19	34.913 ± 0.023	278 ± 4	8.008 ± 0.009	2304 ± 2	2152 ± 2	20.2 ± 2.3
2002	1719	3.38 ± 0.22	34.915 ± 0.027	278 ± 4	7.996 ± 0.009	2303 ± 2	2155 ± 2	23.3 ± 2.6
2004	1769	3.35 ± 0.21	34.908 ± 0.022	277 ± 3	7.991 ± 0.008	2302 ± 3	2157 ± 2	24.2 ± 2.5
2006	1713	3.42 ± 0.22	34.917 ± 0.028	278 ± 2	7.990 ± 0.008	2302 ± 2	2157 ± 3	25.7 ± 3.4
2008	1746	3.43 ± 0.22	34.920 ± 0.025	279 ± 3	7.989 ± 0.008	2306 ± 2	2161 ± 2	28.0 ± 2.7
2010	1754	3.45 ± 0.24	34.923 ± 0.025	277 ± 2	7.987 ± 0.008	2306 ± 2	2161 ± 2	27.8 ± 3.1
2012	1742	3.52 ± 0.22	34.934 ± 0.024	273 ± 2	7.982 ± 0.006	2304 ± 2	2161 ± 2	26.2 ± 2.9
2014	1752	3.51 ± 0.23	34.934 ± 0.019	273 ± 1	7.982 ± 0.009	2307 ± 2	2164 ± 2	27.6 ± 3.2
ISOW ($\sigma_0 \geq 27.81 \text{ kg}\cdot\text{m}^{-3}$)								
1991	2493	2.93 ± 0.14	34.962 ± 0.012	269 ± 5	7.978 ± 0.017	2315 ± 6	2164 ± 6	15.2 ± 2.6
1994	2537	2.91 ± 0.13	34.958 ± 0.007	269 ± 5	7.976 ± 0.016	2314 ± 6	2164 ± 7	15.2 ± 5.5
1997	2557	2.91 ± 0.13	34.959 ± 0.008	272 ± 4	7.983 ± 0.015	2313 ± 5	2160 ± 6	14.7 ± 1.6
2002	2560	2.91 ± 0.13	34.960 ± 0.012	271 ± 5	7.972 ± 0.016	2313 ± 6	2164 ± 6	17.4 ± 2.2
2004	2650	2.88 ± 0.13	34.952 ± 0.017	270 ± 7	7.969 ± 0.013	2312 ± 8	2165 ± 7	17.7 ± 3.7
2006	2561	2.92 ± 0.13	34.960 ± 0.015	271 ± 5	7.968 ± 0.013	2312 ± 6	2165 ± 6	18.3 ± 3.0
2008	2568	2.94 ± 0.09	34.961 ± 0.013	273 ± 5	7.966 ± 0.017	2316 ± 4	2169 ± 5	22.1 ± 3.5
2010	2533	2.94 ± 0.13	34.960 ± 0.016	273 ± 5	7.964 ± 0.016	2314 ± 4	2169 ± 4	22.0 ± 4.2
2012	2489	2.98 ± 0.13	34.968 ± 0.015	270 ± 4	7.961 ± 0.015	2313 ± 4	2168 ± 5	20.7 ± 2.7
2014	2613	2.94 ± 0.12	34.964 ± 0.014	269 ± 6	7.961 ± 0.016	2318 ± 7	2173 ± 7	21.6 ± 3.1

Table S2. Changes of pH in total scale at in situ conditions of temperature and pressure (pH_{Tis}) caused by the changes in the drivers ($\frac{\partial \text{pH}_{\text{Tis}}}{\partial \text{var}}$) considered in the study (in situ temperature, T_{is} , in °C; salinity; total alkalinity, A_{T} , in $\mu\text{mol}\cdot\text{kg}^{-1}$; and total dissolved inorganic carbon, DIC, in $\mu\text{mol}\cdot\text{kg}^{-1}$) for the layers considered in the Irminger basin and the Iceland basin. Last row represents the average values for all layers. The ‘±’ values represent the error of the slope, except for the last row that represents the standard deviation of the values in all layers. Consult Fig. 1 of the main text for layer acronyms.

Layer	$\frac{\partial \text{pH}_{\text{Tis}}}{\partial T_{\text{is}}}$	$\frac{\partial \text{pH}_{\text{Tis}}}{\partial S}$	$\frac{\partial \text{pH}_{\text{Tis}}}{\partial A_{\text{T}}}$	$\frac{\partial \text{pH}_{\text{Tis}}}{\partial \text{DIC}}$
Irminger basin				
SPMW	-0.0160 ± 0.0048	-0.0120 ± 0.0007	0.0024 ± 0.0287	-0.0025 ± 0.0456
uLSW	-0.0161 ± 0.0020	-0.0120 ± 0.0002	0.0025 ± 0.0108	-0.0026 ± 0.0326
cLSW	-0.0160 ± 0.0019	-0.0121 ± 0.0002	0.0025 ± 0.0198	-0.0026 ± 0.0255
ISOW	-0.0158 ± 0.0014	-0.0122 ± 0.0002	0.0025 ± 0.0229	-0.0026 ± 0.0255
DSOW	-0.0157 ± 0.0032	-0.0123 ± 0.0003	0.0025 ± 0.0298	-0.0026 ± 0.0314
Iceland basin				
SPMW	-0.0159 ± 0.0082	-0.0119 ± 0.0009	0.0024 ± 0.0506	-0.0025 ± 0.0678
uLSW	-0.0159 ± 0.0012	-0.0119 ± 0.0002	0.0025 ± 0.0287	-0.0026 ± 0.0246
cLSW	-0.0159 ± 0.0012	-0.0121 ± 0.0002	0.0025 ± 0.0384	-0.0026 ± 0.0541
ISOW	-0.0157 ± 0.0005	-0.0122 ± 0.0001	0.0025 ± 0.0407	-0.0026 ± 0.0557
All	-0.0159 ± 0.0001	-0.0121 ± 0.0001	0.0024 ± 0.0001	-0.00257 ± 0.00005

Figure S1. Observed temporal changes of pH at total scale and in situ conditions (in situ temperature and pressure) along the section for the period 1991–2015. All the trends are calculated based on the annually interpolated values and are in 10^{-3} pH units \cdot yr $^{-1}$. The dashed vertical lines represent the Longitude axis marks, and isopycnals delineating the layers are shown as black lines.

