Supplementary

Ocean Acidification trends and Carbonate System dynamics in the North Atlantic Subpolar Gyre during 2009-2019

Curbelo-Hernández, D.[1], Pérez, F.F.[2], González-Dávila, M.[1,*], Gladyshev, S.[3], González, A. G.[1], González-Santana, D.[1], Velo, A.[2], Sokov, A.[3], and Santana-Casiano, J. M.[1].

[1] Instituto de Oceanografía y Cambio Global (IOCAG), Universidad de Las Palmas de Gran Canaria (ULPGC). Las Palmas de Gran Canaria, Spain.

[2] Instituto de Investigaciones Marinas (IIM), CSIC, Vigo, Spain.

[3] P. P. Shirshov Institute of Oceanology, Russian Academy of Sciences, Moscow, Russian Federation

*Corresponding Author: Melchor González-Dávila (melchor.gonzalez@ulpgc.es)
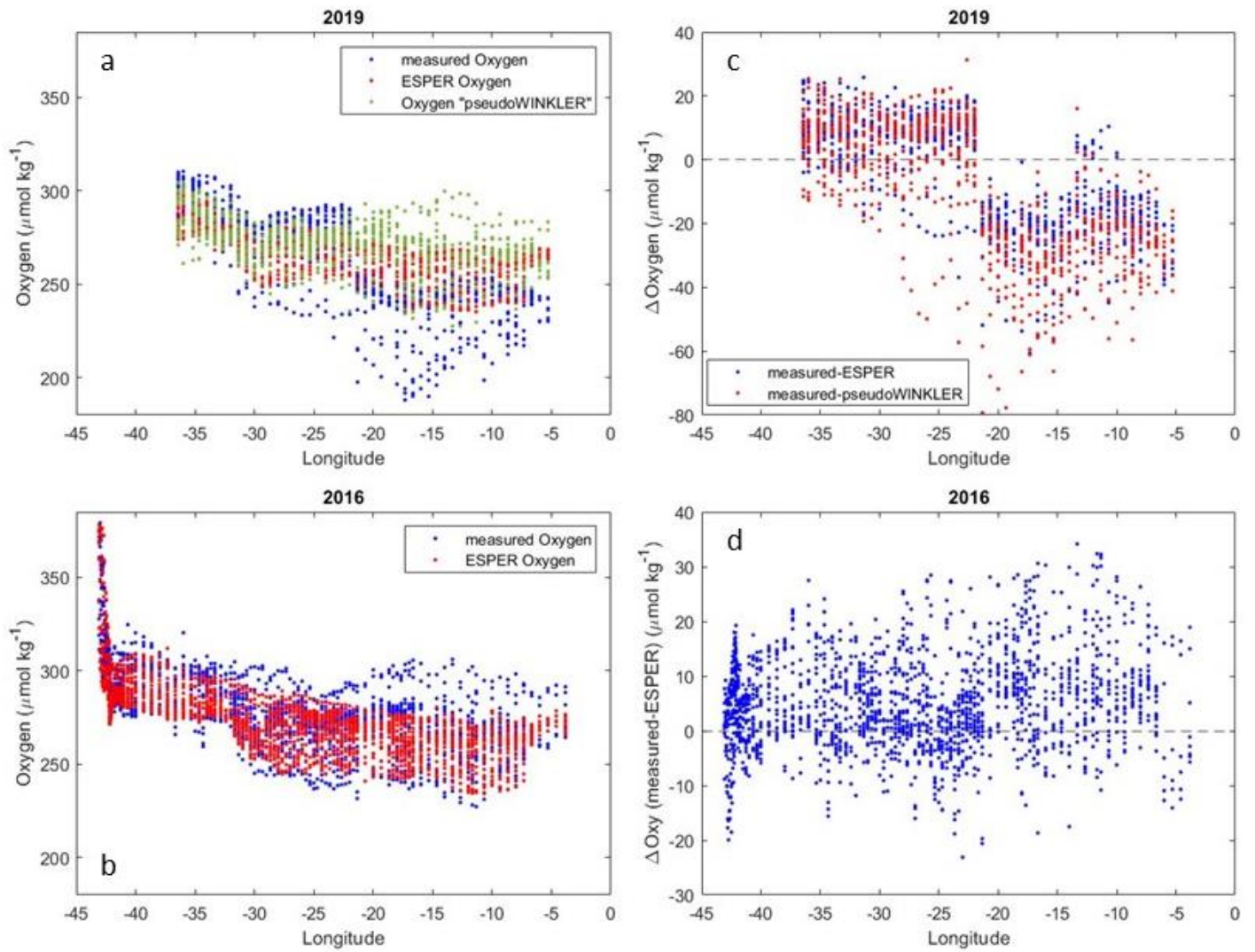
Figure S1. Longitudinal distribution of (a) sensor-measured, ESPER-estimated and pseudo-WIKLER oxygen data for 2019, (b) WINKLER-measured and ESPER-estimated data for 2016, (c) $\Delta DO_{meas\text{-}ESPER}$ and $\Delta DO_{meas\text{-}pseudoWINLKER}$ for the cruise of 2019 and (d) $\Delta DO_{meas\text{-}ESPER}$ for the cruise of 2016.
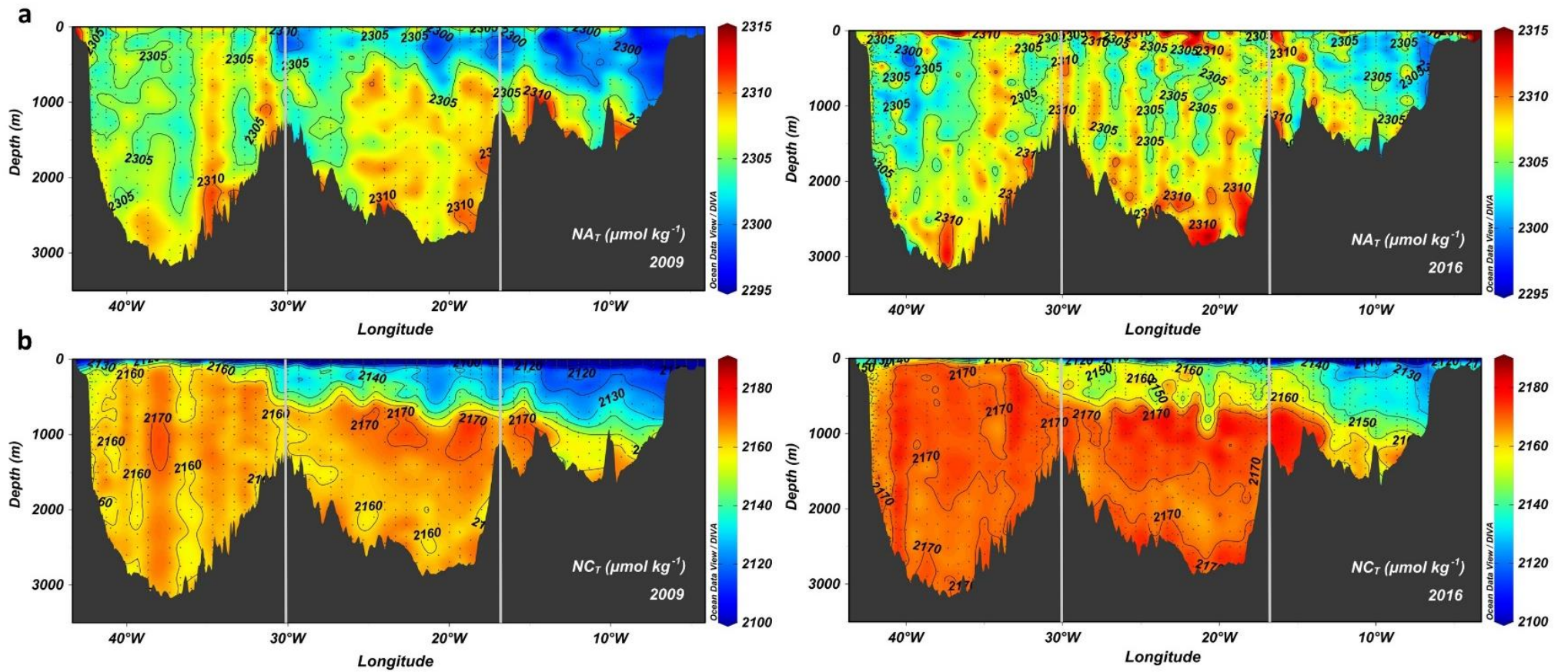
Figure S2. Water-column distribution along the longitudinal transect of (a) NA$_T$ and (b) NC$_T$ for the cruises of 2009 (left plots) and 2016 (right plots). The vertical while lines show the limits between basins.
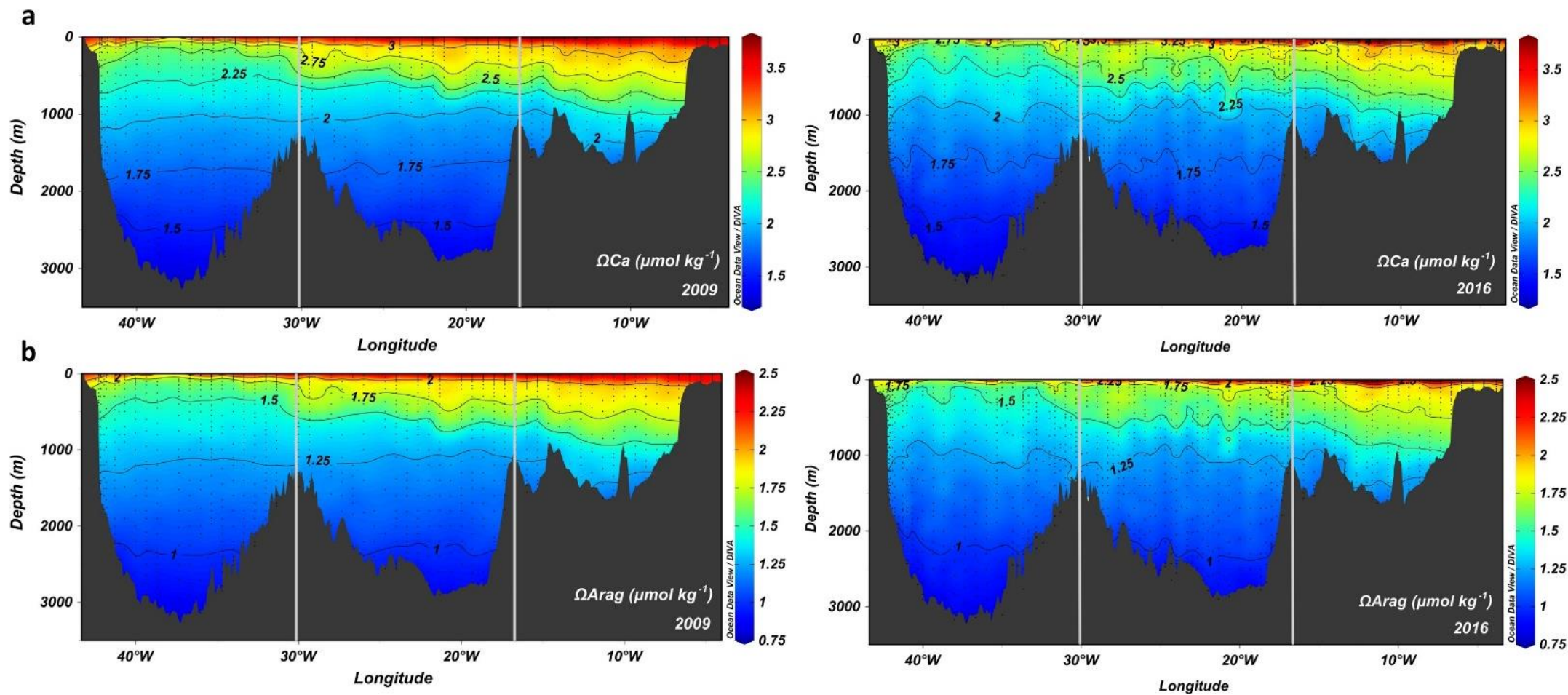
Figure S3. Same as Figure S2 but for (a) ΏCa and (b) ΏArag.
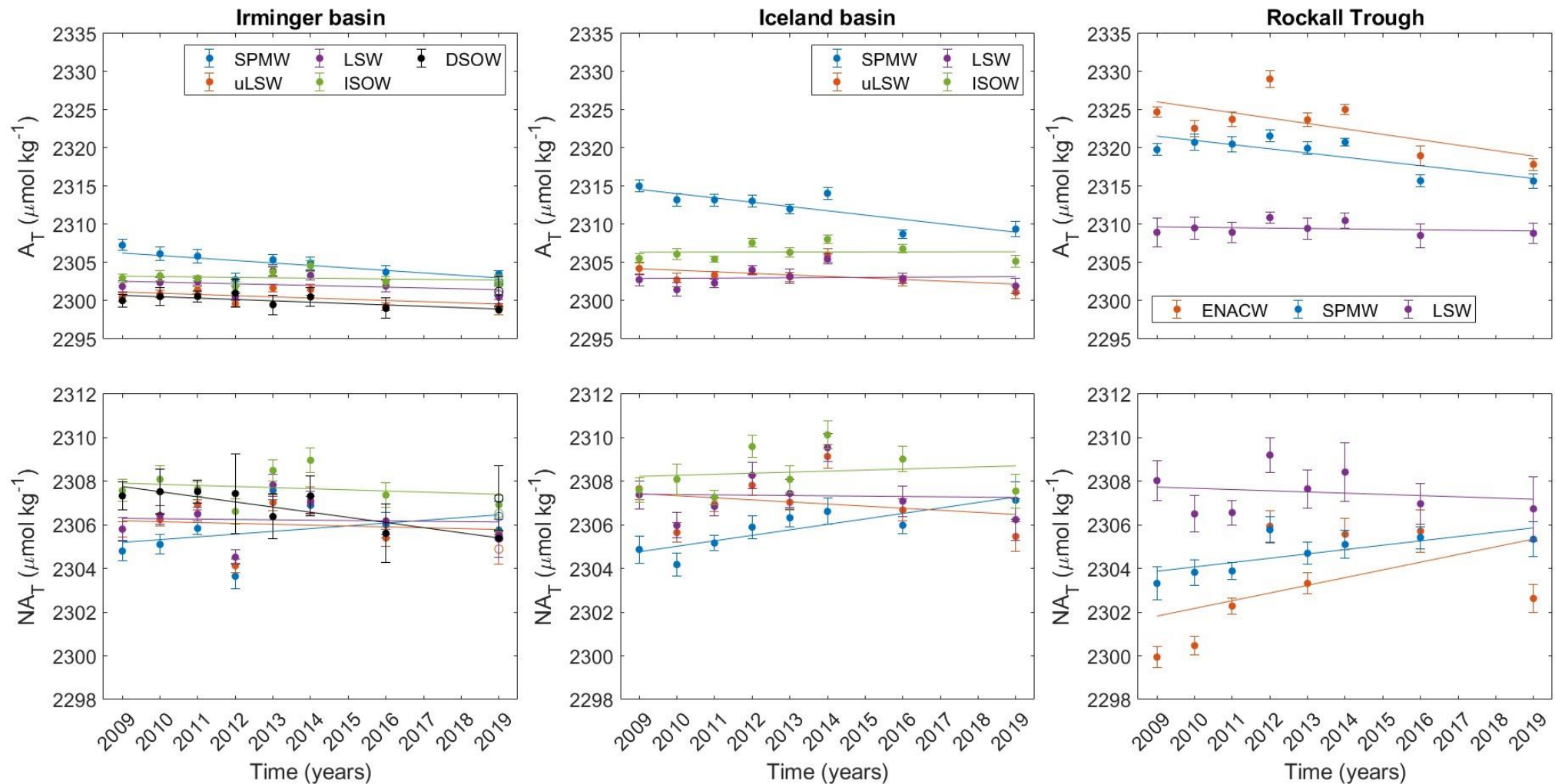
Figure S4. Temporal distribution (2009-2019) of the average A_T and NA_T in each of the layers considered for the Irminger (left plot column), Iceland (central plot column) and Rockall basins (right plot column). The average values were calculated for each cruise and layer and represented with coloured points together with their respective error bars at the time of each cruise (the method used for calculations was described in section 3.2). In the Irminger plots, the empty points represent the average values for 2019 calculated with the measured data available in the easternmost part of the basin (sampled part during this cruise), while the coloured points for 2019 represent the average values corrected with A25-OVIDE-2018 data. The interannual trend were given by linear regression of the average values, with the values of the slope, the standard error of estimate and the r^2 presented in Table S4.
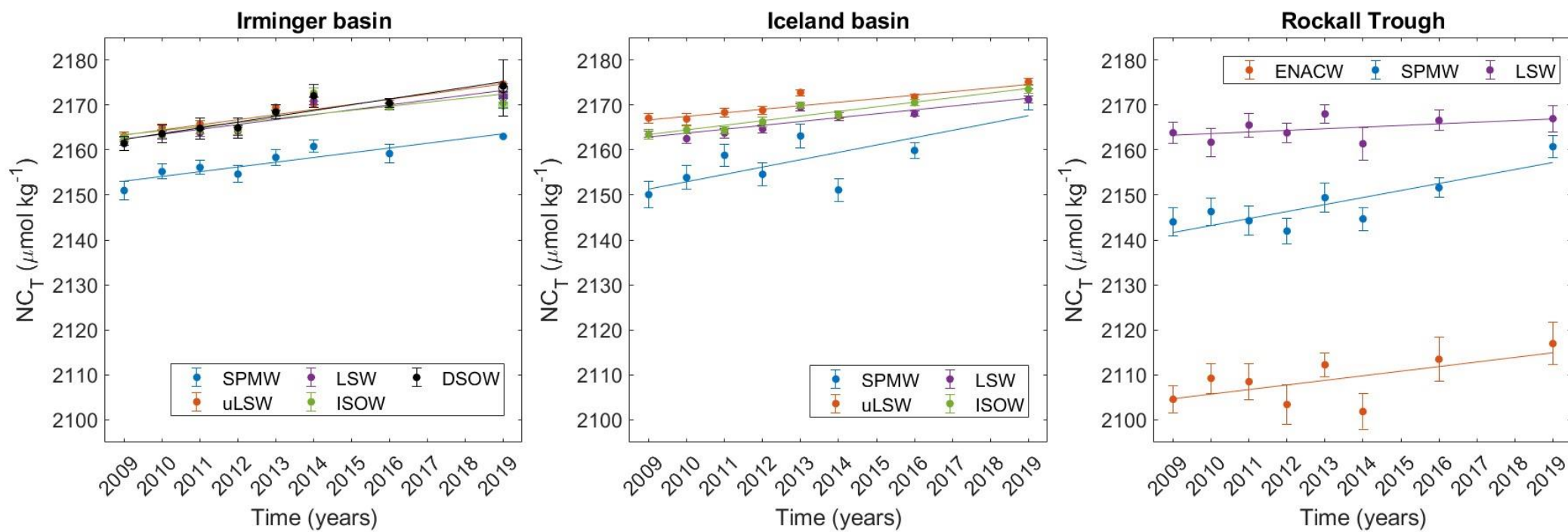
Figure S5. Same as Figure S4 but for NC_T.
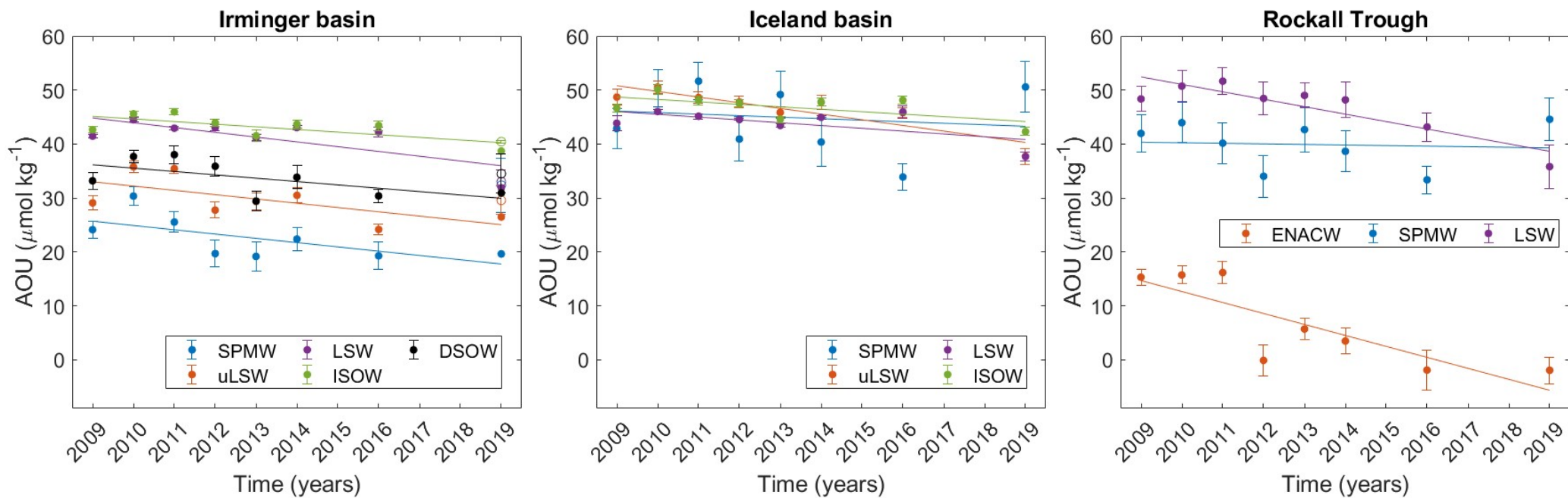
Figure S6. Same as Figure S4 but for AOU.

Table S1. Amount of data (n), mean values and standard deviation (std) per cruise for the measured temperature, salinity, A_T, C_T, pH_T, DO, PO_4 and $Si(OH)_4$.

Cruise	Temperature (°C)			Salinity			A_T (μmol kg^{-1})			C_T (μmol kg^{-1})			pH_T			DO (μmol kg^{-1})			PO_4 (μmol kg^{-1})			$Si(OH)_4$ (μmol kg^{-1})		
	n	mean	std	n	mean	std	n	mean	std	n	mean	std	n	mean	std	n	mean	std	n	mean	std	n	mean	std
2009	1307	6.509	2.966	1307	35.094	0.177	1087	2308.5	12.8	559	2149.3	20.2	975	8.0066	0.0387	1297	266.8	16.7	1319	0.96	0.22	1326	8.83	3.44
2010	1192	6.711	3.233	1192	35.089	0.173	870	2309.7	9.0	763	2151.8	23.5	1071	7.9978	0.0365	1178	261.3	15.7	1220	0.88	0.27	1228	8.42	3.96
2011	1279	5.979	2.770	1279	35.055	0.156	1080	2308.4	9.5	851	2154.1	21.1	1035	7.9994	0.0350	1167	266.2	15.4	1284	0.92	0.23	1284	8.67	3.72
2012	1205	5.867	2.541	1205	35.056	0.159	1189	2308.3	12.5	900	2152.9	20.8	1126	8.0105	0.0438	1136	272.9	17.6	1163	0.91	0.21	1158	8.22	3.49
2013	1235	6.354	2.592	1235	35.057	0.158	1324	2306.8	15.3	1206	2154.4	19.4	1199	8.0077	0.0394	1223	272.3	19.0	1248	0.90	0.21	1279	7.56	3.73
2014	1226	6.476	2.889	1226	35.068	0.157	1059	2307.7	16.1	983	2151.0	25.5	1196	8.0089	0.0454	1211	271.3	16.5	1272	0.90	0.27	1275	6.82	3.67
2016	1849	5.514	2.253	1849	35.001	0.114	1557	2305.6	8.2	1465	2157.9	19.0	1349	7.9959	0.0415	1676	276.1	16.5	1608	0.96	0.23	1642	7.49	2.97
2019	768	6.721	3.146	768	35.042	0.126	808	2307.3	8.6	768	2155.3	28.6	755	7.9796	0.0480	768	271.4	16.2						

Table S2. Average differences between measured and computed variables with CANYON estimations for each of the cruises. The number of data available to apply CANYON-B (n) and the standard deviation (std) were shown.

Cruise	A_T measured - A_T canyon (μmol kg^{-1})			C_T measured - C_T canyon (μmol kg^{-1})			C_T computed - C_T canyon (μmol kg^{-1})			C_T new - C_T canyon (μmol kg^{-1})			pH_T measured - pH_T canyon (units)			PO_4 measured - PO_4 canyon (μmol kg^{-1})			SiO_4 measured - SiO_4 canyon (μmol kg^{-1})		
	n	mean	std	n	mean	std	n	mean	std	n	mean	std	n	mean	std	n	mean	std	n	mean	std
2009	1047	-2.5	2.5	558	0.4	4.0	974	-1.5	4.5	1052	-0.4	4.6	964	-0.0021	0.0097	1271	0.03	0.03	1290	1.08	0.95
2010	940	-3.1	3.0	821	-0.4	4.8	1140	-2.1	3.8	1146	-0.3	4.5	1113	-0.0003	0.0082	1282	-0.04	0.03	1316	0.38	0.92
2011	972	-2.4	2.1	769	0.7	4.264	1011	-2.6	3.6	1027	-0.3	4.5	982	0.0015	0.0082	1138	-0.04	0.04	1162	0.37	0.82
2012	1105	-1.6	3.6	864	2.1	3.932	1106	-4.4	4.4	1109	0.1	5.2	1085	0.0074	0.0087	1088	-0.04	0.03	1114	0.20	0.71
2013	1200	-1.3	2.5	1192	3.9	3.876	1191	-0.2	4.1	1213	3.9	3.9	1164	-0.0016	0.0097	1112	-0.02	0.04	1154	0.32	1.02
2014	968	-0.9	3.1	951	0.7	5.833	1166	-0.3	5.9	1176	1.0	6.2	1151	-0.0009	0.0151	1180	0.00	0.03	1202	-0.53	0.49
2016	1359	-2.2	3.3	1352	2.8	4.183	1334	-0.9	4.9	1344	2.5	4.8	1297	-0.0026	0.0114	1435	0.02	0.04	1514	-0.26	0.85
2019	765	-2.8	3.8	754	5.4	5.213	-	-	-	754	5.4	5.2	-	-	-	-	-	-	-	-	-

Table S3. Amount (n) and percentage of measured and computed C_T data per cruise used to compilate the C_T "new". No computed C_T data were used for the 2019 cruise due to lack of measured pH needed for computation.

Cruise		C_T "new"			
		C_T measured		C_T computed	
	n total				
		n	percentage	n	percentage
2009	1088	501	46.05%	587	53.95%
2010	1202	709	58.98%	493	41.02%
2011	1029	645	62.68%	384	37.32%
2012	1161	735	63.31%	426	36.69%
2013	1307	1095	83.78%	212	16.22%
2014	1285	713	55.49%	572	44.51%
2016	1421	1223	86.07%	198	13.93%
2019	754	754	100%	0	0%

Table S4. Interannual trends of A_T, NA_T, NC_T and AOU in each of the layers and basins. The ratios of change were based on linear regressions applied to the average values (as represented in Supplementary Figures 2-4) and presented together with its Standard error of estimate. The correlation coefficients r^2 and p-values were also provided. Values in bold denotes that trends are statistically significant at the 95% level of confidence.

Basin	Layer	A_T ratio (μmol kg-1 yr-1)		r2	p-value	NA_T ratio (μmol kg^{-1} yr^{-1})		r^2	p-value	NC_T ratio (μmol kg^{-1} yr^{-1})		r^2	p-value	AOU ratio (μmol kg^{-1} yr^{-1})		r^2	p-value
Irminger	SPMW	**-0.33**	± **0.17**	0.50	0.05	0.13	± 0.18	0.12	0.41	**1.05**	± **0.25**	0.82	<0.01	-0.79	± 0.47	0.43	0.08
	uLSW	-0.16	± 0.13	0.27	0.19	-0.04	± 0.16	0.02	0.76	**1.14**	± **0.20**	0.89	<0.01	-0.80	± 0.48	0.42	0.08
	LSW	**-0.11**	± **0.18**	0.09	0.47	-0.02	± 0.15	0.00	0.90	**1.08**	± **0.24**	0.85	<0.01	**-0.89**	± **0.41**	0.55	0.04
	ISOW	-0.05	± 0.13	0.04	0.64	-0.05	± 0.12	0.05	0.61	**0.90**	± **0.33**	0.66	<0.01	**-0.49**	± **0.26**	0.48	0.05
	DSOW	**-0.18**	± **0.09**	0.53	0.04	**-0.24**	± **0.07**	0.75	<0.01	**1.28**	± **0.23**	0.89	<0.01	-0.62	± 0.40	0.38	0.10
Iceland	SPMW	**-0.56**	± **0.19**	0.70	<0.01	**0.25**	± **0.08**	0.72	<0.01	**1.63**	± **0.70**	0.59	0.03	-0.28	± 0.98	0.02	0.73
	uLSW	-0.20	± 0.19	0.22	0.24	-0.10	± 0.18	0.07	0.53	**0.79**	± **0.25**	0.72	<0.01	**-1.05**	± **0.28**	0.79	<0.01
	LSW	0.02	± 0.20	0.00	0.88	-0.02	± 0.18	0.00	0.92	**0.86**	± **0.22**	0.80	<0.01	-0.52	± 0.32	0.40	0.09
	ISOW	0.01	± 0.16	0.00	0.96	0.05	± 0.16	0.02	0.72	**1.03**	± **0.16**	0.91	<0.01	-0.45	± 0.29	0.38	0.10
Rockall	ENACW	-0.71	± 0.41	0.44	0.07	0.35	± 0.32	0.24	0.21	1.02	± 0.63	0.41	0.09	**-2.03**	± **0.69**	0.70	<0.01
	SPMW	-0.55	± 0.22	0.62	0.02	**0.20**	± **0.09**	0.54	0.04	**1.56**	± **0.50**	0.72	<0.01	-0.10	± 0.66	0.01	0.85
	LSW	**-0.05**	± **0.13**	0.05	0.61	-0.06	± 0.15	0.03	0.66	0.36	± 0.33	0.24	0.22	**-1.38**	± **0.37**	0.78	<0.01