Acidification and deoxygenation north-western part of the Japan/East Sea

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Supplementary Information

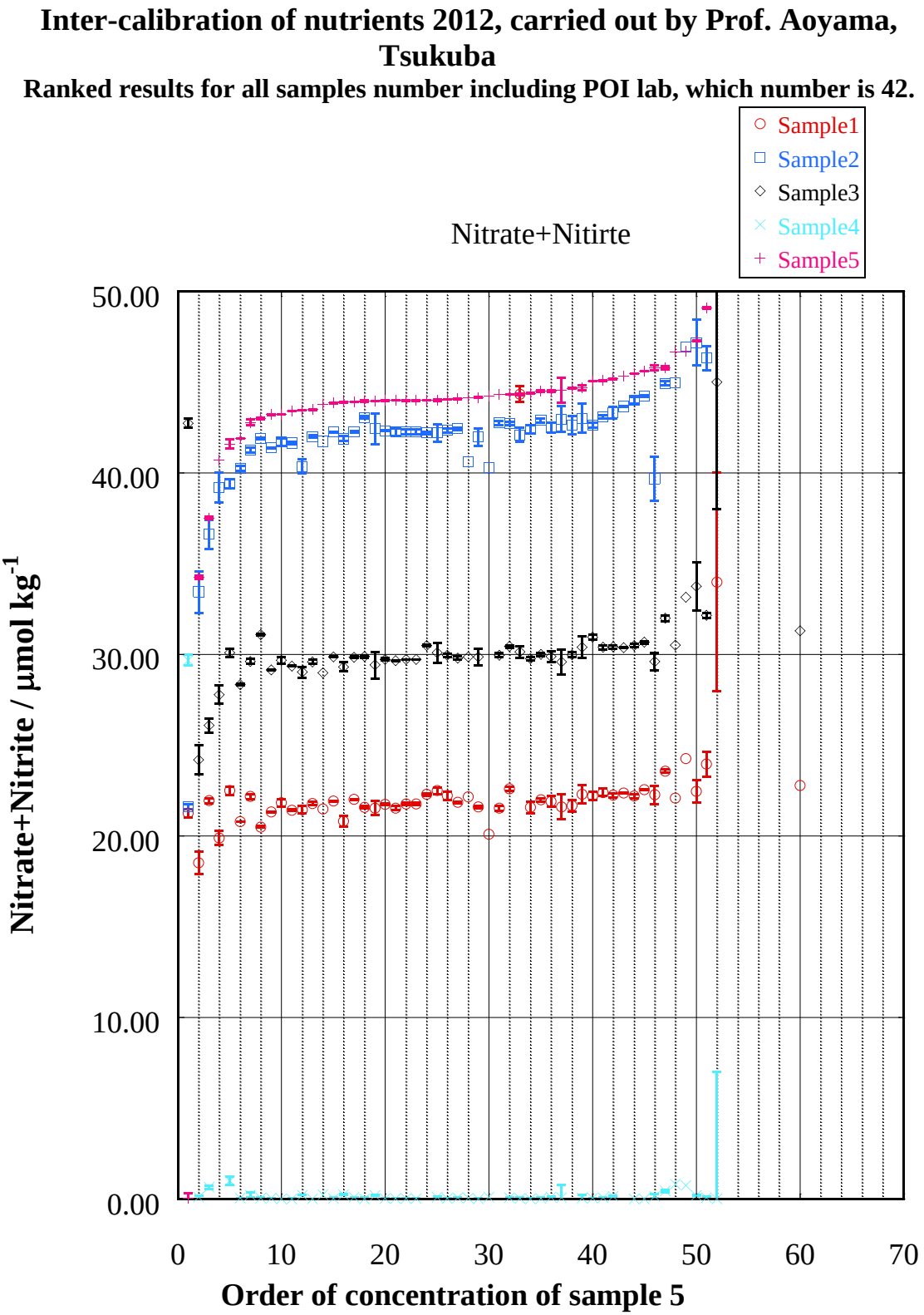

Figure S1 Ranked Nitrite+Nitrate results for all samples: Reported concentrations were sorted using concentration of sample 5

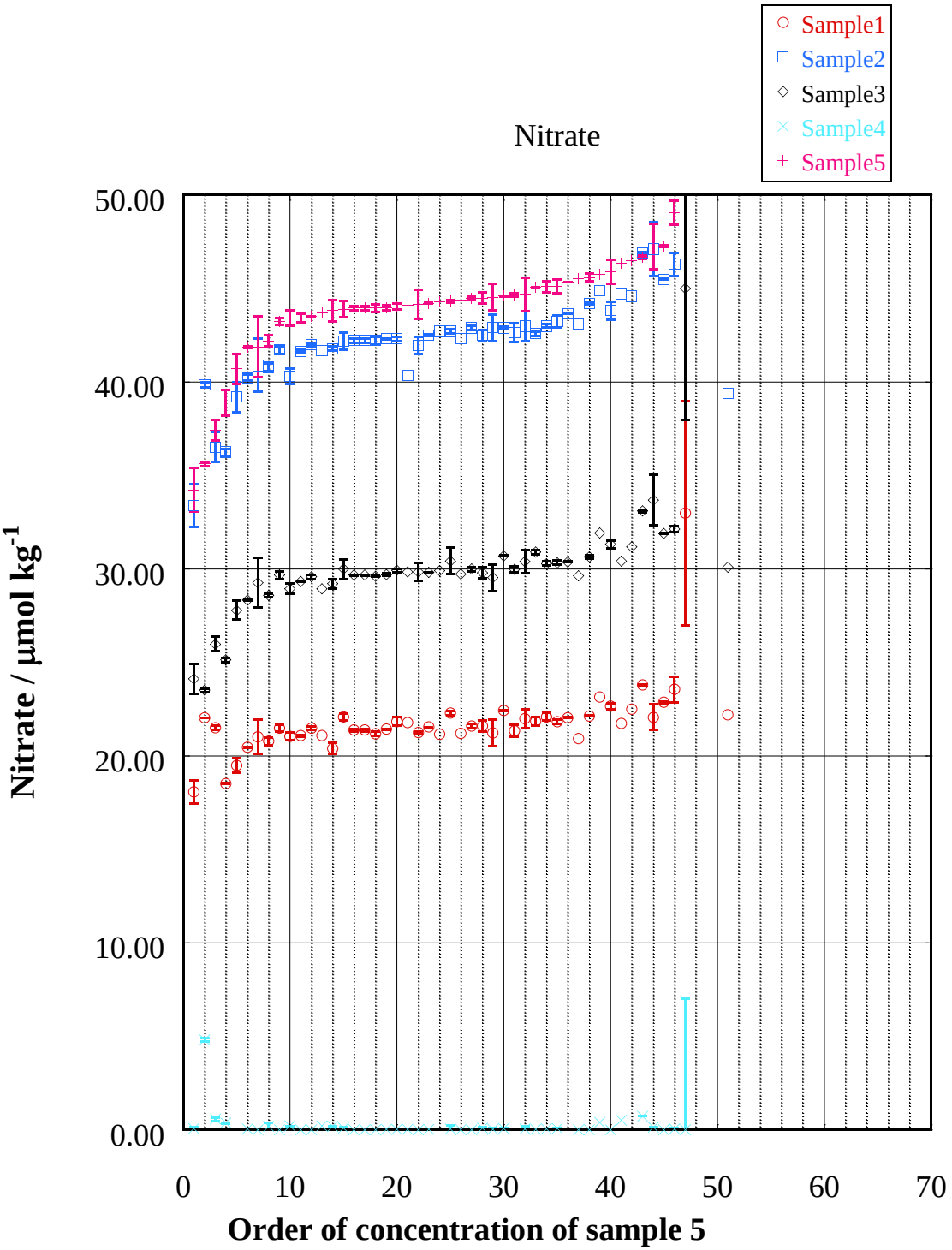

Figure S2 Ranked Nitrate results for all samples: Reported concentrations were sorted using concentration of sample 5

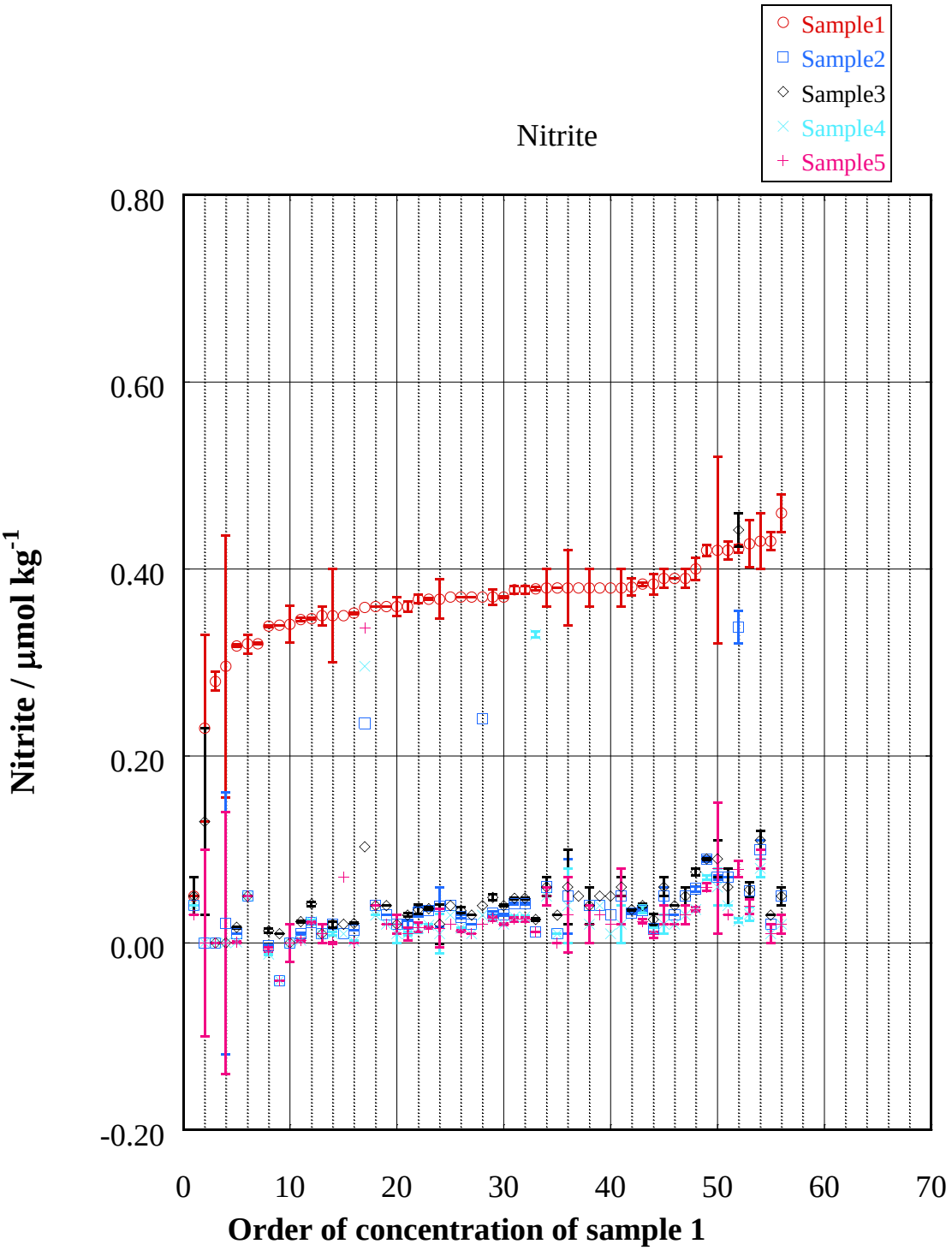

Figure S3 Ranked Nitrite results for all samples: Reported concentrations were sorted using concentration of sample1

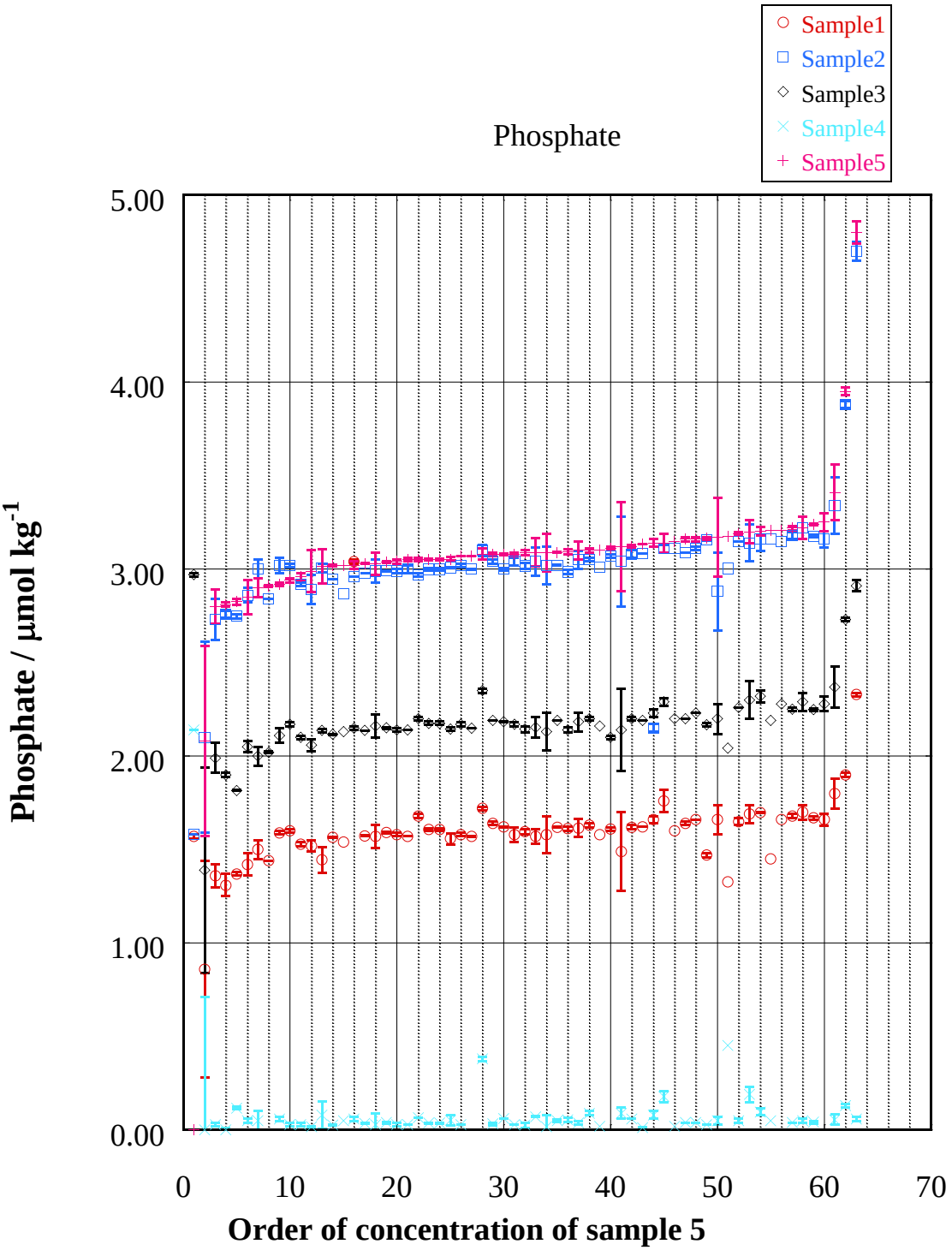

Figure S4 Ranked Phosphate results for all samples: Reported concentrations were sorted using concentration of sample 5

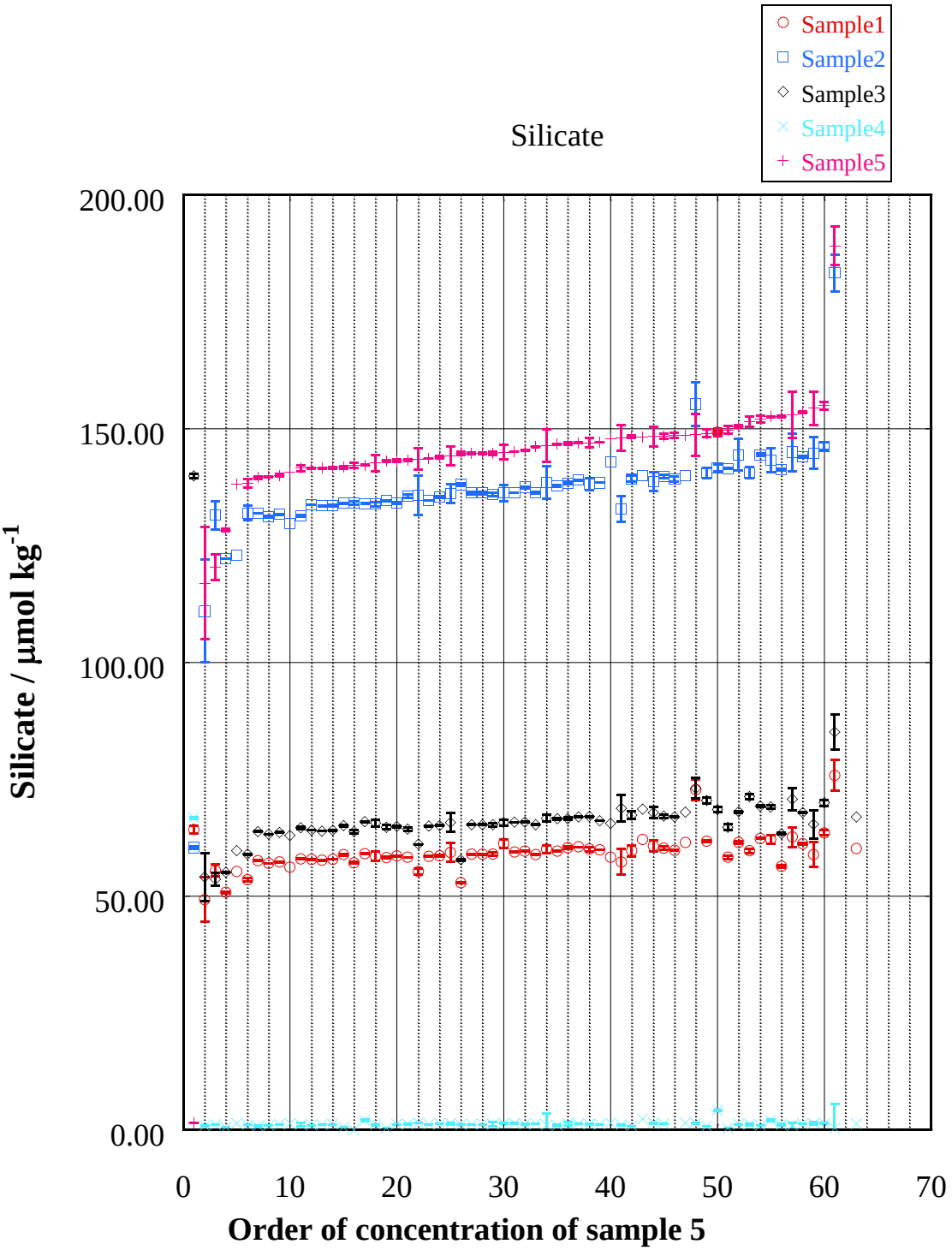

Figure S5 Ranked silicate results for all samples: Reported concentrations were sorted using concentration of sample 5

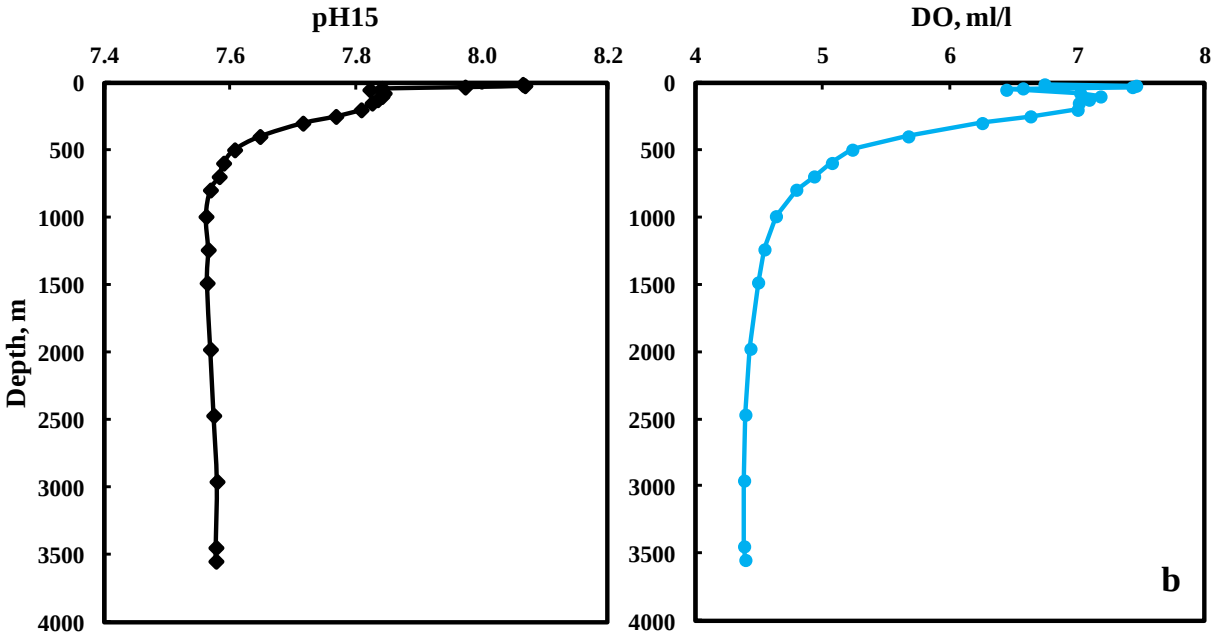

Figure S6. Vertical distribution of $pH_T(15^oC)$ – **a** and dissolved oxygen – **b** in water column. Station CR-02, KH-10-02 cruise, June 2010, Japan/East Sea.

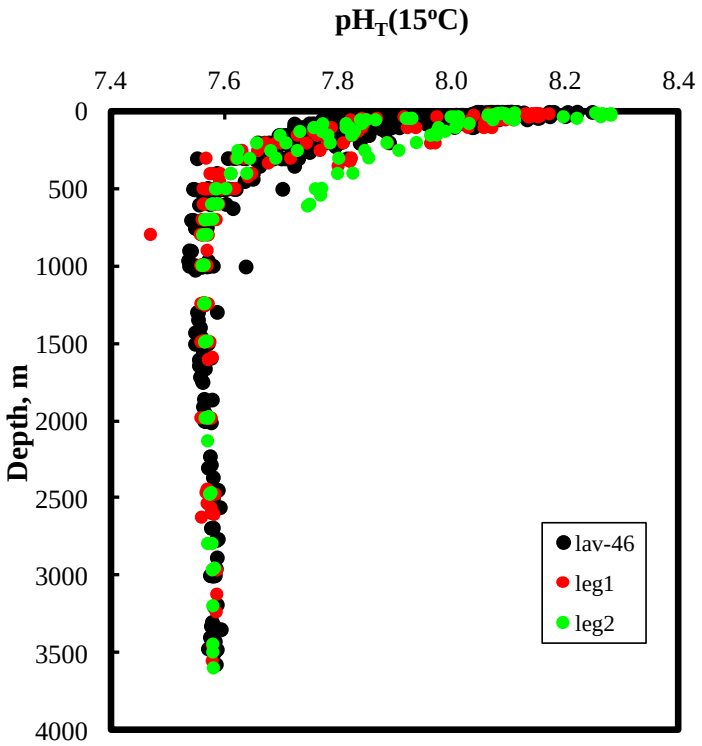

Figure S7. Comparison of pH data measured at 15 ^oC in "total hydrogen concentration scale" for Japan Sea. **Black** is Lavretyev cruise in 2009; **Red** is Hakuho-Maru 2010, leg1; **Green** is Hakuho-Maru 2010, leg 2**.**

Figures S6, S7 suggest that precision and accuracy of pH measurements are very similar and better than 0.01 unit pH. Data demonstrated by Fig. S7 were obtained different peoples in different time, using different equipment. Scatter of data below 2000 m is within 0.01 pH unit.

Table S1. The average hydrochemical parameters ([DO], [P], $[NO_3^-]$, [Si], n- number of measurements, $3s.d.(mean) = 3s.d./\sqrt{n}$, where s.d. – standard deviation) of the JES at nominal depths for different years

Depth, m	year	[DO], μmol/kg	n	3s.d. (mean)	[P], μmol/kg	n	3s.d. (mean)	$[NO_3^-]$, μmol/kg	n	3s.d. (mean)	[Si], μmol/kg	n	3s.d. (mean)
750	1999	226.2	106	4.98	1.86	106	0.039	23.49	106	0.49	53.74	106	1.99
750	2001	234.6	32	6.87	1.80	32	0.044	22.78	32	0.60	50.43	32	1.90
750	2001	245.4	21	7.28	1.70	21	0.056	21.74	21	0.82	47.38	21	4.12
750	2004	230.5	25	8.06	1.82	25	0.042	23.20	25	0.46	53.31	25	2.72
750	2005	228.3	12	8.84	1.80	12	0.075	22.92	4	0.97	50.03	12	3.18
750	2007	221.7	14	7.07	1.87	14	0.053	22.45	14	0.58	54.14	14	3.23
750	2009	222.5	24	4.97	1.88	24	0.041	22.23	24	1.39	54.84	24	3.45
750	2010	216.5	12	7.22	1.91	12	0.050	24.97	12	0.60	55.27	12	2.63
750	2014	210.4	29	3.94	1.97	25	0.035	24.54	29	0.52	54.92	0	2.37
750	2014	213.3	20	8.16	1.92	20	0.068	24.54	20	0.52	54.92	20	2.37
1250	1999	216.3	50	1.58	1.93	50	0.011	24.39	50	0.11	66.89	50	1.91
1250	2001	218.5	30	3.20	1.93	30	0.019	24.26	30	0.19	67.32	30	0.95
1250	2001	220.6	16	3.63	1.87	16	0.025	23.97	16	0.28	64.87	16	3.77
1250	2004	214.1	21	1.85	1.93	20	0.039	24.48	19	0.27	67.56	20	2.05
1250	2005	212.8	11	3.21	1.89	12	0.060	24.27	6	1.06	64.95	12	2.22
1250	2007	211.9	7	1.48	1.94	7	0.013	23.31	7	0.18	69.16	7	2.40
1250	2007	211.3	14	1.31	1.94	14	0.020	23.63	14	0.24	70.12	14	1.30
1250	2009	209.8	18	1.66	1.99	18	0.029	23.50	18	1.30	71.82	18	2.16
1250	2010	203.7	10	1.58	2.00	10	0.013	26.08	10	0.14	69.97	10	0.97
1250	2014	202.9	28	0.57	2.01	24	0.017	24.98	28	0.44	70.08	28	1.14
1250	2014	202.4	15	0.92	2.01	15	0.018	25.20	15	0.39	68.59	15	1.66
1750	1999	210.4	36	0.87	1.97	36	0.008	24.91	36	0.07	76.17	36	1.27
1750	2001	211.3	28	0.56	1.96	28	0.017	24.77	28	0.14	77.05	28	0.84
1750	2001	212.4	16	1.62	1.93	16	0.017	24.43	15	0.20	75.30	16	1.95

1750	2004	210.6	18	1.46	1.98	16	0.026	24.72	15	0.28	76.19	16	1.78
1750	2005	209.4	9	1.85	1.92	10	0.057	24.41	5	0.70	73.92	10	1.58
1750	2007	208.7	7	1.19	1.97	7	0.010	23.66	7	0.15	78.28	7	2.34
1750	2009	207.6	16	0.62	1.98	16	0.016	24.17	16	1.11	80.77	16	0.87
1750	2010	202.2	8	1.12	2.02	8	0.008	26.39	8	0.18	78.31	8	1.37
1750	2014	202.6	26	0.40	2.01	22	0.017	25.13	25	0.43	77.56	25	0.67
1750	2014	202.3	15	0.94	2.02	15	0.017	25.42	15	0.28	76.22	15	0.90
2250	1999	209.7	36	0.77	1.99	36	0.005	25.11	36	0.04	81.36	36	0.85
2250	2001	210.6	24	0.52	1.99	24	0.021	25.15	24	0.15	83.01	24	1.14
2250	2001	213.4	18	2.74	1.94	18	0.031	24.41	18	0.29	78.58	18	2.52
2250	2004	209.5	17	1.71	2.00	16	0.001	24.83	16	0.25	81.45	16	1.66
2250	2005	209.2	8	0.78	1.94	8	0.061	24.73	5	0.61	78.55	8	1.64
2250	2007	208.5	7	0.45	1.99	7	0.013	23.85	7	0.18	83.09	7	1.09
2250	2009	207.5	15	1.02	2.04	15	0.014	24.76	15	0.94	85.86	15	0.79
2250	2010	202.7	6	0.54	2.03	6	0.011	26.51	6	0.17	82.48	6	0.73
2250	2014	203.1	19	0.61	2.03	15	0.013	25.52	19	0.40	82.40	19	0.60
2250	2014	202.6	11	0.97	2.04	11	0.017	25.32	11	0.52	80.74	11	1.29
3000	1999	211.1	51	0.38	1.99	51	0.003	25.11	51	0.03	82.70	51	0.24
3000	2001	212.9	32	1.03	1.99	33	0.021	25.15	33	0.15	83.01	33	1.14
3000	2001	214.2	32	1.42	1.94	32	0.019	24.70	32	0.22	82.23	32	1.20
3000	2004	211.0	29	0.35	2.00	30	0.012	24.85	29	0.20	83.26	30	0.56
3000	2005	210.3	14	0.56	1.96	13	0.014	24.68	7	0.53	80.39	13	0.78
3000	2007	209.0	11	0.50	1.98	11	0.009	23.82	11	0.16	85.33	11	0.28
3000	2007	209.1	14	0.95	1.97	14	0.013	24.64	14	0.95	85.59	14	3.09
3000	2009	208.4	19	0.94	2.03	18	0.013	24.74	19	1.04	87.77	19	0.65
3000	2010	203.4	11	0.36	2.04	11	0.007	26.65	11	0.12	85.09	11	0.52
3000	2014	205.1	22	0.68	2.05	17	0.022	25.67	21	0.41	84.66	21	0.68
3000	2014	204.4	14	0.85	2.05	14	0.012	25.54	14	0.33	85.81	14	1.11

Table S2. The average carbonate system parameters ([NTA], [pH], [NDIC], pCO_2, n- number of measurements, $3s.d.(mean) = 3s.d./\sqrt{n}$, where s.d. – standard deviation) of the JES at nominal depths for different years

Depth, m	year	NTA, μmol/kg	n	3s.d. (mean)	pH	n	3s.d. (mean)	NDIC, μmol/kg	n	3s.d. (mean)	pCO_2, μatm	n	3s.d. (mean)
750	1999	2333.3	56	1.3	7.762	56	0.015	2292.4	56	5.5	711.4	56	24.3
750	2001	2333.0	30	2.0	7.788	30	0.020	2284.5	30	6.0	668.9	30	30.2
750	2001	2333.4	21	2.7	7.815	21	0.018	2276.4	21	6.5	626.2	21	25.7
750	2004	2332.3	25	2.7	7.782	25	0.018	2285.2	25	7.3	677.0	25	32.2
750	2005	2331.6	12	2.1	7.767	12	0.009	2289.3	12	4.6	702.2	12	15.5
750	2007	2329.8	14	4.5	7.760	14	0.016	2289.6	14	7.4	713.7	14	26.4
750	2009	2331.6	24	1.5	7.750	24	0.009	2294.1	24	3.6	730.8	24	15.0
750	2010	2332.1	12	2.4	7.740	12	0.012	2297.7	12	4.2	749.7	12	20.6
750	2014	2333.0	29	1.9	7.733	29	0.007	2300.4	29	3.0	761.4	29	12.6
750	2014	2333.0	20	2.9	7.741	20	0.017	2298.0	20	5.5	748.8	20	28.8
1250	1999	2338.0	27	2.5	7.725	27	0.008	2303.4	27	3.0	732.1	27	13.7
1250	2001	2339.4	30	1.9	7.741	30	0.005	2299.5	30	2.4	705.9	30	8.3
1250	2001	2338.3	16	4.2	7.751	16	0.013	2295.1	16	6.8	688.7	16	20.7
1250	2004	2339.1	21	2.4	7.736	21	0.004	2300.3	21	3.2	713.7	21	7.7
1250	2005	2339.6	12	1.8	7.729	12	0.003	2303.2	12	2.7	727.1	12	5.9
1250	2007	2335.9	8	9.1	7.730	8	0.004	2299.3	8	9.4	724.1	8	8.0
1250	2009	2338.6	19	2.0	7.717	19	0.004	2305.4	19	2.8	747.1	19	7.1
1250	2010	2338.4	10	2.6	7.706	10	0.005	2308.3	10	2.1	768.2	10	8.2
1250	2014	2339.7	29	2.3	7.714	29	0.005	2307.4	29	2.9	754.2	29	8.7
1250	2014	2339.6	16	2.9	7.712	15	0.008	2307.7	15	2.4	756.7	15	14.1
1750	1999	2344.6	22	1.7	7.701	22	0.005	2309.9	22	2.2	732.6	22	9.1
1750	2001	2345.2	28	2.2	7.716	28	0.002	2306.6	28	2.3	708.5	28	3.4
1750	2001	2344.8	16	5.3	7.721	16	0.004	2304.6	16	5.7	700.3	16	7.7
1750	2004	2345.8	21	2.6	7.715	21	0.003	2307.4	21	3.4	710.7	21	5.7
1750	2005	2345.5	10	1.4	7.710	10	0.002	2308.6	10	1.8	720.3	10	3.3

1750	2007	2344.3	7	9.4	7.713	7	0.003	2307.0	7	9.2	713.9	7	6.0
1750	2009	2344.7	18	1.6	7.701	18	0.002	2310.3	18	1.6	734.8	18	4.2
1750	2010	2345.2	8	2.8	7.693	8	0.005	2312.9	8	1.6	749.2	8	9.0
1750	2014	2344.0	26	2.6	7.702	26	0.006	2309.2	26	3.5	732.4	26	10.3
1750	2014	2343.8	15	2.9	7.698	15	0.007	2310.1	15	2.9	739.8	15	12.5
2250	1999	2348.5	19	1.5	7.683	19	0.006	2312.9	19	2.3	723.6	19	10.1
2250	2001	2349.1	23	2.1	7.700	23	0.002	2308.8	23	2.1	697.1	23	4.5
2250	2001	2346.3	18	5.4	7.705	18	0.004	2304.8	18	5.8	687.5	18	6.5
2250	2004	2349.8	17	3.2	7.699	17	0.004	2310.0	17	4.1	699.1	17	7.0
2250	2005	2347.7	8	1.2	7.694	8	0.003	2309.4	8	1.5	707.2	8	5.9
2250	2007	2351.5	7	3.7	7.698	7	0.003	2312.5	7	4.2	700.8	7	6.0
2250	2009	2348.6	16	0.9	7.687	16	0.001	2312.3	16	0.8	719.0	16	2.3
2250	2010	2348.0	6	1.8	7.675	6	0.003	2314.9	6	2.2	740.0	6	5.6
2250	2014	2347.6	19	2.1	7.690	19	0.007	2310.3	19	3.3	714.0	19	12.3
2250	2014	2347.7	11	4.2	7.685	11	0.010	2311.9	11	3.8	722.8	11	16.3
3000	1999	2349.3	14	1.6	7.659	14	0.004	2311.3	14	1.6	703.7	14	7.1
3000	2001	2349.2	28	1.5	7.683	28	0.005	2304.7	28	2.7	666.1	28	8.9
3000	2001	2350.3	32	3.5	7.681	32	0.004	2306.3	32	3.9	669.5	32	6.4
3000	2004	2348.7	30	2.1	7.673	30	0.001	2307.1	30	2.1	681.9	30	2.7
3000	2005	2348.8	14	1.1	7.667	14	0.002	2308.9	14	1.3	692.6	14	4.3
3000	2007	2352.2	11	2.8	7.673	11	0.003	2310.9	11	3.3	681.5	11	4.5
3000	2009	2349.5	21	0.5	7.666	21	0.003	2310.1	21	1.3	694.6	21	5.5
3000	2010	2350.0	11	1.0	7.648	11	0.005	2314.7	11	1.4	725.6	11	7.9
3000	2014	2347.9	21	2.1	7.666	21	0.007	2308.1	21	2.9	693.6	21	12.7
3000	2014	2349.7	14	3.3	7.666	14	0.008	2309.9	14	3.1	693.9	14	12.9

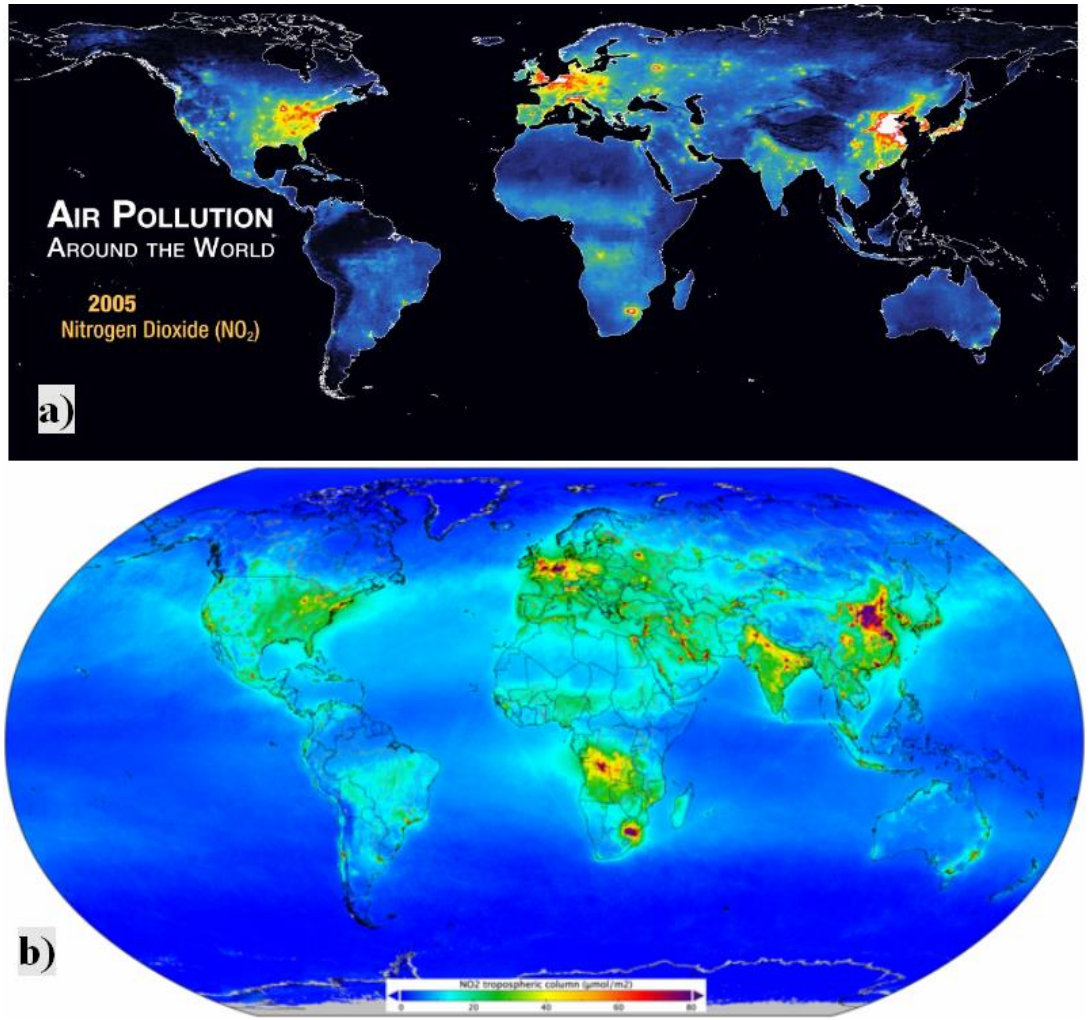

Figure S8. Nitrogen dioxide worldwide pollution. **a)** NASA's Earth-observing satellite, Aura, measures the air pollutant nitrogen dioxide (NO2) from high above Earth's surface. (https://eospso.gsfc.nasa.gov/sites/default/files/publications/NO2GlobalLenticular_508.pdf);

b) Measurements gathered by the Copernicus Sentinel-5P mission between April and September 2018 have been averaged to reveal nitrogen dioxide in the atmosphere, ESA data. (http://www.esa.int/spaceinimages/Images/2019/03/Nitrogen_dioxide_worldwide).