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Ocean alkalinity enhancement in an open-ocean ecosystem: biogeochemical responses and carbon storage durability

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Table S1: Overview on linear regression analyses for all variables included in the summary table (Table 1) in the main manuscript. Phase 0 refers to before alkalinity addition, Phase I corresponds to Days 7-19 (short-term response), and Phase II corresponds to Days 21-33 (longer-term response). Bold data indicate significant positive impact of OAE detected, and italics indicates significant negative impact of OAE was detected.

Group of variables	Variable	Phase	F-Statistic (df = 7)	p value	Multiple R^2	Estimate (intercept)	Estimate (x-coefficient)	Data transformation
nutrients	Nitrate + nitrite [NO_x^-]	0	0.975	0.3563		-0.9211	0.0001	none
		I	0.204	0.6655	0.028	-0.0667	0.0000	none
		II	2.10	0.1275	0.299	-0.0331	0.0000	log(y + 1)
	Inorganic phosphate [PO_4^{3-}]	0	1.54	0.2548		0.1383	0.0000	none
		I	0.123	0.7357	0.017	0.0454	0.0000	log(y + 1)
		II	3.56	0.1012	0.337	0.0438	0.0000	none
	Dissolved silicate [$Si(OH)_4^-$]	0	5.337	0.0537		0.4067	0.0000	log(y + 1)
		I	1.64	0.2416	0.189	0.2233	0.0000	log(y + 1)
		II	0.096	0.7662	0.057	0.2626	0.0000	log(y + 1)
POM	Chl a	0	0.375	0.5594		2.4030	0.0000	log(y)
		I	1.58	0.2487	0.184	13.6721	-0.0003	$^2\sqrt{(y+1)}$
		II	0.269	0.6199	0.037	7.1817	0.0005	$^3\sqrt{(y+1)}$
	POC	0	0.295	0.6040		89.7057	-0.0024	none
		I	4.82	0.0642	0.408	76.1748	-0.0042	none
		II	0.009	0.9262	0.001	4.3100	0.0000	$^3\sqrt{(y+1)}$
	PON	0	1.12	0.3100		1.1570	0.0000	none
		I	*11.3*	*0.0121*	*0.617*	*11.8449*	*-0.0009*	*none*
		II	0.099	0.7620	0.014	1.1460	0.0000	log(y)
	POP	0	3.33	0.1109	0.322	0.0000	0.0000	y^4
		I	3.24	0.1151	0.316	0.0000	0.0000	y^4
		II	0.041	0.8452	0.006	0.0027	0.0000	y^2
POM stoichiometry	POC:PON	0	0.514	0.4965		6.2026	0.0002	none
		I	3.52	0.1027	0.335	6.3983		none
		II	**62.1**	**0.0001**	**0.899**	**5.5810**	**0.0005**	**none**
	POC:POP	0	1.685	0.2354	0.194	160.5000	0.0053	none
		I	0.717	0.4252	0.093	161.5000	0.0026	none
		II	0.434	0.5308	0.058	134.3000	0.0065	none
	PON:POP	0	0.829	0.4136	0.097	19.8492	0.0011	none
		I	0.022	0.7535	0.015	21.7250	-0.0002	none
		II	0.030	0.8201	0.008	20.2446	-0.0003	none
	PON:BSi	0	1.91	0.2090		0.0191	0.0000	$1/y^2$
		I	0.024	0.8819	0.003	78.5541	-0.0059	y^2
		II	1.45	0.2670	0.172	16.8356	0.0038	none
Biomineralisation	BSi	0	0.345	0.5754		0.1694	0.0000	none
		I	0.457	0.5208	0.061	0.5058	0.0000	none
		II	0.828	0.3927	0.106	0.4056	0.0000	$^3\sqrt{(y)}$
	PIC	0	0.490	0.5063		1.9852	0.0002	$^3\sqrt{(y)}$
		I	0.107	0.7530	0.015	1.7730	0.0000	$^3\sqrt{(y+5)}$
		II	0.135	0.7287	0.000	0.6581	0.0001	$^2\sqrt{(y)}$
	PIC:POC	0	0.465	0.5173		0.0897	0.0000	log(y + 1)
		I	**7.32**	**0.0304**	**0.511**	**0.0012**	**0.0001**	**none**
		II	1.13	0.3237	0.139	0.6655	0.0000	$^4\sqrt{(y)}$
	POC:BSi	0	0.982	0.3547		0.0000	0.0000	$1/y^3$
		I	0.009	0.9267	0.001	65.9248	-0.0007	none
		II	1.997	0.2005	0.222	115.4953	0.0353	none
	δ^{15}N-PON	0	4.54	0.0707	0.393	20.6250	-0.0043	y^2
		I	0.580	0.4712	0.077	41760.69	-8.9000	$10^{(y+1)}$
		II	0.416	0.5395	0.056	80.2658	-0.0183	y^3

Table S2: Overview of carbonate system variables as mean ± S.D. for each OAE treatment level for the experiment phases. "Short-term" refers to Days 7-19 (short-term response), and "longer term" refers to Days 21-33 with "Pre-treatment" corresponding to days prior to alkalinity addition.

		OAE								
		0	300	600	900	1200	1500	1800	2100	2400
Phase	Variable									
Pre-treatment	TA ($\mu mol\ kg^{-1}$)	2393.1 ± 6.5	2395.0 ± 3.5	2398.3 ± 3.1	2398.1 ± 1.2	2399.3 ± 2.1	2399.4 ± 2.6	2397.4 ± 3.8	2400.3 ± 3.5	2398.7 ± 1.4
	DIC ($\mu mol\ kg^{-1}$)	2110.0 ± 10.2	2107.9 ± 5.7	2121.2 ± 9.9	2114.5 ± 5.1	2117.4 ± 7.1	2115.3 ± 5.1	2117.7 ± 7.8	2113.1 ± 7.3	2115.0 ± 4.2
	pCO_2 (μatm)	448 ± 11	441 ± 11	464 ± 18	449 ± 13	453 ± 15	449 ± 12	458 ± 14	443 ± 21	449 ± 7
	Ω_{Ar}	3.17 ± 0.06	3.21 ± 0.08	3.11 ± 0.12	3.18 ± 0.09	3.16 ± 0.10	3.18 ± 0.09	3.14 ± 0.09	3.21 ± 0.17	3.18 ± 0.04
Short term	TA ($\mu mol\ kg^{-1}$)	2415.0 ± 14.2	2693.4 ± 9.5	2988.2 ± 12.5	3282.7 ± 9.2	3587.7 ± 13.3	3865.8 ± 18.5	4139.9 ± 17.2	4439.2 ± 17.8	4707.3 ± 15.7
	DIC ($\mu mol\ kg^{-1}$)	2114.4 ± 11.7	2344.4 ± 12.7	2588.1 ± 17.1	2830.6 ± 20.3	3076.4 ± 20.3	3296.5 ± 29.6	3508.1 ± 31-0	3743.6 ± 25.2	3951.3 ± 27.3
	pCO_2 (μatm)	421 ± 11	426 ± 16	435 ± 17	445 ± 23	448 ± 18	449 ± 27	444 ± 24	446 ± 15	447 ± 17
	Ω_{Ar}	3.35 ± 0.07	4.00 ± 0.09	4.71 ± 0.11	5.44 ± 0.16	6.27 ± 0.14	7.08 ± 0.23	7.96 ± 0.22	8.88 ± 0.14	9.74 ± 0.15
Longer term	TA ($\mu mol\ kg^{-1}$)	2440.3 ± 5.6	2699.8 ± 55.2	3017.3 ± 33.8	3310.3 ± 10.7	3618.0 ± 30.7	3895.6 ± 35.6	4174.0 ± 42.9	4469.3 ± 29.5	4615.8 ± 92.2
	DIC ($\mu mol\ kg^{-1}$)	2124.1 ± 12.5	2352.0 ± 12.3	2598.3 ± 19.1	2829.1 ± 16.2	3082.4 ± 19.0	3294.9 ± 24.0	3502.9 ± 25.8	3754.0 ± 30.1	3892.2 ± 52.2
	pCO_2 (μatm)	412 ± 8	443 ± 86	419 ± 29	415 ± 29	428 ± 19	422 ± 35	417 ± 32	436 ± 30	464 ± 40
	Ω_{Ar}	3.47 ± 0.05	3.98 ± 0.50	4.91 ± 0.28	5.76 ± 0.26	6.54 ± 0.24	7.44 ± 0.43	8.37 ± 0.46	9.10 ± 0.36	9.28 ± 0.67

Table S3: Overview of experiment information for unpublished data sets included in Fig. 8. For KOSMOS2016 and KOSMOS2017, pH was calculated on the total scale from measured TA and DIC using CO2SYS as described in the methods. NO_x was determined spectrophotometrically and Chlorophyll a fluorometrically.

Experiment name	Location	Experiment description	Mesocosms/time period selected
KOSMOS2014 (published)	Gran Canaria (Gando Bay)	Ocean acidification with simulated upwelling of deep water. See Taucher et al. (2018) for more information	All mesocosms on all sampling days before deep water addition (Day 27). $[NO_x] < 0.20$ µM
KOSMOS2016 (unpublished)	Gran Canaria (Taliarte Harbour)	Ocean acidification with simulated upwelling of deep water.	All mesocosms on all sampling days before deep water addition (Day 19). $[NO_x] < 0.20$ µM
KOSMOS2017 (unpublished)	Gran Canaria (Taliarte Harbour)	Ocean artificial upwelling simulated upwelling of deep water.	Only controls mesocosms (M1, M6 - no nutrient addition) on all sampling days (Days 1-29) included. $[NO_x] < 0.12$ µM
KOSMOS2021 (this study)	Gran Canaria (Taliarte Harbour)	Ocean Alkalinity Enhancement	All mesocosms on all sampling days included. $[NO_x] < 0.10$ µM

Fig. S1: Photo showing two deployed mesocosms with the lids open at the pier in Taliarte Harbour, Gran Canaria, Spain (A) and an underwater photo showing the deployed mesocosms and the attached sediment trap (B).

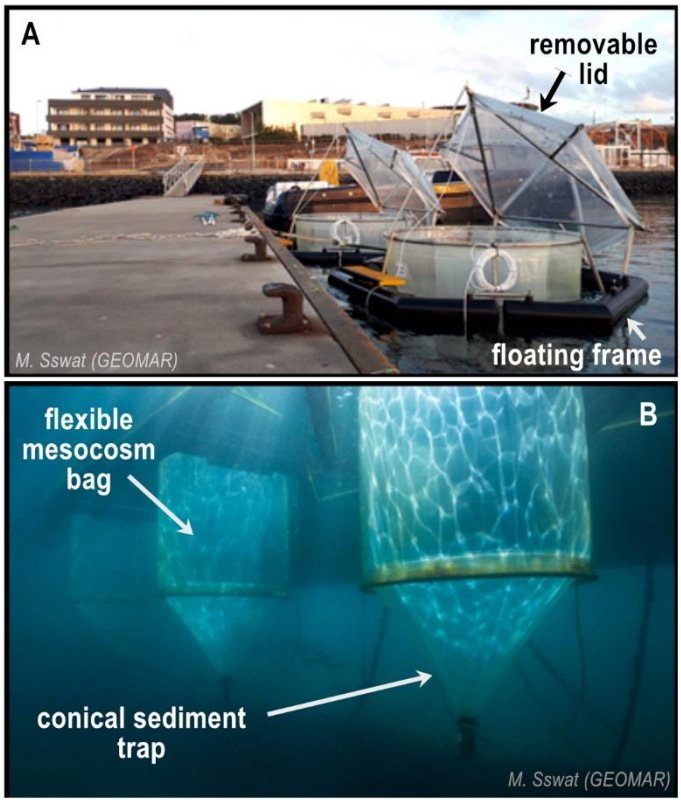

Fig. S2: Photo of white particles attached to mesocosm walls in OAE2400 on Day 28.

Fig. S3: Analysis of two mesocosm wall precipitates (samples A/B) by FTIR (Bruker Vertex 70 FTIR, University of Hamburg) against a pure anhydrous aragonite reference material, and a calcite reference. Overlap of the sample peaks at with the aragonite standard at 854 cm^{-1}, 1083 cm^{-1} and 1424-1474 cm^{-1} confirm aragonite as the primary crystal polymorph in the mesocosm particles (Chakrabarty and Mahapatra, 1999; Jovanovski et al., 2002). Peaks around 3500 cm^{-1} are water (H_2O).

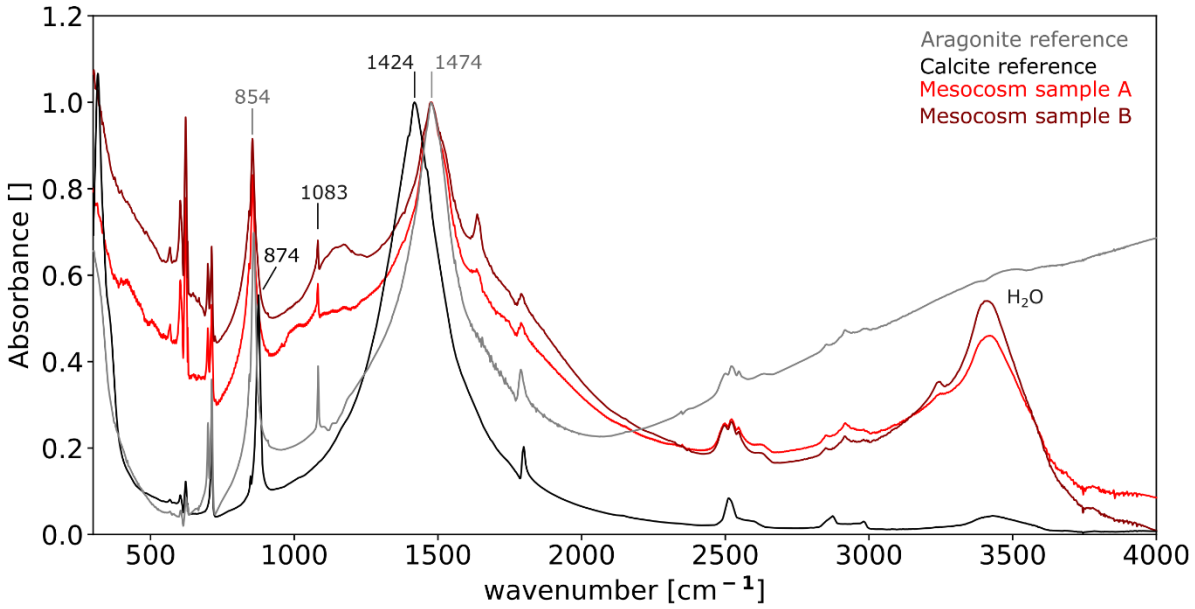

Fig. S4: Measured change in alkalinity (TA) and aragonite saturation state (Ω_{ar}) for up to 150 days after the end of the incubation experiment. Dashed lines refer “fresh” seawater with solid lines for mesocosm water. Error bars indicate the standard deviation of treatment duplicates. Sampling was continued in Gran Canaria until Day 45. Thereafter the bottles were sealed and transported by container back to Hamburg where sampling and analysis was continued on Day 81, 125, 195. These incubations were not temperature controlled so there is some uncertainty in the pH correction applied (Badocco et al., 2021) that was used to calculate saturation states for aragonite (Ω_{ar}). Hence, the Ω_{ar} values have a higher uncertainty associated with them but the general trends over time are considered representative of changes over time.