Supplementary Material

Supplementary Tables

Table S1 | Carbonate Chemistry Parameters of Day 7 in all OAE levels, calculated by CO2SYS (Pierrot et al., 2011) corrected for water-column averaged temperature and salinity. Carbonate dissociation constants (K1 and K2) from Lueker et al (2000) were chosen. pH is on total scale and calculated from hydrogen ion concentrations in mol kg^{-1}.

OAE	TA ($\mu mol\ kg^{-1}$)	DIC ($\mu mol\ kg^{-1}$)	pH_T	pCO_2 (μatm)	HCO_3^- ($\mu mol\ kg^{-1}$)	CO_3^{2-} ($\mu mol\ kg^{-1}$)	R-Factor	Ω_{Ca}	Ω_{Ar}
0	2402.7	2096.5	8.053	404.5	1865.8	218.79	9.74	5.19	3.41
300	2690.5	2327.9	8.098	400.5	2049.5	266.60	9.56	6.32	4.15
600	2968.1	2558.7	8.123	412.6	2237.8	308.70	9.53	7.32	4.81
900	3272.0	2795.1	8.165	404.2	2415.9	367.27	9.34	8.71	5.72
1200	3576.2	3051.1	8.180	424.4	2625.6	412.97	9.37	9.79	6.43
1500	3837.9	3253.5	8.209	420.2	2775.1	465.96	9.26	11.05	7.26
1800	4107.4	3463.0	8.233	419.6	2930.2	520.42	9.17	12.34	8.10
2100	4417.5	3699.4	8.261	416.0	3099.6	587.47	9.07	13.93	9.15
2400	4689.3	3911.5	8.279	419.6	3256.1	643.00	9.020	15.24	10.01

Table S 2| Output of two-tailed Welch´s t-test for unequal variances in sample population. Differing total alkalinity in ΔTA_{2400} was tested for in two phases (Phase I: Day 4 – 17, Phase II: Day 19 – 33)

Response variable	sample estimates		t	df	p-value
	Phase I	Phase II			
Total Alkalinity	4701.07	4610.54	2.565	7.823	**0.0340**

Table S3| Output of fit parameters and p-values levels from linear mixed effects models of numerous parameters fitted to the predictor variables ΔTA and Day with Mesocosm assigned as a random effect. Associated data transformation is shown if assumptions of normally distributed data were violated. Note, that all models excluded the highest treatment level ΔTA_{2400}.

Response variable	Fixed effect	df_{Num}	df_{Den}	F-ratio	p-Value	$R^2_{marginal}$
a) POC Flux	ΔTA	1	6	0.0576	0.8184	
(sqrt transformed)	Day	14	82	4.5633	**< 0.0001**	0.604
	ΔTA × Day	14	82	0.2514	0.9970	
b) POC PON Ratio	ΔTA	1	6	0.6817	0.4406	
(not transformed)	Day	14	82	1.6459	0.0841	0.359
	ΔTA × Day	14	82	0.4666	0.9444	
c) mean sinking velocity	ΔTA	1	6	1.1549	0.3238	
25 – 100 µm	Day	14	81	2.2554	**0.0120**	0.476
(log-10 transformation)	ΔTA × Day	14	81	0.9939	0.4674	
d) mean sinking velocity	ΔTA	1	6	4.3185	0.0830	
100 – 250 µm	Day	14	81	1.9048	**0.0377**	0.461
(log-10 transformation)	ΔTA × Day	14	81	0.5583	0.8892	
e) mean sinking velocity	ΔTA	1	6	1.4978	0.2669	
250 – 1000 µm	Day	14	76	4.8663	**< 0.0001**	0.612
(log-10 transformation)	ΔTA × Day	14	76	1.0107	0.4520	
f) remineralization rates	ΔTA	1	6	0.0014	0.9710	
(not transformed)	Day	6	36	1.2418	0.3085	0.255
	ΔTA × Day	6	36	0.9193	0.4926	
g) PIC Flux	ΔTA	1	6	6.8286	**0.0400**	
(sqrt transformed)	Day	14	82	2.4045	**0.0072**	0.569
	ΔTA × Day	14	82	1.1647	0.3178	
h) PIC POC Ratio	ΔTA	1	6	7.3777	**0.0348**	
(sqrt transformed)	Day	14	82	1.9930	**0.0282**	0.615
	ΔTA × Day	14	82	0.7323	0.7362	
i) BSi Flux	ΔTA	1	6	0.00224	0.9638	
(sqrt transformed)	Day	14	82	8.13492	**<0.0001**	0.688
	ΔTA × Day	14	82	0.25376	0.9969	
j) POP Flux	ΔTA	1	6	0.0126	0.9142	
(log-10 transformation)	Day	14	82	2.8311	**0.0017**	0.441
	ΔTA × Day	14	82	0.167	0.9997	
k) PON Flux	ΔTA	1	6	0.13688	0.7241	
(sqrt transformation)	Day	14	82	397.312	**<.0001**	0.547
	ΔTA × Day	14	82	0.20413	0.9991	
l) BSi:POC Ratio	ΔTA	1	6	0,0607	0.8136	
(log-10 transformation)	Day	14	82	1,9865	**0.0288**	0.359
	ΔTA × Day	14	82	1,4083	0.1681	
m) SV_{avg}	ΔTA	1	6	1.1297	0.3287	
(log-10 transformation)	Day	14	82	3.299	**0.0004**	0.508
	ΔTA × Day	14	82	1.0908	0.3784	

Table S4| Fit parameters and correlation coefficients for the linear regressions shown in Figures 1, 2, 3, 4. Time periods of respective regressions are given in brackets. Note, that all regressions excluded the highest treatment level ΔTA_{2400}.

Dataset	Intercept	Slope	R^2	p-value
POC_{ST} Flux vs. OAE (Day 4 – 33)	0.428	0.00001	0.02	0.721
POC_{ST}:PON_{ST} Ratio vs. OAE (Day 4 – 33)	9.80	0.0002	0.11	0.426
C_{remin} vs. OAE (Day 4 – 33)	0.127	-0.000001	0.02	0.738
PIC_{ST} Flux vs. OAE (Day 19 – 31)	0.019	0.00002	0.55	**0.036**
PIC_{ST}:POC_{ST} Ratio vs. OAE (Day 19 – 31)	0.075	0.00005	0.61	**0.021**
PIC_{ST}:POC_{ST} Ratio vs. $SV_{25\text{-}100\mu m}$ (Day 19 – 31)	13.4	18.1	0.46	0.065
PIC_{ST}:POC_{ST} Ratio vs. $SV_{100\text{-}250\mu m}$ (Day 19 – 31)	20.03	30.1	0.53	**0.039**
PIC_{ST}:POC_{ST} Ratio vs. $SV_{250\text{-}1000\mu m}$ (Day 19 – 31)	48.6	25.1	0.04	0.639
POP_{ST} Flux vs. OAE (Day 4 – 33)	1014.0	8548.4	0.0004	0.960
BSi_{ST} Flux vs. OAE (Day 4 – 33)	976.5	3848.6	0.0004	0.961
POC_{WC} vs. OAE (Day 4 – 33)	684.8	55.4	0.005	0.857
Chl a vs. OAE (Day 4 – 33)	562.9	1453.1	0.20	0.265
PON_{ST} vs. OAE (Day 4 – 33)	1807	-18473	0.04142	0.629
BSi_{ST}:POC_{ST} vs. OAE (Day 4 – 33)	-128,8	26152,5	0,105	0,433
$SV_{avg.}$ vs. OAE (Day 4 – 33)	1017,758	2,993	0,07491	0.984

Supplementary Figures

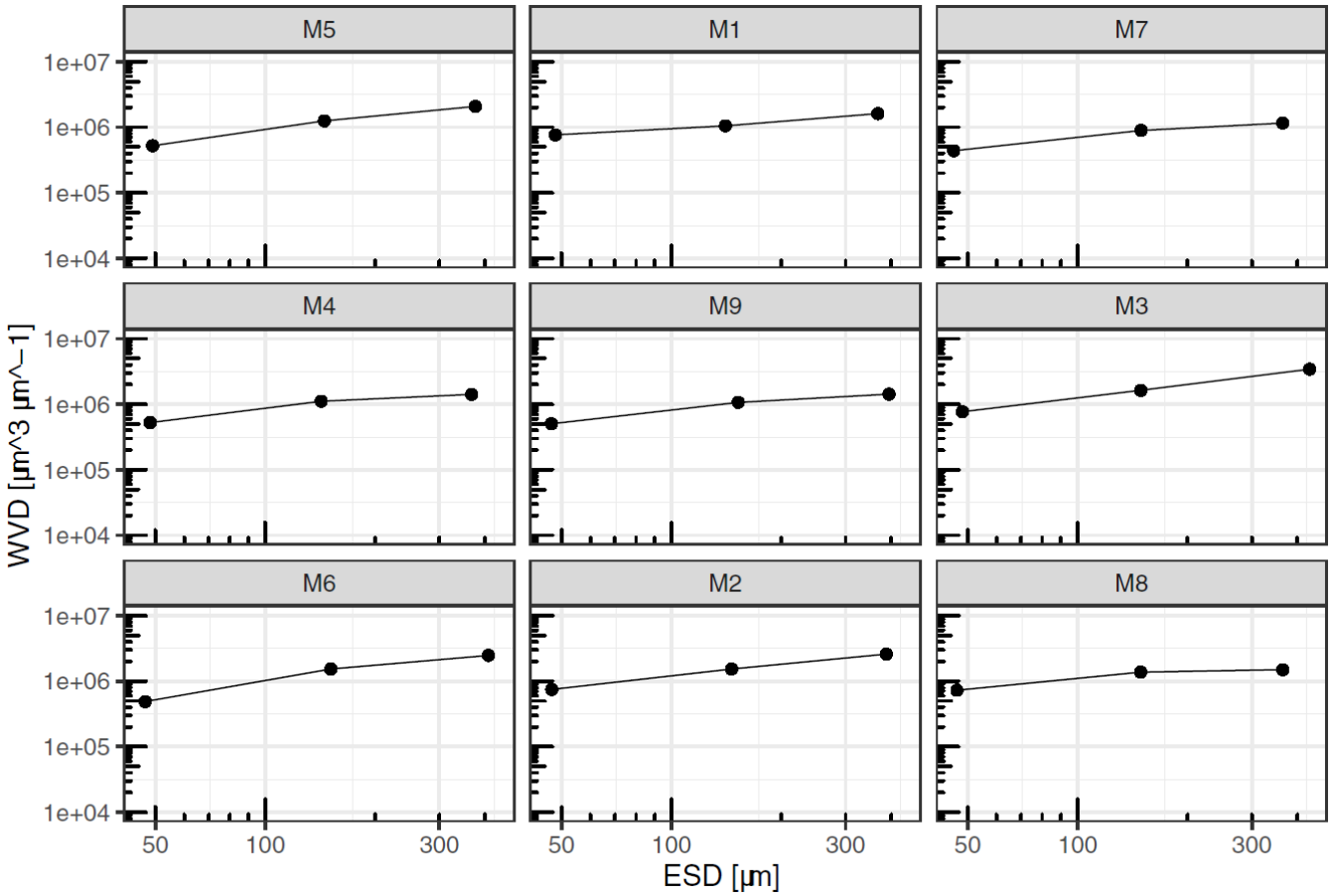

Figure S1 | Exemplary weighted bio-volume distribution (WVD) of sediment trap particles of all mesocosm on Day 9. The cumulative biovolume of all particles within a given size class is plotted against the respective size class mean equivalent spherical diameter (ESD)

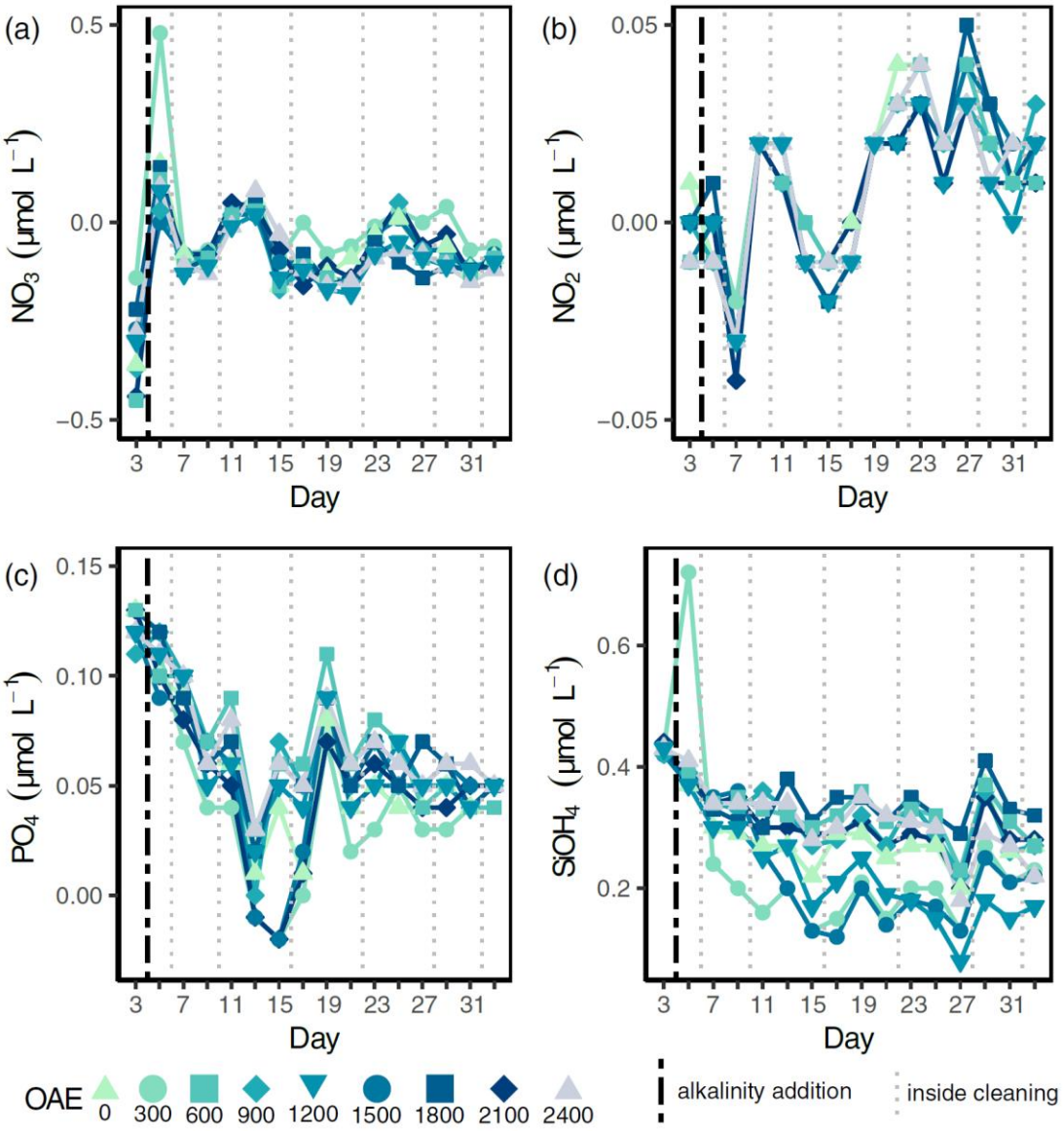

Figure S2| Dissolved Inorganic Nutrients. Concentration of inorganic nitrate (a), nitrite (b) phosphate (C) and silicate (d) over time. Note the negative values for nitrate, nitrite and phosphate, indicating that measured concentrations were low and below the detection limit of the spectrophotometrically determination method. The vertical black line indicates the alkalinity addition and the grey lines the inside cleaning of the mesocosm walls.

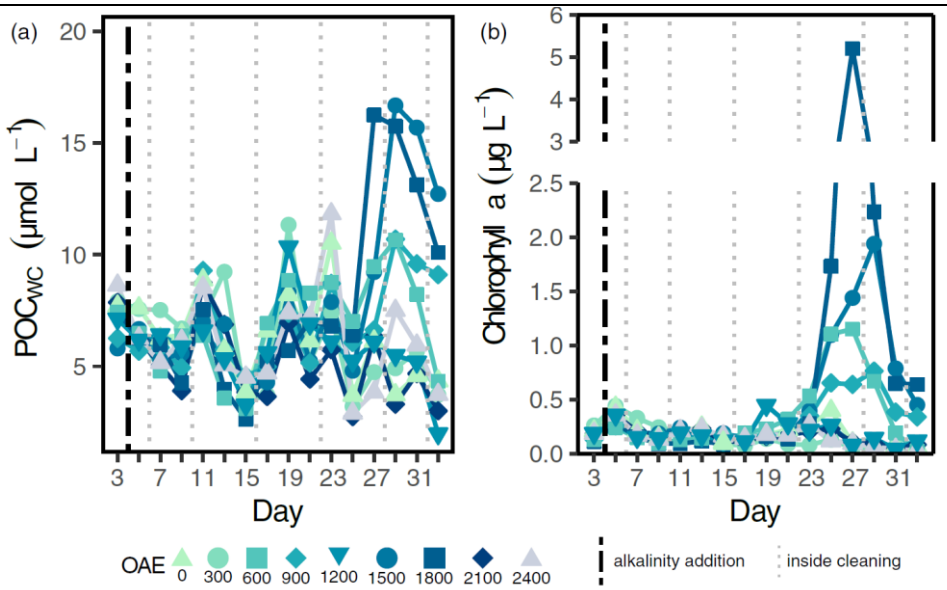

Figure S3| Biotic responses of the water column under the application of OAE. Development of carbon biomass (a) and chlorophyll *a* (b) of the plankton community over time. The vertical black line indicates the alkalinity treatment addition, the grey lines specify days of the inside cleaning of the mesocosm walls.

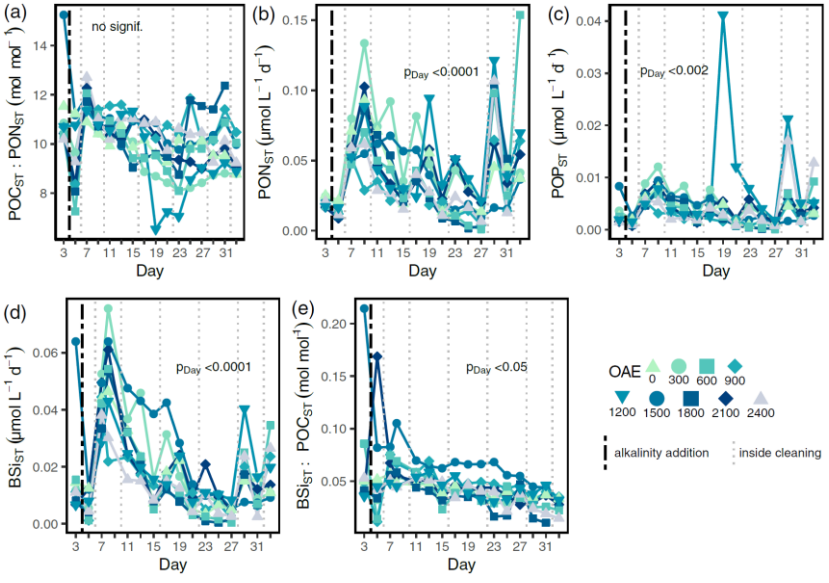

Figure S4| Temporal trends of organic matter fluxes and stoichiometry under the application of OAE. Major elemental fluxes (PON_{ST}, POP_{ST}, BSi_{ST}) in response to OAE over time (b,c,d). Quality and opal ballasting of the export flux (POC_{ST}:PON_{ST}, BSi_{ST}:POC_{ST}) in response to OAE over time (a, e). p-value in (a) represents the output of linear mixed effects model (Table S3b,i,j,k,l). The vertical black line indicates the alkalinity addition and the grey lines the inside cleaning of the mesocosm walls

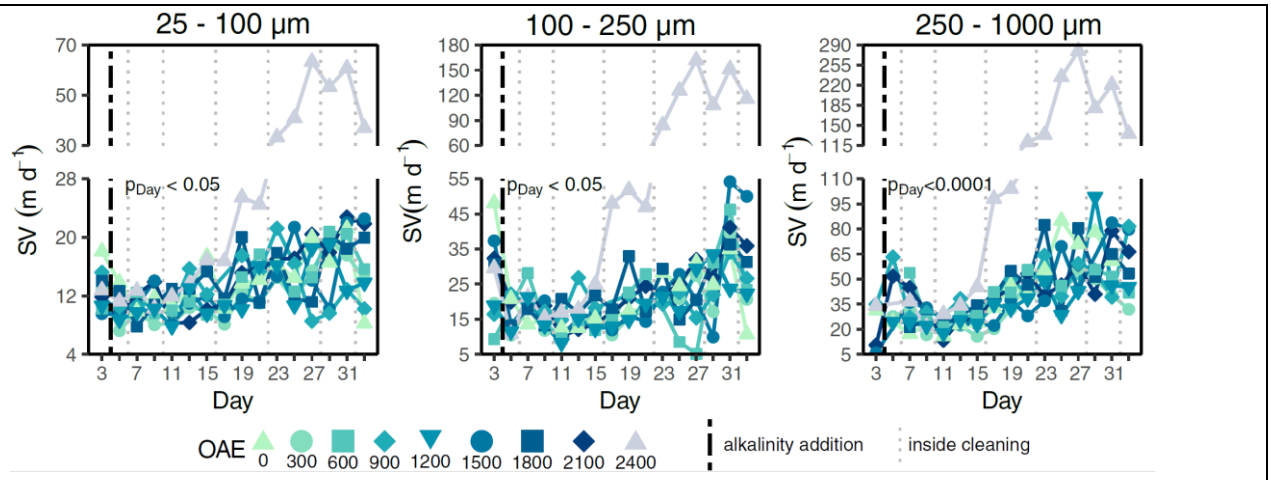

Figure S5| Particle sinking velocities in response to OAE. Development of average particle sinking velocities of different size classes over the application period of OAE. p-value in (a) represents the output of linear mixed effects model (Table S3c,d,e). The vertical black line indicates the alkalinity addition and the grey lines the inside cleaning of the mesocosm walls.

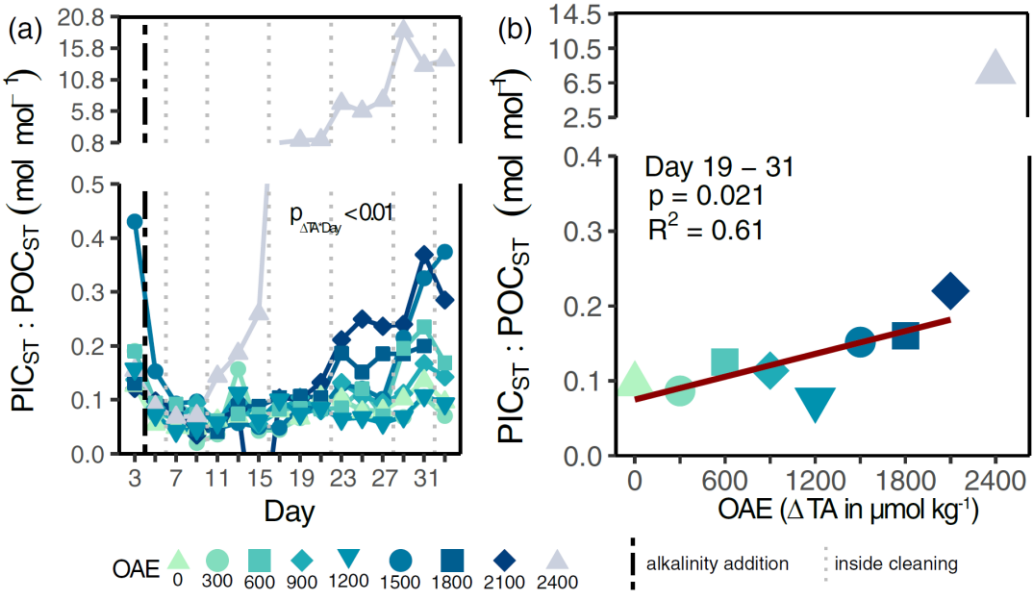

Figure S6| Response of carbonate ballasting to OAE. Carbonate ballasting and its response to OAE over time (a) and averaged over the treatment period (b). p-value in (a) represents the output of linear mixed effects model (Table S3h). The vertical black line indicates the alkalinity addition and the grey lines the inside cleaning of the mesocosm walls.

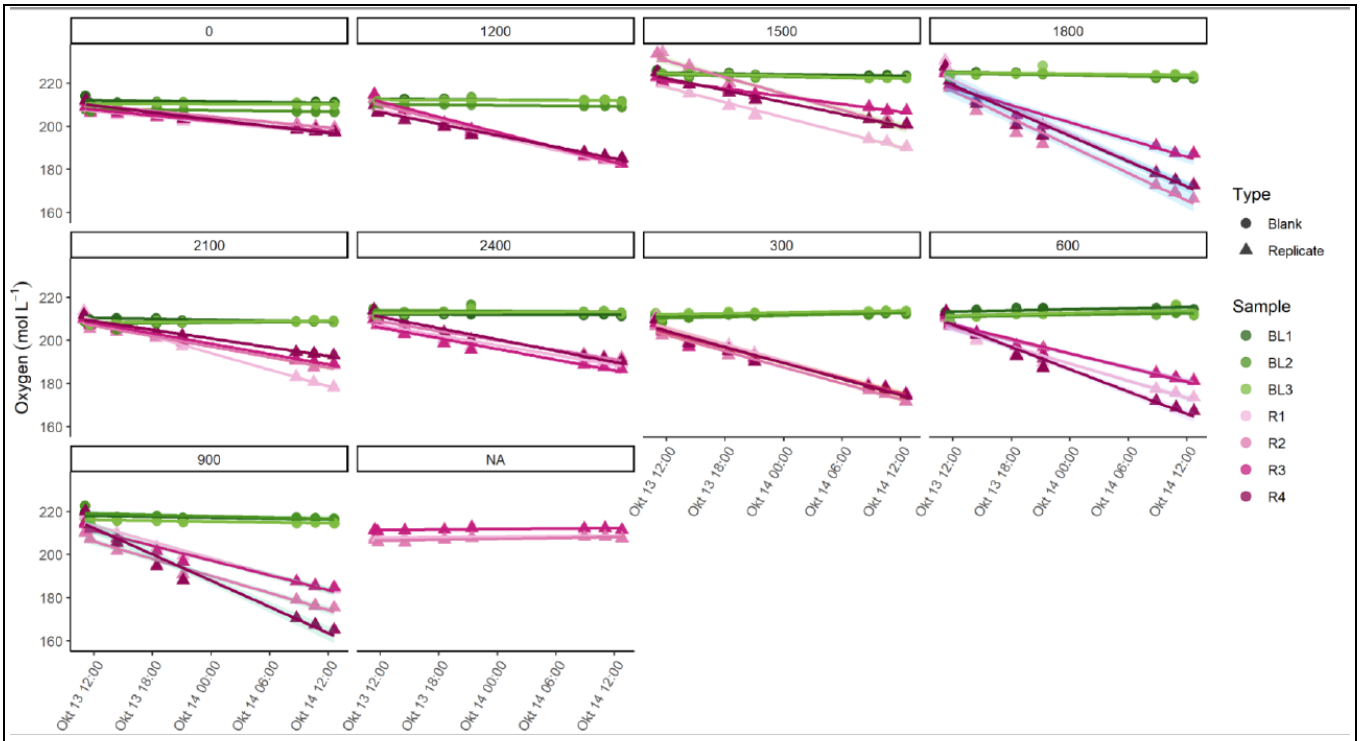

Figure S7| Example of O_2 consumption rates on Day 33. O_2 consumption rates within incubation bottles on the plankton wheel measured with the Fibox 4 on Day 33. Pink dots and regressions represent sediment incubated bottles, while green dots and regressions represent the blank bottle O_2 consumptions. Single data points represent averages of n > 3 measurements and have been corrected for temperature and salinity. Note, that there is no discernible difference in regression lines of ΔTA_{2400} compared to other treatment levels.

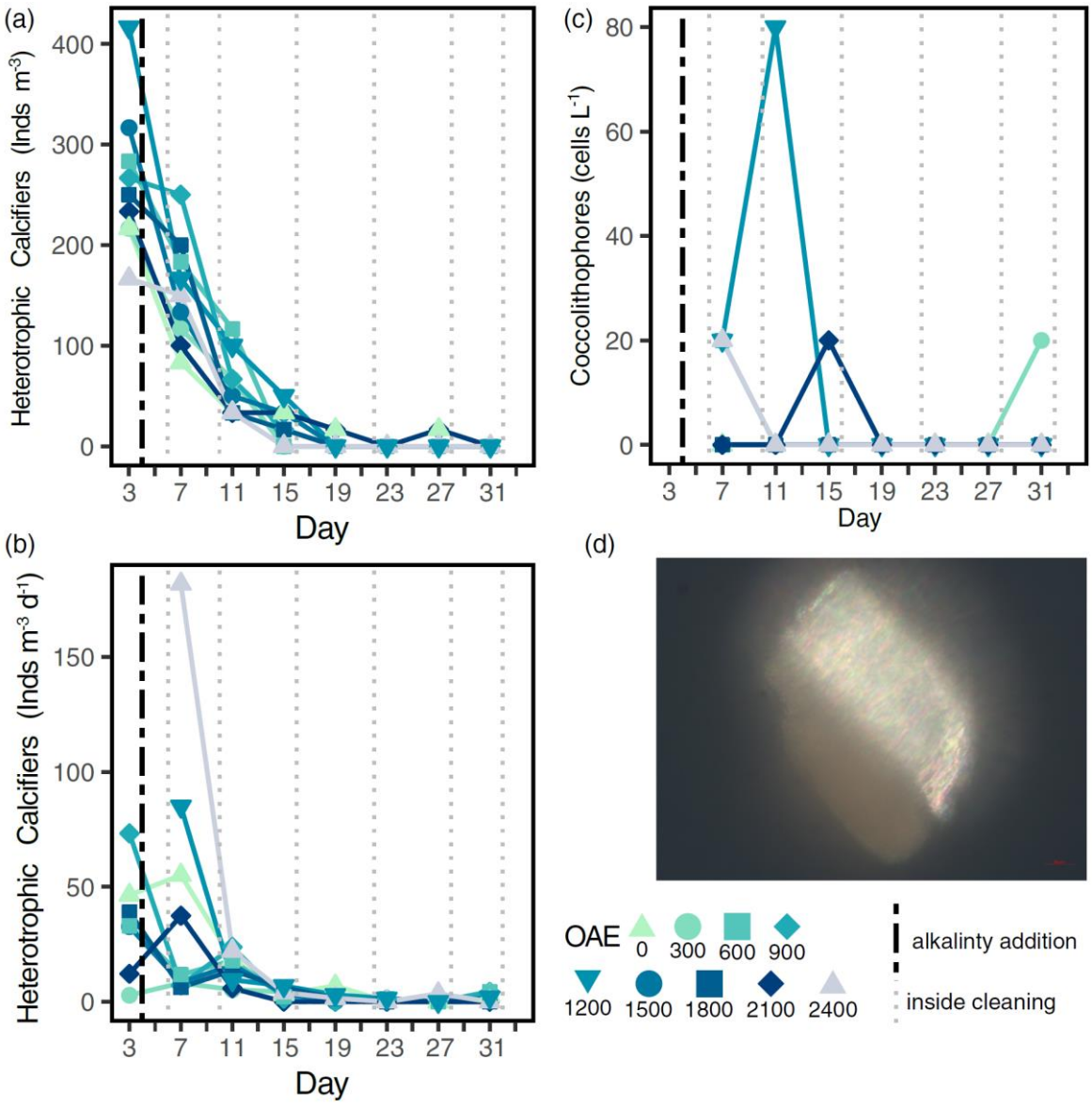

Figure S8| Absence of auto- and heterotrophic calcifiers and presence of white precipitates under OAE. Abundance of heterotrophic calcifiers (taxa: *Foraminifera, Pteropoda*) found in the water column (a) and flux of heterotrophic calcifiers to the sediment trap (b) as well as coccolithophores in the water column (c) over time. Particle of white precipitate sample from ΔTA_{2400} as seen under an inverted microscope with a make-shift polarisation filter in front of the light source (d). The vertical black line indicates the alkalinity addition and the grey lines the inside cleaning of the mesocosm walls.