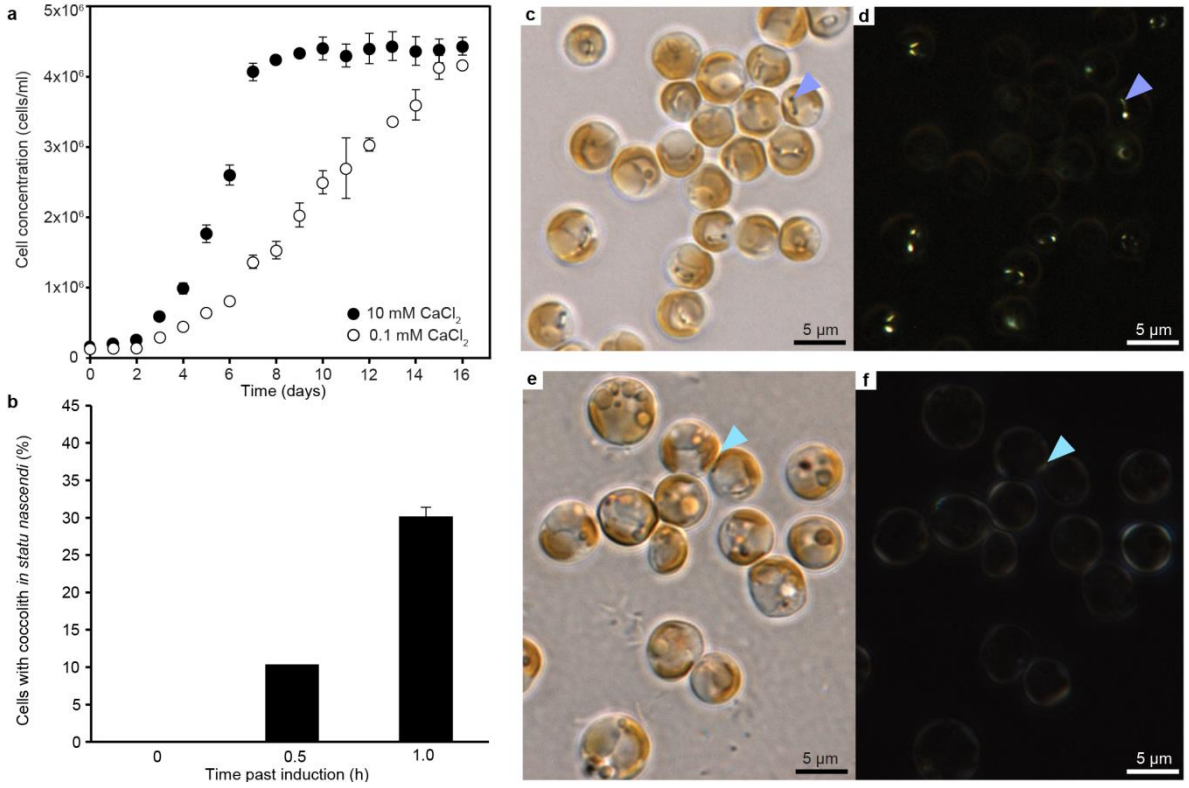
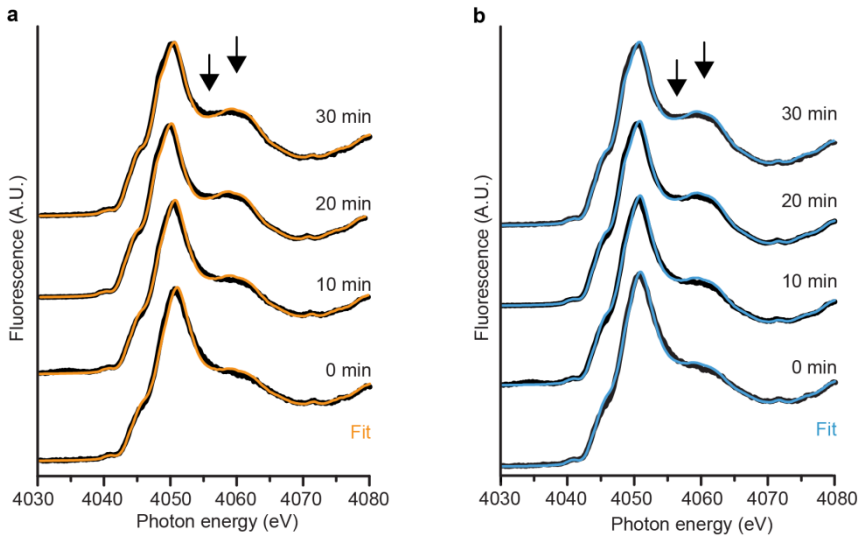
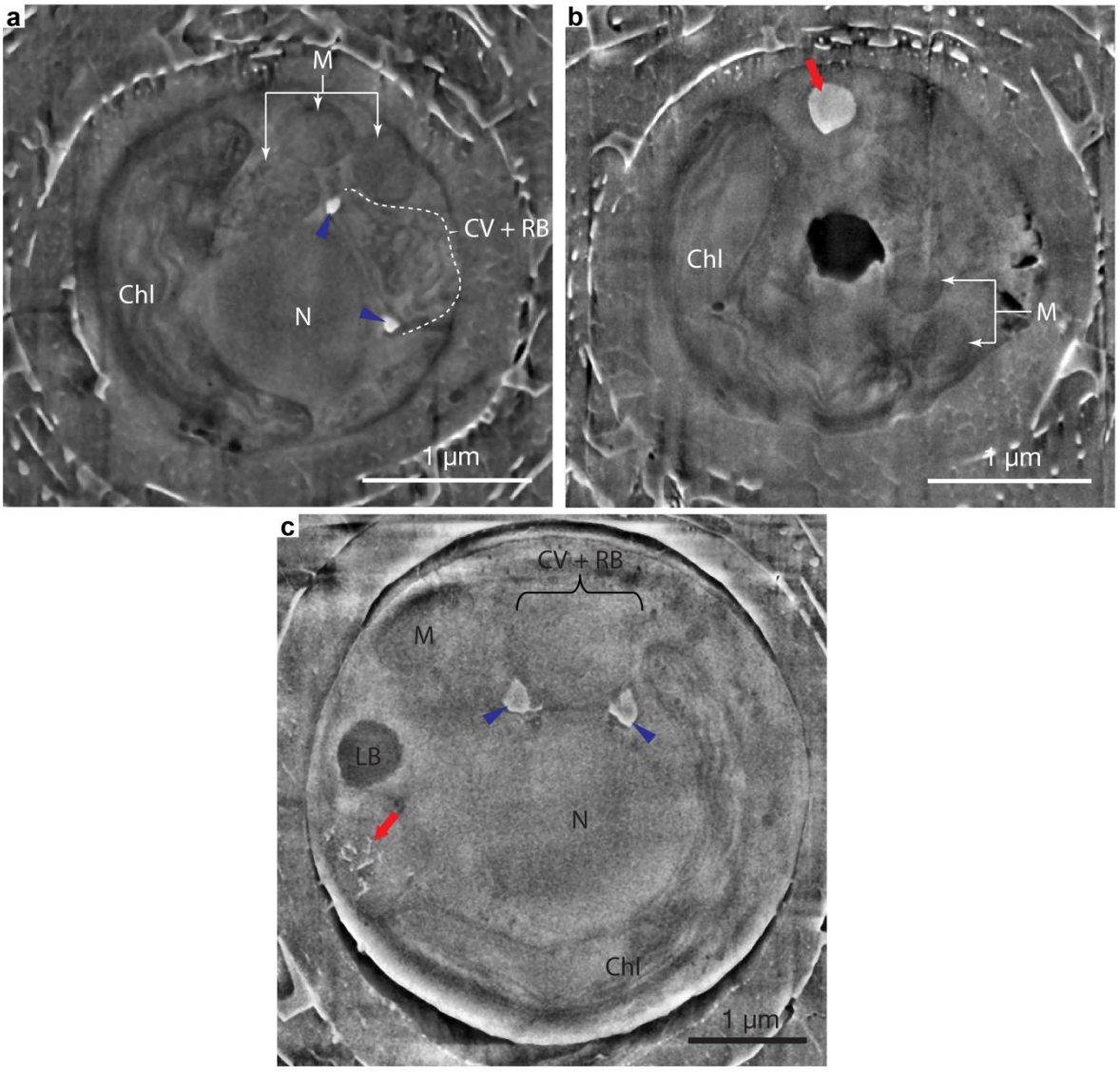


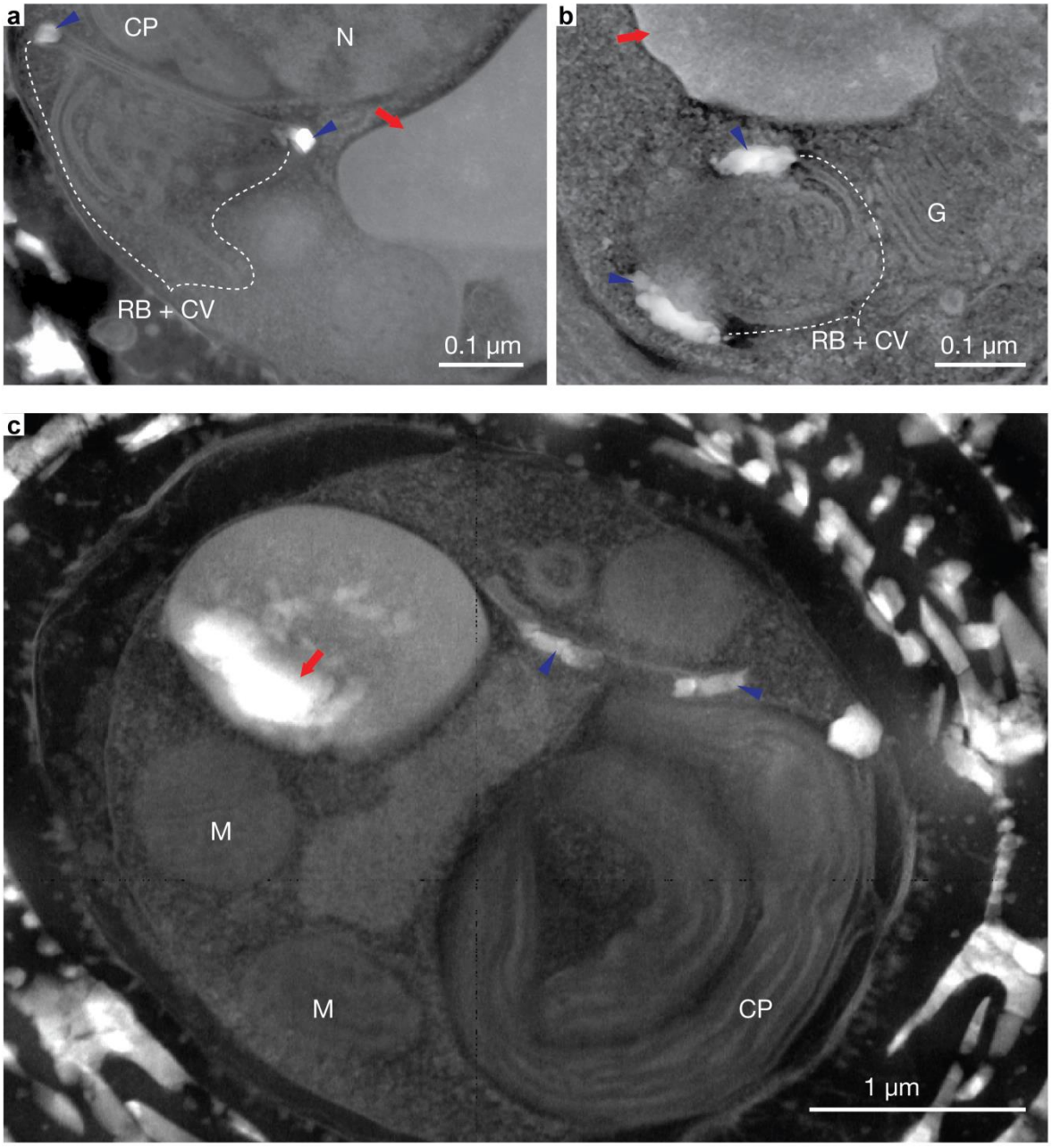
Supplementary Figure 1 | Turning coccolith formation 'on' and 'off'. (**a**) Growth behavior of *Emiliania huxleyi* in standard (10 mM $CaCl_2$) and calcium-depleted (0.1 mM $CaCl_2$) artificial seawater. The values are the mean of triplicate samples, and the bar indicates the standard error. (**b**) Plot showing the proportion of cells with an intracellular coccolith at different time points after induction of coccolith formation in calcium-depleted *E. huxleyi*. Bar graphs showing mean±s.d. of triplicate experiments with >500 cells in each experiment analyzed. (**c,d**) Light micrographs of EDTA-decalcified *E. huxleyi* cells. Arrowhead points to an intracellular coccolith and indicate the same position in the bright-field (**c**) and the cross-polarized light (**d**) image. Note that in cross-polarized light imaging contrast is related to birefringent material such as calcite. (**e**,**f**) Light micrographs of *E. huxleyi* cells grown for two weeks in calcium-depleted medium, which turns coccolith formation 'off'. Arrowhead indicates the same position in the bright-field (**e**) and the cross-polarized light (**f**) image.



Supplementary Figure 2 | Speciation of cellular calcium during the early stages of coccolith formation in *E. huxleyi*. Time-resolved evolution of the XANES spectra (black) of cells induced to form calcite and of the calculated fits using linear combinations of (**a**) the two reference standards (orange), coccolith calcite and free calcium ions, and (**b**) the three reference standards (blue), coccolith calcite, free calcium ions, and amorphous calcium phosphate. Arrows point to regions of highest divergence between fit and measured data.



Supplementary Figure 3 | Cryo FIB SEM imaging of vitrified *E. huxleyi* cells. 2D-slices from two cells ((a,b) cell 1; (c) cell 2) acquired with the In-lens secondary electron detector showing a cross-section of the coccolith *in statu nascendi* (blue arrowheads), a cross-section of the coccolith vesicle (CV) – reticular body (RB), a pool of diluted concentrations of calcium (red arrow), a lipid body (LB), nucleus (N), chloroplast (Chl), and mitochondria (M).



Supplementary Figure 4 | Ultrastructural features in unstained cell sections of *E. huxleyi*. HAADF-STEM images of thin-sectioned cells showing a cross-section of the coccolith *in statu nascendi* (blue arrowheads), a cross-section of the coccolith vesicle (CV) – reticular body (RB), a pool of densely concentrated calcium (red arrow), the Golgi apparatus (G), nucleus (N), chloroplast (CP), and mitochondria (M).

Supplementary Table 1 | Quantities of calcium and phosphorus in calcium-rich bodies calculated from the EDX data set of five cells

	Element	Weight %	Atomic %	Uncertainty %	P/Ca ratio
Cell 1	P(K)	63.17	66.3	1.06	2.76
	Ca(K)	29.54	23.96	0.71	
Cell 2	P(K)	31.60	29.18	1.03	1.83
	Ca(K)	22.37	15.97	0.84	
Cell 3	P(K)	59.46	62.3	1.39	2.65
	Ca(K)	29.07	23.53	0.92	
Cell 4	P(K)	60.08	59.88	1.63	3.49
	Ca(K)	22.26	17.15	1.06	
Cell 5	P(K)	53.44	55.76	0.67	2.22
	Ca(K)	31.20	25.16	0.46	

The quantities of calcium and phosphorus were determined from the equation: $C_A/C_B = K_{AB} * (I_A/I_B)$ where C stands for the concentration and I for the intensity of the components A and B, using the Tecnai imaging and analysis software TAI.

39