| Salt                                 | Concentration (mmol L <sup>-1</sup> ) |
|--------------------------------------|---------------------------------------|
| NaCl                                 | 419.6                                 |
| Na <sub>2</sub> SO <sub>4</sub>      | 28.9                                  |
| MgCl <sub>2</sub> .6H <sub>2</sub> O | 25.6                                  |
| KCl                                  | 9.3                                   |
| CaCl <sub>2</sub> .2H <sub>2</sub> O | 8.0                                   |
| KBr                                  | 2.4                                   |

**SUPPLEMENTARY TABLE 1** | Composition of the artificial seawater.

**SUPPLEMENTARY TABLE 2** | Calculated concentrations of DIC and inorganic carbon species for different carbonate alkalinities at equilibrium with 410 ppm CO<sub>2</sub> and after acidifying to pH 7 used in the kinetic experiments. The volume of HCl added to 2 mL of artificial seawater was determined from a linear regression between volume of acid and alkalinity (volume of acid ( $\mu$ L) = 3.512 (0.066) alkalinity (mequiv L<sup>-1</sup>) + 0.329 (0.162); R<sup>2</sup> = 0.999) where values represent one standard error.

|                           |            |                            | mmol L <sup>-1</sup> |          |                                  |               |  |  |  |  |  |
|---------------------------|------------|----------------------------|----------------------|----------|----------------------------------|---------------|--|--|--|--|--|
|                           | Vol 0.1 M  |                            |                      |          |                                  |               |  |  |  |  |  |
| Initial                   | HCl added  |                            |                      |          |                                  |               |  |  |  |  |  |
| Alkalinity                | to chamber |                            |                      |          |                                  |               |  |  |  |  |  |
| (mequiv L <sup>-1</sup> ) | (µl)       | pН                         | [DIC]                | $[CO_2]$ | [HCO <sub>3</sub> <sup>-</sup> ] | $[CO_3^{2-}]$ |  |  |  |  |  |
|                           |            | After equilibrium with air |                      |          |                                  |               |  |  |  |  |  |
| 0.2                       | 0          | 7.07                       | 0.210                | 0.013    | 0.195                            | 0.002         |  |  |  |  |  |
| 0.5                       | 0          | 7.46                       | 0.499                | 0.013    | 0.473                            | 0.013         |  |  |  |  |  |
| 1                         | 0          | 7.74                       | 0.963                | 0.013    | 0.902                            | 0.048         |  |  |  |  |  |
| 2                         | 0          | 8.01                       | 1.845                | 0.013    | 1.669                            | 0.164         |  |  |  |  |  |
| 5                         | 0          | 8.33                       | 4.273                | 0.013    | 3.529                            | 0.731         |  |  |  |  |  |
|                           |            |                            | After acidification  |          |                                  |               |  |  |  |  |  |
| 0.2                       | 1.5        | 7.00                       | 0.210                | 0.015    | 0.193                            | 0.002         |  |  |  |  |  |
| 0.5                       | 3.0        | 7.00                       | 0.499                | 0.036    | 0.458                            | 0.004         |  |  |  |  |  |
| 1                         | 5.5        | 7.00                       | 0.963                | 0.069    | 0.885                            | 0.009         |  |  |  |  |  |
| 2                         | 10.5       | 7.00                       | 1.845                | 0.132    | 1.697                            | 0.016         |  |  |  |  |  |
| 5                         | 2.6*       | 7.00                       | 4.273                | 0.305    | 3.930                            | 0.038         |  |  |  |  |  |

\* 1 M HCl

**SUPPLEMENTARY TABLE 3** | pH and concentrations of  $CO_2$  and  $HCO_3^-$ , and their ratio, calculated using the CO2SYS\_calc program (Pierrot and Wallace, 2006) for past, present and future (2100) atmospheric  $CO_2$  partial pressures relating to different shared socioeconomic pathways (SSPs) based on (Meinshausen et al., 2020). Values are calculated for 20°C, an alkalinity of 2.228 mequiv L<sup>-1</sup> and a salinity of 36.

| Scenario | Atmosphere CO <sub>2</sub> | pН   | CO <sub>2</sub> | HCO <sub>3</sub> - | Ratio HCO3 <sup>-</sup> : |
|----------|----------------------------|------|-----------------|--------------------|---------------------------|
|          | (ppm)                      |      | $(mmol L^{-1})$ | $(mmol L^{-1})$    | $\rm CO_2$                |
| Past     | 280                        | 8.17 | 0.009           | 1.705              | 189                       |
| Present  | 410                        | 8.03 | 0.013           | 1.830              | 141                       |
| SSP1-2.6 | 446                        | 8.00 | 0.014           | 1.855              | 133                       |
| SSP2-4.5 | 603                        | 7.89 | 0.019           | 1.939              | 102                       |
| SSP3-7.0 | 867                        | 7.75 | 0.027           | 2.023              | 75                        |
| SSP5-8.5 | 1135                       | 7.64 | 0.036           | 2.075              | 58                        |

**SUPPLEMENTARY TABLE 4** | Morphological and pigment characteristics of *P. oceanica* and *Z. marina*. Values are the means of three replicates (17 replicates for fresh weight to dry weight) with standard deviation in parentheses. The ratio of *P. oceanica* to *Z. marina* (P:Z) is given and the significance of the difference between the two species was determined with a t-test and presented as: NS, not significant, \* P<0.05, \*\* P<0.01.

| Characteristic                                   | P. oceanica   | Z. marina     | P:Z                   |
|--------------------------------------------------|---------------|---------------|-----------------------|
| 2-sided area per g FW ( $cm^2 g^{-1}$ )          | 141.3 (11.0)  | 181.0 (10.4)  | $0.78(0.1)^{*}$       |
| Dry weight per fresh weight (g g <sup>-1</sup> ) | 0.181 (0.006) | 0.197 (0.017) | $0.92 (0.1)^{**}$     |
| Chla per g FW ( $\mu$ g g <sup>-1</sup> )        | 711 (71)      | 405 (44)      | $1.76(0.3)^{**}$      |
| Chla :Chlb                                       | 1.72 (0.06)   | 1.76 (0.12)   | $0.98 (0.1)^{\rm NS}$ |

**SUPPLEMENTARY TABLE 5** | Protein ID and predicted location of carbonic anhydrase proteins in *Z. marina* determined using WoLF PSORT. Locations are: chloroplast (Chlo); extracellular (Extr); nucleus (Nucl); cytoplasm (Cyto); mitochondria (Mito); endoplasmic reticulum (E.R.); golgi (Golgi); vacuole (Vacu); plasma membrane (Plas). Proteins that shuttle between two locations are given dual localisation separated by an 'underscore'. The number of proteins of known localization with the most similar localization features to the query is given; the highest number(s) for each sequence are highlighted in bold and underlined. Sequences in red were also given a location in NCBI (<u>https://www.ncbi.nlm.nih.gov/protein/</u>), which is indicated by grey shading and underlined. The total number of carbonic anhydrase proteins in each location based on the most similar localization is given, with values in parentheses taking into account the different annotated locations.

|            | Predicted Location |          |            |          |       |      |       |          |      |          |           |            |           |
|------------|--------------------|----------|------------|----------|-------|------|-------|----------|------|----------|-----------|------------|-----------|
|            | Chlo               | Extr     | Nucl       | Cyto     | Mito  | E.R. | Golgi | Vacu     | Plas | E.R.     | Mito_plas | Golgi_plas | Cyto_Nucl |
| Protein ID |                    |          |            |          |       |      |       |          |      | _plas    |           |            |           |
| KMZ59859   |                    | 1        | 1          | <u>4</u> | 1     | 3.5  |       |          | 3.5  | <u>4</u> |           |            |           |
| KMZ63672   |                    | 3        |            | 2        |       | 2    |       | 7        |      |          |           |            |           |
| KMZ64507   |                    | <u>7</u> | 3          |          | 1.5   | 1    |       | 1        |      |          | 1.5       |            |           |
| KMZ69362   | 2                  | 1        | 1          | <u>3</u> | 2     | 1    |       | 2        | 2    |          |           |            |           |
| KMZ71968   | 2                  | 2        | 1          | 1        | 1     | 1    | 1     | <u>5</u> |      |          |           |            |           |
| KMZ73035   | <u>8</u>           | 3        | 2          | 1        |       |      |       |          |      |          |           |            |           |
| KMZ75401   | 1                  | <u>5</u> | 1          | 1        |       | 1    | 1     | 4        |      |          |           |            |           |
| KMZ56823   | <u>7</u>           | 1        |            | 2        | 2     |      |       | 1        | 1    |          |           |            |           |
| KMZ56166   | <u>6</u>           |          | 2          | 2        | 1     |      |       | 2        | 1    |          |           |            |           |
| KMZ73424   | 4                  |          | <u>6.5</u> |          | 2     |      |       |          |      |          |           | 1          | 4         |
| Number     | 3 (4)              | 2        | 1          | 2 (4)    | 0 (2) | 0    | 0     | 2        | 0    | 1        | 0         | 0          | 0         |



**SUPPLEMENTARY FIGURE 1** | Example results for one replicate of *Z. marina*. A, Measured rate of net photosynthesis versus the concentration of inorganic carbon (DIC) at air-equilibrium (open squares) and after acidification to pH 7.0 (closed triangle). B, Modelled versus measured rate of net photosynthesis with linear regression of the relationship. C, Modelled rate of net photosynthesis versus concentration of  $CO_2$ . D, Modelled rate of net photosynthesis versus concentration of  $HCO_3^-$ .

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