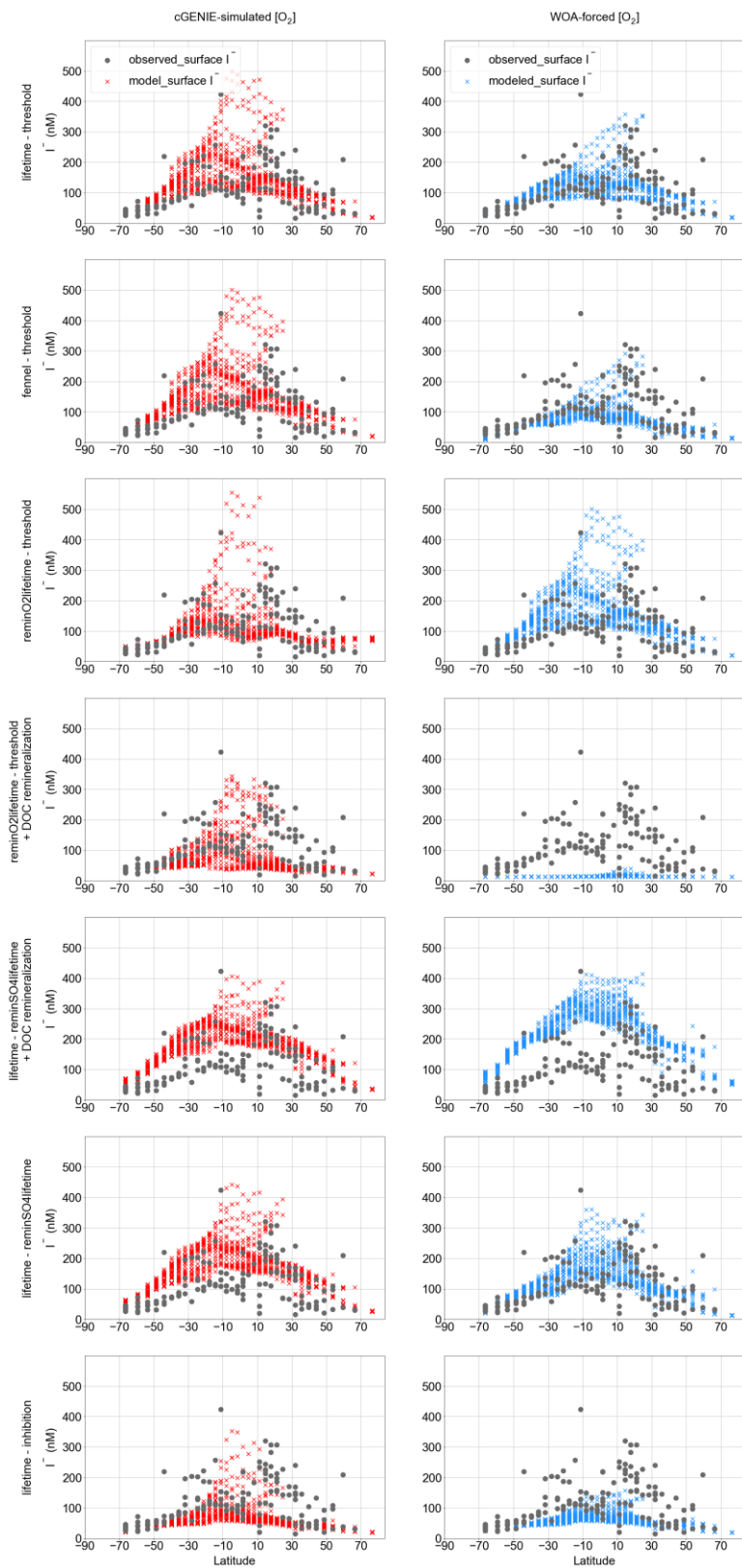
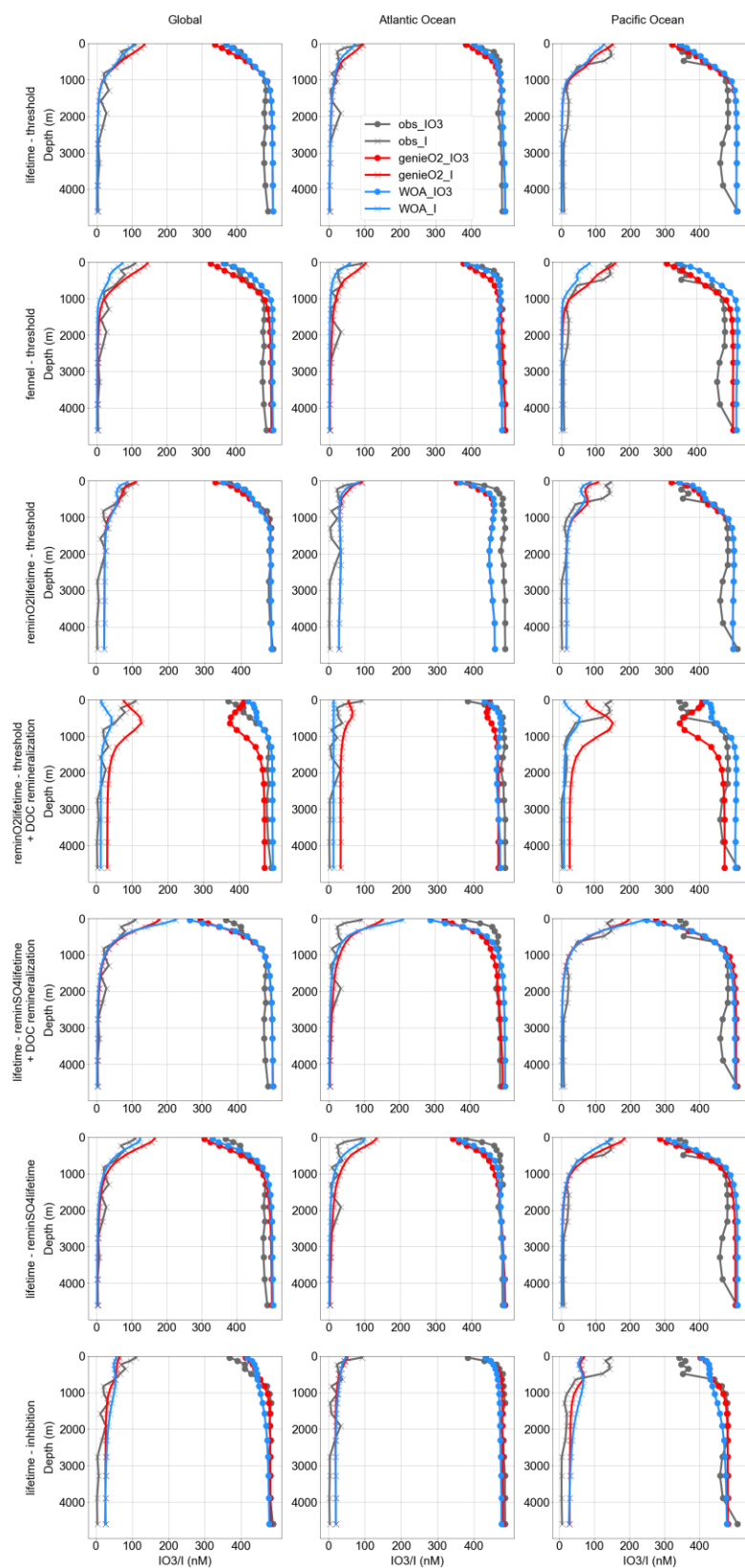


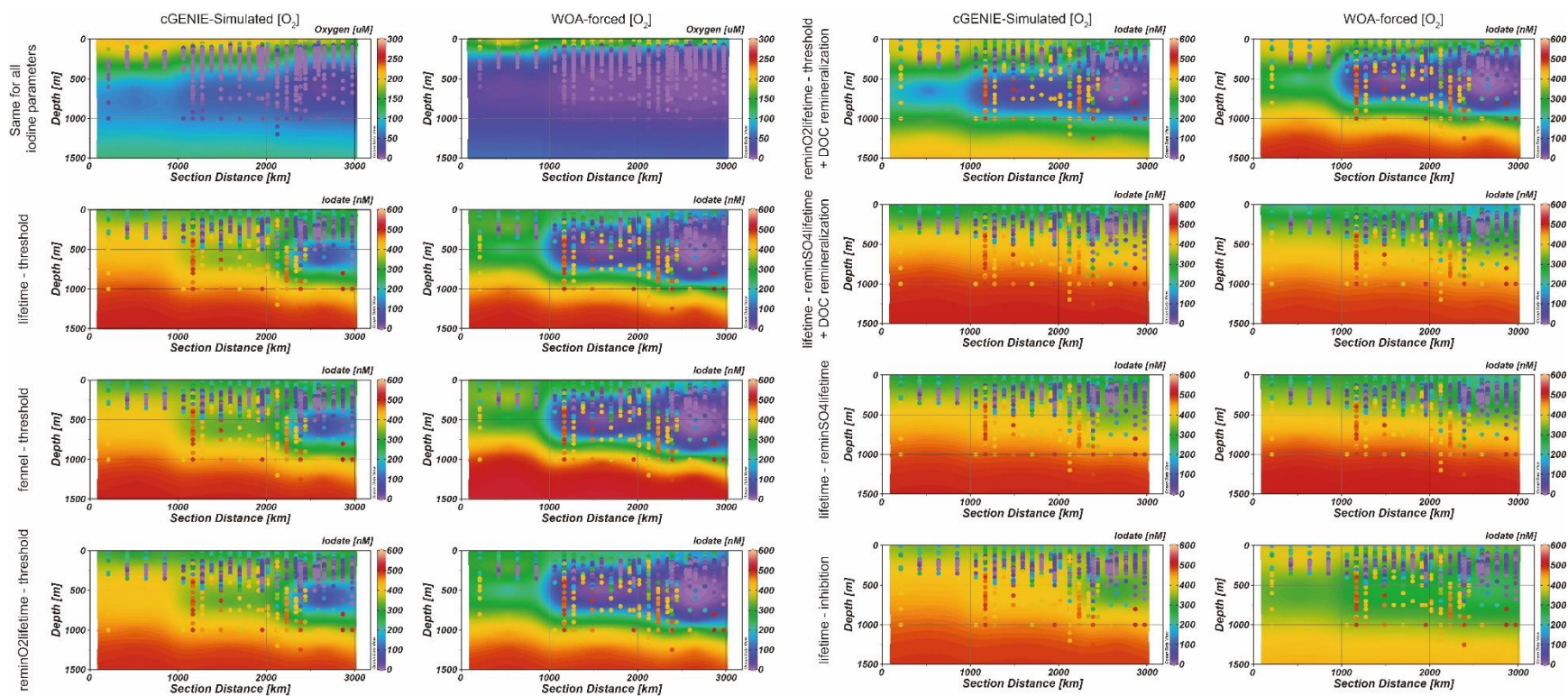
Figure S1. The three-dimensional model skill score array of all the experiment ensembles tested in this study.



**Figure S2. Modeled latitudinal surface iodide distribution compared with observation with the cGENIE simulated [O<sub>2</sub>] and the WOA-[O<sub>2</sub>] restoring forcing.**

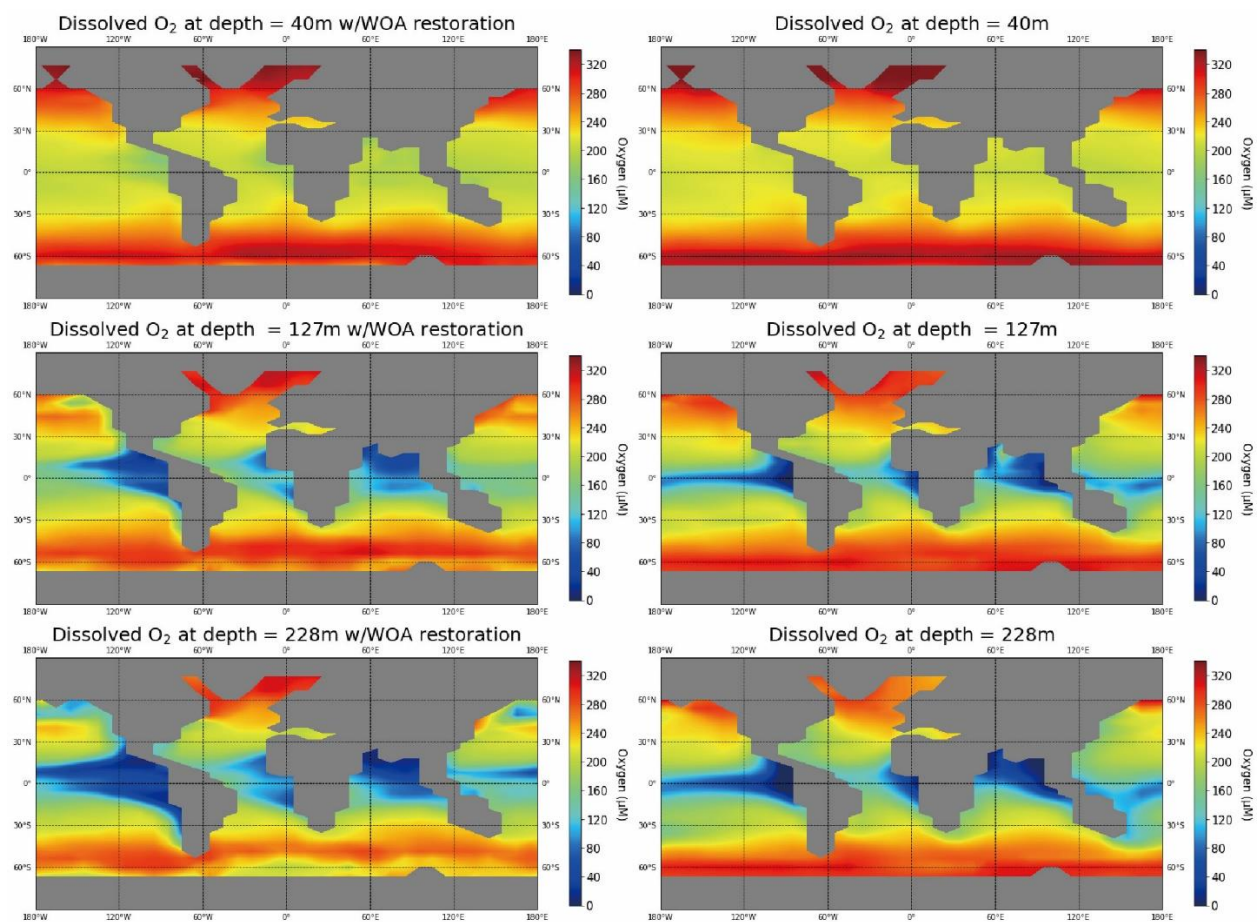


**Figure S3. Modeled averaged iodine (including iodate and iodide) depth profile among global ocean, the Pacific, the Atlantic compared with observation.**

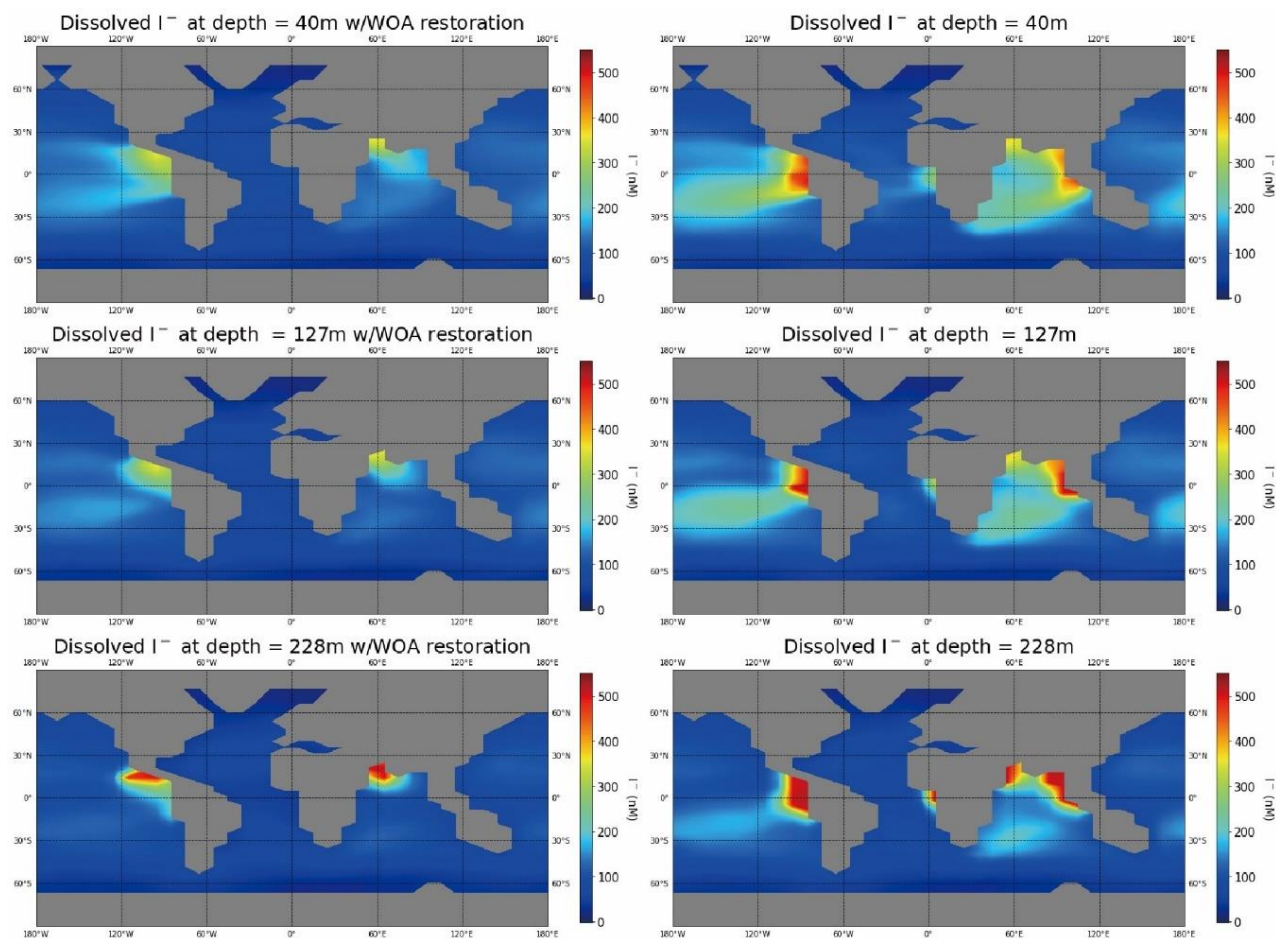


**Figure S4. Modeled (contour) and observed (colored dots) transects of  $\text{IO}_3^-$  in the ETNP.**

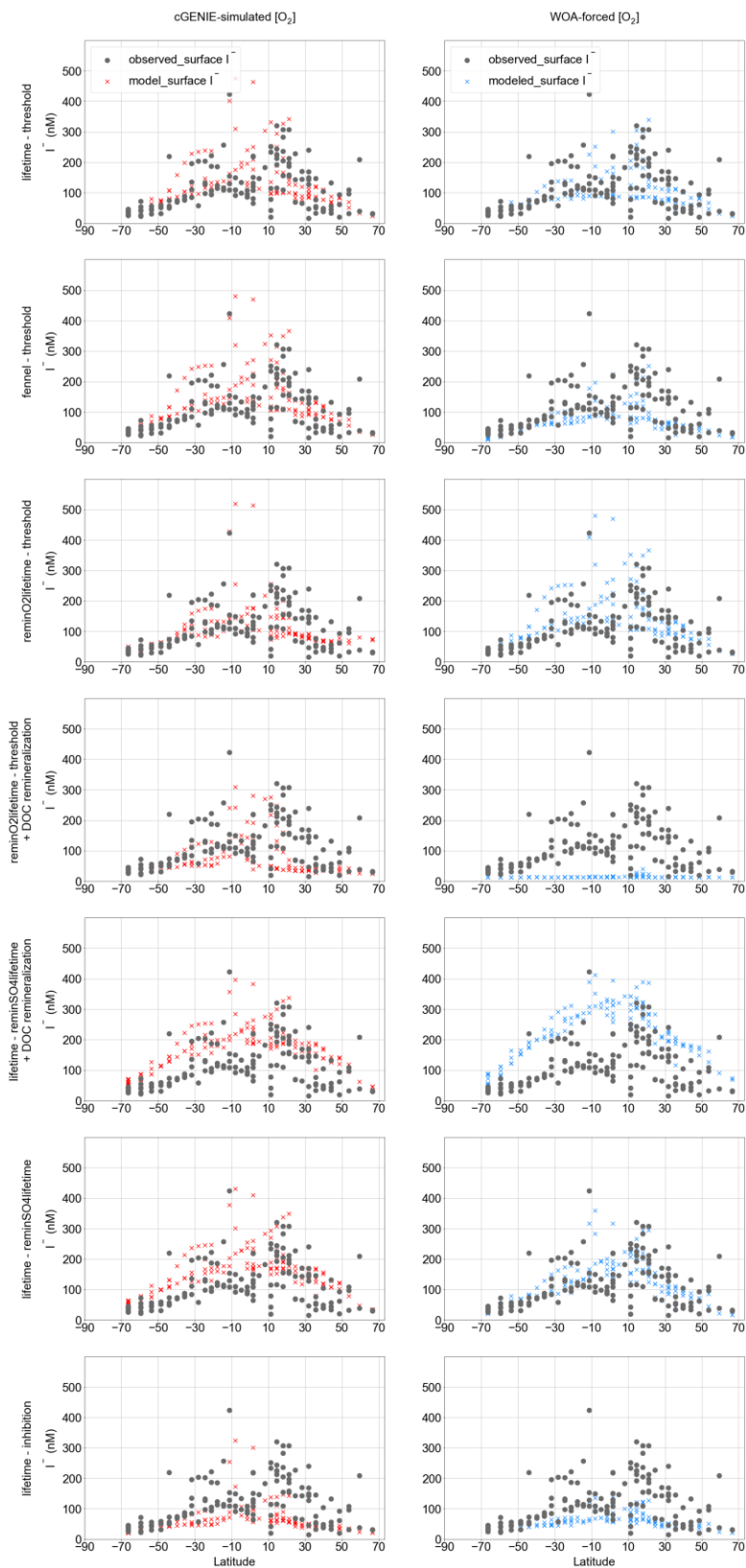




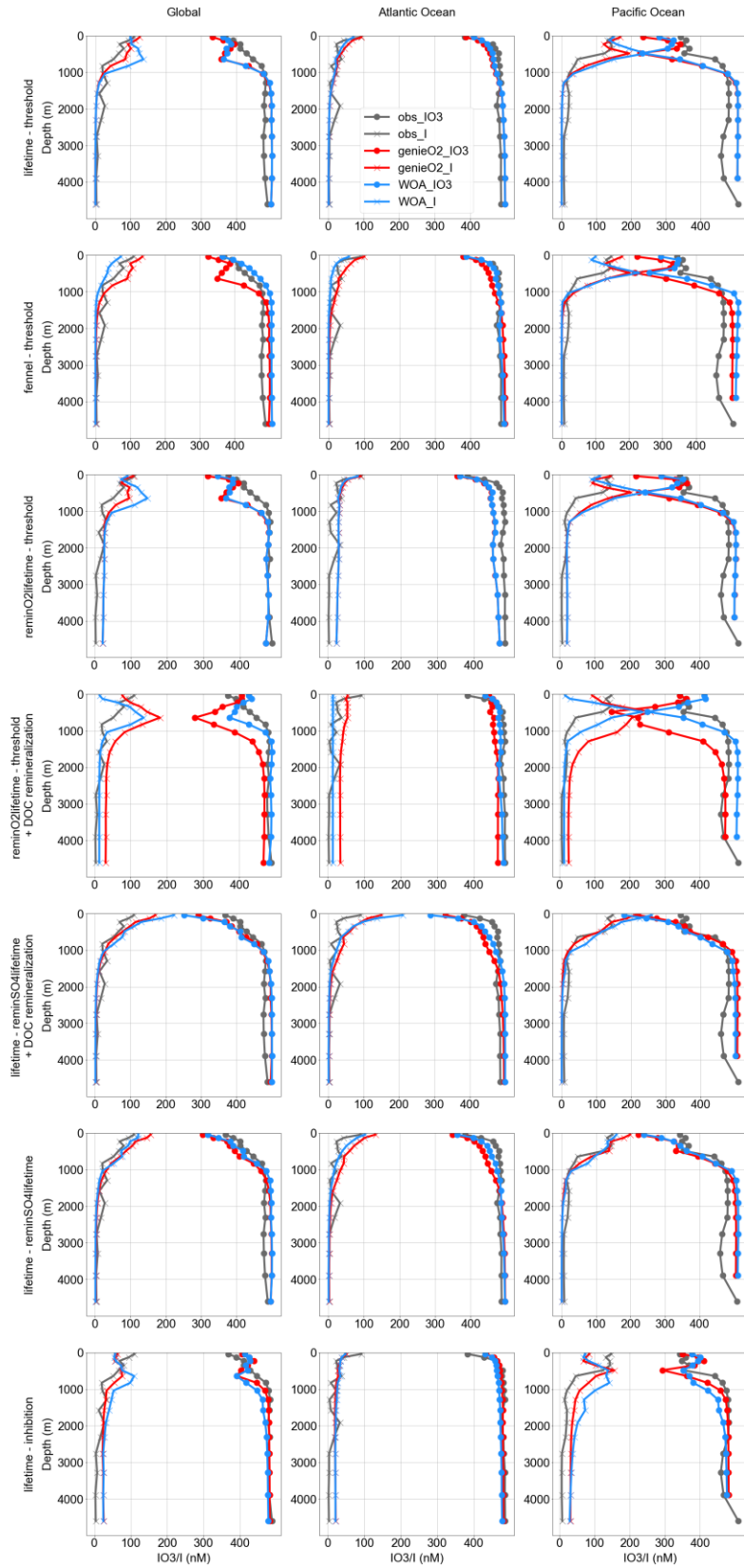
**Figure S5.** The modeled  $O_2$  distribution in the surface and subsurface with  $[O_2]$  forced to restore the WOA observation and without forcing.



**Figure S6.** The modeled  $I^-$  distribution in the surface and subsurface with  $[O_2]$  forced to restore the WOA observation and without forcing. Associated iodine cycling parameters: “lifetime” = 50 yrs; “threshold” =  $10 \times 10^{-6}$  mol; I:C ratio =  $1.5 \times 10^{-4}$  mol/mol. This is an example of cGENIE overestimating surface  $I^-$  when the overall model performance is tuned to the best.

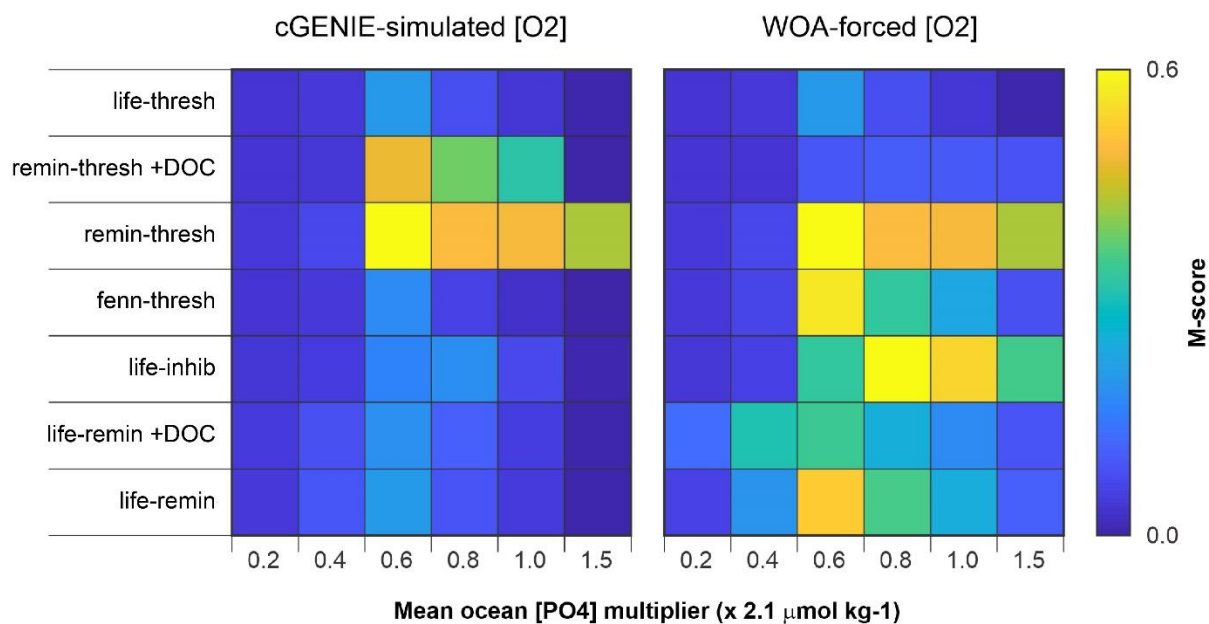


**Figure S7. Modeled latitudinal surface iodide distribution compared with observation with the cGENIE simulated  $[O_2]$  (masked only include grid points with corresponding observations) and the  $[O_2]$  restoring forcing.**



**Figure S8. Modeled averaged iodine (including iodate and iodide, and masked only include grid points with corresponding observations) depth profile among global ocean, the Pacific, the Atlantic compared with observation.**





**Figure S9.** The model skill scores between modeled and measured I:Ca during the pre-OAE2. The iodine cycling parameters are derived from modern simulations with cGENIE-simulated [O<sub>2</sub>] and WOA-forced [O<sub>2</sub>], respectively. lifetime-thresh = lifetime-threshold; remin-thresh +DOC = reminO2lifetime-threshold +DOC remineralization; remin-thresh = reminO2lifetime-threshold; fenn-thresh = fennel-threshold; life-inhib = lifetime-inhibition; life-remin +DOC = lifetime-reminSO4lifetime +DOC remineralization; life-remin = lifetime-reminSO4lifetime

| Parameter description |   | Iodine oxidation parameters |   |  | Iodine reduction parameters |                                  |  | I:C ratio (× 10 <sup>-4</sup> mol/mol) |  |
|-----------------------|---|-----------------------------|---|--|-----------------------------|----------------------------------|--|--|--|
|                       |   | 'lifetime' (years)          | 'reminO2lifetime' (× 10 <sup>-5</sup> mol/kg) | 'Fennel' (Inhibition μM O <sub>2</sub> ) | constant/                   | 'threshold' (μM O <sub>2</sub> ) | 'inhibition' (μM/ year <sup>-1</sup> ) |  | 'reminSO4lifetime' (× 10 <sup>-6</sup> mol/kg) |
| Simulation 1          | cGENIE O <sub>2</sub>                     | 10-170                      | \   | \  |                             | 1-110                            | \                                      | \                                      | 0.5-3.5  |
|                       | WOA                                       | 10-170                      | \   | \  |                             | 1-110                            | \                                      | \                                      | 0.5-3.5  |
|                       | no excess I <sup>-</sup> filtration       | 10-170                      | \   | \  |                             | 1-110                            | \                                      | \                                      | 0.5-3.5  |
|                       | no excess I <sup>-</sup> filtration + WOA | 10-170                      | \   | \  |                             | 1-110                            | \                                      | \                                      | 0.5-3.5  |
| Simulation 2          | cGENIE O <sub>2</sub>                     | \                           | 0.01-1  | \  |                             | 1-100                            | \                                      | \                                      | 0.5-3.5  |
|                       | WOA                                       | \                           | 0.01-1  | \  |                             | 1-100                            | \                                      | \                                      | 0.5-3.5  |
|                       | cGENIE O <sub>2</sub> +DOC remin.         | \                           | 0.01-1  | \  |                             | 1-100                            | \                                      | \                                      | 0.5-3.5  |
|                       | WOA +DOC remin                            | \                           | 0.01-1  | \  |                             | 1-100                            | \                                      | \                                      | 0.5-3.5  |
| Simulation 3          | cGENIE O <sub>2</sub>                     | 10-170 (1/k)                | \   | 20                                       |                             | 1-110                            | \                                      | \                                      | 0.5-3.5  |
|                       | WOA                                       | 10-170 (1/k)                | \   | 20                                       |                             | 1-110                            | \                                      | \                                      | 0.5-3.5  |
|                       | WOA(alternative)                          | 10-170 (1/k)                | \   | 20                                       |                             | 1-110                            | \                                      | \                                      | 0.5-3.5  |
| Simulation 4          | cGENIE O <sub>2</sub>                     | 0.5-50                      | \   | \  |                             | \                                | 10/0.1-10                              | \                                      | 0.5-3.5  |
|                       | WOA                                       | 0.5-50                      | \   | \  |                             | \                                | 10/1.0-10                              | \                                      | 0.5-3.5  |
| Simulation 5          | cGENIE O <sub>2</sub>                     | 10-170                      | \   | \  |                             | \                                | \                                      | 0.01-1                                 | 0.5-3.5  |
|                       | WOA                                       | 10-170                      | \   | \  |                             | \                                | \                                      | 0.01-1                                 | 0.5-3.5  |
|                       | cGENIE O <sub>2</sub> +DOC remin.         | 10-170                      | \   | \  |                             | \                                | \                                      | 0.01-1                                 | 0.5-3.5  |
|                       | WOA +DOC remin                            | 10-170                      | \   | \  |                             | \                                | \                                      | 0.01-1                                 | 0.5-3.5  |

**Table S1. The cGENIE iodine redox options and the associated range of parameters of these options in this research. Only simulations 1-3 are chosen for detailed discussion in the manuscript for higher M scores and reasonably replicating iodine ocean gradients (See Discussion).**

| Parameter description |   | Iodine oxidation parameters |   |  | Iodine reduction parameters |                                  |  | I:C ratio (× 10 <sup>-4</sup> mol/mol) | Model skill score (global) |  |
|-----------------------|---|-----------------------------|---|--|-----------------------------|----------------------------------|--|--|----------------------------|--|
|                       |   | 'lifetime' (years)          | 'reminO2lifetime' (× 10 <sup>-5</sup> mol/kg) | 'Fennel' (Inhibition μM O <sub>2</sub> ) | constant/                   | 'threshold' (μM O <sub>2</sub> ) | 'inhibition' (μM/ year <sup>-1</sup> ) |  |                            | 'reminSO4lifetime' (× 10 <sup>-6</sup> mol/kg) |
| Simulation 1          | cGENIE O <sub>2</sub>                     | 50                          | \   | \  |                             | 10                               | \                                      | \                                      | 1.5                        | 0.305  |
|                       | WOA                                       | 50                          | \   | \  |                             | 10                               | \                                      | \                                      | 1.5                        | 0.385  |
|                       | no excess I <sup>-</sup> filtration       | 50                          | \   | \  |                             | 10                               | \                                      | \                                      | 2.5                        | 0.316  |
|                       | no excess I <sup>-</sup> filtration + WOA | 50                          | \   | \  |                             | 10                               | \                                      | \                                      | 2.5                        | 0.393  |
| Simulation 2          | cGENIE O <sub>2</sub>                     | \                           | 0.1   | \  |                             | 10                               | \                                      | \                                      | 3.5                        | 0.266  |
|                       | WOA                                       | \                           | 0.1   | \  |                             | 10                               | \                                      | \                                      | 3.5                        | 0.365  |
|                       | cGENIE O <sub>2</sub> +DOC remin.         | \                           | 1   | \  |                             | 50                               | \                                      | \                                      | 0.5                        | 0.213  |
|                       | WOA +DOC remin                            | \                           | 0.1   | \  |                             | 10                               | \                                      | \                                      | 3.5                        | 0.302  |
| Simulation 3          | cGENIE O <sub>2</sub>                     | 50 (1/k)                    | \   | 20                                       |                             | 10                               | \                                      | \                                      | 1.5                        | 0.308  |
|                       | WOA                                       | 10 (1/k)                    | \   | 20                                       |                             | 10                               | \                                      | \                                      | 3.5                        | 0.385  |
|                       | WOA(alternative)                          | 50 (1/k)                    | \   | 20                                       |                             | 10                               | \                                      | \                                      | 1.5                        | 0.379  |
| Simulation 4          | cGENIE O <sub>2</sub>                     | 50                          | \   | \  |                             | \                                | 10/0.1                                 | \                                      | 1.5                        | 0.289  |
|                       | WOA                                       | 10                          | \   | \  |                             | \                                | 10/1.0                                 | \                                      | 1.5                        | 0.289  |
| Simulation 5          | cGENIE O <sub>2</sub>                     | 50                          | \   | \  |                             | \                                | \                                      | 0.5                                    | 1.5                        | 0.307  |
|                       | WOA                                       | 10                          | \   | \  |                             | \                                | \                                      | 0.1                                    | 2.5                        | 0.363  |
|                       | cGENIE O <sub>2</sub> +DOC remin.         | 50                          | \   | \  |                             | \                                | \                                      | 1                                      | 1.5                        | 0.300  |
|                       | WOA +DOC remin                            | 10                          | \   | \  |                             | \                                | \                                      | 0.1                                    | 0.5                        | 0.337  |

**Table S2. The performance of the cGENIE iodine simulations of all the combinations of parameters in this research and associated parameterization when the model reaches the best global M score.**

| Section         | Site location     | Depth      |       |      | Pre-CIE d13C  | CIE d13C      | Post-CIE d13C | Pre-CIE I:Ca | CIE I:Ca | Post-CIE I:Ca |
|-----------------|-------------------|------------|-------|------|---------------|---------------|---------------|--------------|----------|---------------|
| Raia del Pedale | Shallow water     | Few meters |       |      | -0.353        | 2.013         | 0.681         | 0.688        | 0.837    | 1.296         |
| Demerara Rise   | Low lat. pelagic  | Below base | storm | wave | -27.616 (org) | -23.816 (org) | -26.345 (org) | 0.427        | 0.530    | 0.153         |
| Tarfaya         | Low lat. pelagic  | Below base | storm | wave | -27.019 (org) | -24.841 (org) | -26.729 (org) | 0.836        | 0.494    | 0.869         |
| South Ferriby   | Mid. lat. Pelagic | Below base | storm | wave | 3.200         | 3.780         | 3.260         | 3.656        | 4.953    | 3.344         |
| Eastbourne      | Mid. lat. Pelagic | Below base | storm | wave | 2.796         | 4.335         | 3.803         | 2.351        | 3.005    | 4.241         |
| Newfoundland    | Mid. lat. Pelagic | Below base | storm | wave | 2.803         | 3.064         | 2.697         | 2.883        | 1.552    | 2.364         |

**Table S3. The Cretaceous OAE2 sections where I:Ca data were measured from for model-data comparison. The d13C from Demerara Rise and Tarfaya are organic data (d13Corg) instead of carbonate data.**