

**Supplementary information**

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# Rethinking ocean alkalinity enhancement

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# Rethinking Ocean Alkalinity Enhancement

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- Supplementary Tables 1-3
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**Supplementary Table 1.** Anthropogenic analogues of OAE where human activities lead to elevated alkalinity in the ocean not intended for the purposes of carbon dioxide removal.

| Context                     | Purpose                   | Alkalinity source         | Description                                                                                | Reference          |
|-----------------------------|---------------------------|---------------------------|--------------------------------------------------------------------------------------------|--------------------|
| Marine ecosystem management | Pest control              | CaO                       | Quicklime dispersed over reefs to eradicate pest/invasive echinoderms                      | Ref. <sup>1</sup>  |
| Aquaculture                 | pH control                | Various                   | Alkalinity added to marine aquaculture systems to elevate pH                               | Ref. <sup>2</sup>  |
|                             | Biofouling removal        | Ca(OH) <sub>2</sub> , CaO | Biofouling species removed from infrastructure or grow lines by application lime solutions | Ref. <sup>3</sup>  |
|                             | Substrate preparation     | CaO                       | Lime dispersed over settlement substrate to promote larval settlement                      | Ref. <sup>4</sup>  |
|                             | Acid soil neutralisation  | Various lime              | Alkalinity added to pond soils to neutralise acidity                                       | Ref. <sup>5</sup>  |
|                             | Spawning induction        | Various lime              | Alkalinity added culture to induce spawning                                                | Ref. <sup>6</sup>  |
|                             | Coagulation/flocculation  | Various lime              | Alkalinity added to wastewater to remove suspended solids                                  | Ref. <sup>7</sup>  |
| Wastewater                  | pH adjustment             | Various lime              | Alkalinity added to wastewater to increase pH prior to discharge                           | Ref. <sup>8</sup>  |
|                             | Bactericide               | Various lime              | Alkalinity added to wastewater for bactericidal effects                                    | Ref. <sup>9</sup>  |
|                             | Heavy metal removal       | Various lime              | Alkalinity added to wastewater for removal of heavy metals                                 | Ref. <sup>10</sup> |
|                             | Nutrient removal          | Various lime              | Alkalinity added to wastewater for removal of nutrients                                    | Ref. <sup>11</sup> |
| Waste products              | Disposal                  | Steel slag, bauxite       | Disposal of alkaline waste products in coastal regions                                     | Ref. <sup>12</sup> |
|                             | Sand capping              | Steel slag, bauxite       | Capping of polluted sediments with industrial by products to bind with cations             | Ref. <sup>13</sup> |
|                             | Artificial reefs          | Steel slag                | Steel slag used to make artificial reefs for kelp and corals                               | Ref. <sup>14</sup> |
|                             | Hydrogen sulfide removal  | Steel slag                | Steel slag used to remove hydrogen sulfide in sediments                                    | Ref. <sup>15</sup> |
|                             | Habitat restoration       | Steel slag                | Steel slag used as an alternative to sand in restoring seagrass beds and tidal flats       | Ref. <sup>16</sup> |
| Acidic soil neutralisation  | Environmental remediation | Various lime              | Hydrated lime used to neutralise soils in acidified tidal areas for ecosystem restoration  | Ref. <sup>17</sup> |
|                             | Agricultural liming       | Various lime              | Hydrated lime used to neutralise soils in acidic soils to improve crop yield               | Ref. <sup>18</sup> |

**Supplementary Table 2.** Supplementary Table 2 corresponds with that in Figure 3 in the main text and details includes footnotes that clarify how assessments were made.

| Collateral perturbation                             |                                  | Carbonate chemistry <sup>b</sup>                                          |                         | Contaminants <sup>c</sup>            |                  |                                   |                  |                                      |                   |                   | Particulates <sup>d</sup>           |
|-----------------------------------------------------|----------------------------------|---------------------------------------------------------------------------|-------------------------|--------------------------------------|------------------|-----------------------------------|------------------|--------------------------------------|-------------------|-------------------|-------------------------------------|
|                                                     |                                  | pH (NBS)                                                                  | pCO <sub>2</sub> (µatm) | Earth Metals (mol kg <sup>-1</sup> ) |                  | Nutrients (mol kg <sup>-1</sup> ) |                  | Trace Metals (mol kg <sup>-1</sup> ) |                   |                   | Total suspended solids (TSS) (mg/L) |
|                                                     |                                  |                                                                           |                         | Mg                                   | Ca               | Si                                | P                | Ni                                   | Fe                | Mn                |                                     |
| Seawater level <sup>e</sup>                         |                                  | 8.17                                                                      | 433                     | 10 <sup>-2</sup>                     | 10 <sup>-2</sup> | 10 <sup>-6</sup>                  | 10 <sup>-6</sup> | 10 <sup>-9</sup>                     | 10 <sup>-10</sup> | 10 <sup>-10</sup> | Highly variable                     |
| Example guidelines Nova Scotia, Canada <sup>f</sup> |                                  | 8.7                                                                       | 105                     | n.l.                                 | n.l.             | n.l.                              | n.l.             | 1.40 × 10 <sup>-7</sup>              | n.l.              | n.l.              | 25                                  |
| Source material                                     | Alkalinizing agent(s)            | Relative magnitude of collateral perturbations (approximate) <sup>a</sup> |                         |                                      |                  |                                   |                  |                                      |                   |                   |                                     |
| Dunite <sup>g</sup>                                 | Mg <sub>2</sub> SiO <sub>4</sub> | Low                                                                       | Low                     | Med                                  | Med              | High                              | Low              | Med                                  | Med               | Low               |                                     |
| Limestone <sup>h</sup>                              | CaCO <sub>3</sub>                | Low <sup>m</sup>                                                          | Low <sup>m</sup>        | Med                                  | Med              | Low                               | Low              | Low                                  | Low               | Low               |                                     |
| Brucite <sup>i</sup>                                | Mg(OH) <sub>2</sub>              | Med                                                                       | Med                     | Med                                  | Med              | Low                               | Low              | Low                                  | Low               | Low               |                                     |
| Steel slag <sup>j</sup>                             | CaO, MgO                         | Med                                                                       | Med                     | Med                                  | Med              | Med                               | Med              | Low                                  | Med               | Med               |                                     |
| Iron ore <sup>k</sup>                               | Fe <sup>2+</sup>                 | Low                                                                       | Low                     | Low                                  | Low              | Low                               | Low              | Low                                  | High              | Low               |                                     |
| Brine <sup>l</sup>                                  | HCl depleted brine               | High                                                                      | High                    | None                                 | None             | None                              | None             | None                                 | None              | None              | Low                                 |

<sup>a</sup>Scores (none, low, med and high) refer to the relative magnitude of the perturbation between OAE source materials. Importantly, the scores assigned do not indicate the magnitude of biological effects as this will depend on factors such as the scale of the deployment and organismal sensitivity to the particular perturbation. Furthermore, the relative magnitudes will also depend on the scale of the deployment, the specific composition of source materials, their treatment (for example grain size effect on TSS), and the reactive environment to which the corresponding alkalinizing agent is added.

<sup>b</sup>The scores for carbonate chemistry perturbations are based on reactivity of each source material. For example, dunite alters the carbonate system over timescales of months to years, whereas methods involving brines alter the carbonate system near instantaneously.

<sup>c</sup>The scores for dissolution products are based on the weight percentages of elements in the respective source materials as referenced in the footnotes<sup>g-k</sup>. For example, the Si weight percentage of dunite, limestone, brucite, steel slag, iron ore and brine are 16.78 %, < 1 %, < 1 %, 8.07 %, < 1 %, and 0 %, respectively. In this case, olivine was given a score of high, steel slag a med, limestone, brucite, and iron ore all low and brine was given a score of none.

<sup>d</sup>To this date it is not possible to assign a meaningful magnitude of collateral perturbation for TSS, as this will vary on a case by case basis with how much material is added. For brines, however, we can safely assume that NaOH-enriched (HCl-depleted) brine will cause the formation of amorphous brucite, which will usually quickly re-dissolve<sup>19</sup>.

<sup>e</sup>Typical order of magnitude in the ocean in mol kg<sup>-1</sup>. Values for pH are on the NBS scale and values for pCO<sub>2</sub> are in µatm and calculated assuming this pH, a temperature of 25 °C, a salinity of 35, a dissolved inorganic carbon concentration of 2165 µmol kg<sup>-1</sup>. Concentrations for Mg, Ca, Si, P, Ni, Fe, Mn taken from ref.<sup>20</sup>. Values for pH taken from 2024 data point from ref.<sup>21</sup> and converted to the NBS scale assuming a temperature of 25 °C and a salinity of 35.

<sup>f</sup>Example guideline values for each collateral perturbation in the province of Nova Scotia, Canada where a commercial OAE project operates<sup>22,23</sup>. The value for pH is the upper acceptable limit and the pCO<sub>2</sub> (µatm) limits are calculated assuming this pH and a temperature of 25 °C, a salinity of 35, a dissolved inorganic carbon concentration of 2165 µmol kg<sup>-1</sup>. Values for contaminants and TSS refer to increases relative to background values<sup>22</sup>; n.l. refers to no regulatory limits available.

<sup>g</sup>Olivine mineral composition assumed to be identical to ref.<sup>24</sup>, who used olivine-rich rocks from a quarry in Norway.

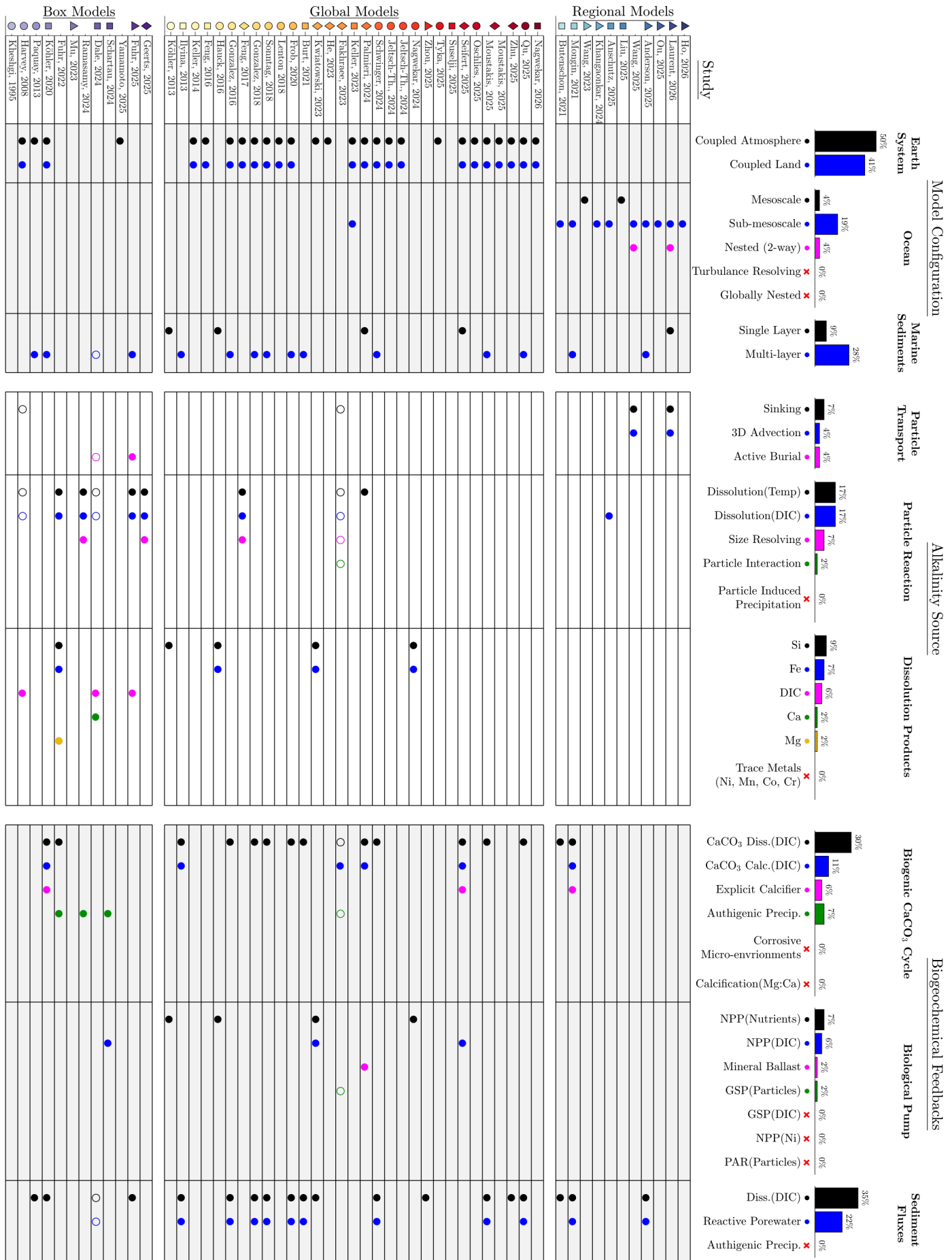
<sup>h</sup>Limestone composition assumed to be identical to ref.<sup>25</sup>.

<sup>i</sup>Assuming Mg(OH)<sub>2</sub> composition according to ref.<sup>26</sup>.

<sup>j</sup>Assuming the use of Basic Oxygen Furnace Slag with a composition identical to ref.<sup>27</sup>.

<sup>k</sup>Assuming the use of hematite with a composition identical to ref.<sup>28</sup> and alkalinity generated in sediments<sup>29</sup>. <sup>l</sup>Assuming the hydrochloric acid removed from the brine is safely neutralised/stored on land. If the acid is neutralised with alkaline minerals (for example via olivine<sup>30</sup>) and this liquid transferred back into the ocean<sup>30</sup>, the relative score for each perturbation (other than carbonate system) would be the equivalent of the neutralising mineral.

<sup>m</sup>Assuming a saturation state for calcite of 0.8



## Supplementary Note 1. Review of OAE Modelling Studies

52 unique OAE modeling studies were considered to review the scale of alkalinity added and carbon removed (**Figure 5**, main text) as well as the configuration of the model used in the simulations (**Figure 6** main text; **Supplementary Table 3**). Two studies<sup>68,79</sup> included simulations in two distinct models, thus 54 total models are considered in **Figure 6** (main text) and **Supplementary Table 3**. In studies with more than one simulation experiment conducted using the same model, we selected 1–4 (median = 2) experiments to plot in **Figure 5b** (main text). Experiments were selected to cover a representative range in the scale/location/type of perturbations conducted, without weighting results too far towards a single study. Note, **Fig. 5a, 6** and **Supplementary Table 3** are agnostic to which experiments were selected.

### Quantification of OAE and CDR Rates

The rate of OAE is reported as the mean total moles alkalinity contained within all anthropogenically added alkaline material added to the ocean per year (**Figure 5**, main text). The rate of OAE is averaged over the duration of the deployment if it is not fixed in time, as in simulations which scale OAE in proportion to projected emissions<sup>31,32,64</sup>. In five cases, alkaline material is added for less than a year, but allowed to equilibrate<sup>32,43,73</sup>, dissolve<sup>46</sup> or both<sup>58</sup> in a longer running simulation. Here, the annual rate is expressed as the cumulative amount added during the first year of the simulation rather than the (higher) time-normalized rate to avoid exaggerating the amount of alkalinity actually added. If a simulation explicitly resolves the dissolution of solid-phase alkalinity, the OAE rate reflects the amount of alkalinity that would be added assuming complete/instantaneous dissolution, such that incomplete/slow dissolution effectively reduces the efficiency of CDR. However, in two cases dissolution is assumed to occur from a fixed, unlimited source of solid alkaline material<sup>51,59</sup>. Here, the OAE rate must be reported as the rate of dissolved alkalinity entering solution. Finally, the rate of OAE is integrated in space over the specified deployment footprint.

The rate of CDR is reported as the mean amount of additional carbon removed from the atmosphere per year, averaged over the length of the simulation following the start of OAE (**Figure 5**, main text). In simulations with a coupled, responsive atmosphere, CDR is reported explicitly as the reduction in atmospheric carbon content relative to the control (27 out of 54 models; **Supplementary Table 3**). In simulations with an offline, forced atmosphere, CDR is inferred from the increase in marine DIC, and thus yields a higher efficiency by ignoring Earth System feedbacks (27 out of 54 models; **Supplementary Table 3**). In models where CDR is not directly reported, it is estimated graphically.

Instead of attempting to constrain a direct comparison of efficiency, OAE and CDR are quantified to most consistently express the scale of alkalinity added and carbon sequestered across studies. This is best expressed as an annualized rate to control for large differences in the time scales of simulations (1 month to 380 years). While the median ratio of CDR to OAE ( $CO_{AE}:Alk_{ant}$ ) surveyed here (0.63; **Figure 5b** (main text), red line) is comparable to the theoretical OAE efficiency of seawater ( $\sim 0.84$ ; **Figure 5b**, black line), this is not an exact comparison. The efficiency of OAE (known as  $\eta$ <sup>33</sup> or  $CO_{AE}:Alk_{ant}$  in **Figure 5**, main text) is typically expressed as the additional moles of carbon sequestered per mole of alkalinity added over a specific timescale<sup>73</sup>. However, here we are comparing time-averaged rates which are not necessarily computed over the same period. In simulations with continuous OAE integrated longer than equilibration timescales, **Figure 5b** (main text) gives a fairly robust estimate of end-member efficiency. However, for short simulations with incomplete equilibration or long simulations following a shorter OAE deployment period (with CDR rates saturating later in the simulation),  $CO_{AE}:Alk_{ant}$  will underestimate the true efficiency. Accordingly, values are reported to compare the scale of perturbations across simulations, not as direct estimates of efficiency.

In four models either  $Alk_{ant}$  or  $CO_{AE}$  are unquantified, either because they are area/mass normalized perturbations<sup>45,55,75</sup> or because alkalinity is added as an inert tracer without resolving the carbonate system<sup>40</sup>. These models are not plotted in **Figure 5** (main text) or included in related statistics regarding the size of simulated perturbations. However, their model attributes are counted in **Figure 6** (main text) and reported in **Supplementary Table 3**.

### Clarification of Model Feature and Feedbacks

Model features and feedbacks are separated into (1) Model Configuration, (2) Alkalinity Source, and (3) Biogeochemical Feedbacks. These categories loosely map onto the first three recommendations for model development suggested by

Fennel et al. (ref.<sup>83</sup>): “(1) supporting multiscale simulations with sufficiently high-fidelity flow fields, (2) faithfully simulating the near-field dynamics associated with alkalinity addition, (3) capturing feedbacks to OAE owing to biological and geochemical responses.”

The frequency of model features and feedbacks included in 54 reviewed models is summarized in in **Figure 6** (main text). The features and feedbacks included in each individual model are reported in **Supplementary Table 3**. Circles denote inclusion in each model. Circle colors are for easier visual alignment with attribute labels. Note, key features that have not yet been included in any surveyed model are included in the table. In three models with an offline forcing for alkalinity dissolution<sup>35,42,50</sup>, an *unfilled marker* denotes the feature is only included in the forcing model for the sourcing of alkalinity, but not in the model that ultimately computes CDR through air-sea gas exchange. The summary statistics/histogram in **Figure 6** is repeated for convenience in the histogram in **Supplementary Table 3**.

## (1) Model Configuration

**Model Configuration** refers to the architecture, resolution, and coupling of the modeling system.

**Earth System** refers to the configuration of non-ocean earth system components. *Coupled Atmosphere* and *Coupled Land* refer to whether the ocean model was coupled to a responsive carbon reservoir in the atmosphere and terrestrial biosphere, respectively, regardless of whether these components were highly resolved or highly idealized<sup>37,63,64,68</sup>. About half the models surveyed (27 of 54) do not include a coupled atmosphere and can be considered to be concentration-driven, rather than emissions-driven, scenarios. Outside the model of Wang et al. (ref.<sup>40</sup>), which focuses on particle dissolution dynamics not carbonate chemistry, all models compute gas exchange based on the disequilibrium between oceanic and atmospheric CO<sub>2</sub>. Most (22 of 27) models with a coupled atmosphere were run as a full Earth System Model and include a coupled biosphere as well. One model also includes a coupled solid earth, in which silicate/carbonate weathering rates were coupled to atmospheric CO<sub>2</sub> (ref.<sup>53</sup>).

**Ocean** refers to the resolution and configuration of the ocean model. *Mesoscale* and *Sub-mesoscale* refer to a nominal resolution of  $\leq 0.25^\circ$  and  $\leq 0.1^\circ$ , respectively. Noting, *Mesoscale* includes eddy-permitting grids ( $\sim 0.25^\circ$ ) and *sub-mesoscale* does not necessarily resolve all sub-mesoscale features. Importantly, resolving mesoscale dynamics primarily alters horizontal velocities. However, further resolving sub-mesoscale dynamics has a larger influence on vertical velocities<sup>84</sup>, which could alter the time-scale or air-sea equilibration. *Nested (2-way)* refers to a nested, down-scaled, high-resolution ( $\leq 0.1^\circ$ ) ocean grid with 2-way coupling of biogeochemical and/or particle tracers, but not necessarily physics. Nested models range from  $<100$  m resolution in nested regional configurations<sup>58</sup> to  $0.1^\circ$  resolution in nested global configurations<sup>77</sup>.

No surveyed OAE models are *Turbulence Resolving* or include *Global to Regional Nesting*. *Turbulence Resolving* refers to either *Large Eddy Simulations* and *Direct Numerical Simulations*, which resolve turbulent flows at small scales. Their omission precludes the explicit representation of how turbulence impacts particle dissolution dynamics, settling rates, and secondary precipitation<sup>83</sup>. *Global to Regional Nesting* refers to the nested inclusion of high-resolution (10-100 m) regional/coastal models within a coarser resolution global ocean model. Such models are required to simultaneously resolve regional-scale feedbacks close to the OAE perturbation where alkalinity may be concentrated and un-equilibrated, as well as downstream equilibration time scales which may be required for carbon accounting and quantification of integrated open-ocean, global-scale feedbacks if many local deployments eventually come online.

**Marine Sediments** refers to the configuration of the marine sediment model (if included) and the fate of particles if they reach the seafloor. *Single Layer* sediment models include a single sediment layer (sometimes effectively the seafloor) that can accumulate and store organic and inorganic particles as they are dissolved or remineralized, but vary in how those rates are derived (see **Sediment Fluxes**). Models without a single sediment layer, instantly dissolve or bury some fraction of particles reaching the seafloor. *Multi-layer* sediment models can further transport solid and dissolved material in the sediments relative to the sediment-ocean boundary. This material can ultimately be buried beneath the bottom sediment layer, at which point it is permanently removed from the model, making the ocean an 'open' system. *Multi-layer* sediment models are often referred to as *1-D Reaction-Transport* or *Early Diagenetic* models but vary in the complexity of reactions included (see **Sediment Fluxes**).

## (2) Alkalinity Source

The **Alkalinity Source** refers to the means by which anthropogenic alkalinity is produced and added to the model. Idealized OAE simulations can be conducted through the direct addition of dissolved alkalinity to a desired grid-cell. Such studies assume that a 'pure' liquid-phase alkalinity source has been added (NaOH, CaO, Ca(OH)<sub>2</sub>) and are agnostic to its means of upstream production. More complex models explicitly simulate the dissolution of solid-phase alkalinity sources to varying degrees and/or include additional reactive dissolution products associated with a given alkalinity source.

Roughly two thirds (38 of 54) of models were forced with idealized alkalinity-only perturbations and thus included no features required to explicitly simulate or distinguish the production or source of alkalinity. The remaining studies differ in the way they resolve the transport and dissolution of anthropogenic alkaline particles and/or their associated dissolution products.

Importantly, the features described here refer exclusively to the transport and dissolution of *anthropogenic* alkaline particles, not necessarily to the way in which *biogenic* particles that may also be represented in the ocean biogeochemical model are represented.

**Particle Transport** refers to the movement of anthropogenic alkaline particles in the water column or sediments. *Sinking* refers to negatively buoyant particles which explicitly sink through the water column at a fixed<sup>40,58</sup> or variable rate<sup>35,42</sup>. *3D Advection* refers to whether particles are coupled to a 3D hydrodynamic ocean model. *Active Burial* refers to the explicit movement and possible burial of anthropogenic particles through the sediment column relative to the sediment-ocean interface through implicit bioirrigation, sediment deposition, or advection in a 1D reaction-transport model. Note, one study<sup>58</sup> allows a fixed fraction of accumulating anthropogenic alkaline particles on the seafloor to be lost to the sediments. However, this burial term is set to 0 in the presented simulations, so no *Active Burial* is included.

**Particle Reaction** refers to the manner in which anthropogenic alkaline particles dissolve and/or interact with the physical and biogeochemical environment. *Dissolution(Temp)* and *Dissolution (DIC)* refer to whether particle dissolution has a functional dependence on temperature and/or carbonate chemistry (i.e. DIC), respectively. The specific carbonate system dependency varies across studies and source material but generally refers to pH for olivine dissolution<sup>42,44,45,55,64</sup> and  $\Omega_{\text{(Calcite)}}$  for CaCO<sub>3</sub> dissolution<sup>35,45,50,59</sup>. *Size Resolving* refers to particles that explicitly change size and properties associated with their size (i.e. sinking speed, dissolution rate) as they dissolve. This differs from models that consider size via parameter selection but do not explicitly resolve changes in size as the model evolves<sup>44</sup>. Note, where models<sup>40,58</sup> include sinking anthropogenic alkaline particles, but no temperature, DIC or size resolving dissolution dependencies, then dissolution rates are fixed. *Particle Interaction* refers to whether anthropogenic particles can interact with other particulate elements of the model. This includes fragmentation and aggregation with anthropogenic or biogenic particles as well as fragmentation and ingestion from zooplankton<sup>42</sup>.

No models resolve *Particle Induced Precipitation*. *Particle Induced Precipitation* refers to the ability of particles to act as nucleation sites for authigenic precipitation, creating a positive feedback that could induce runaway precipitation at substantially lower saturation states ( $\Omega \sim 5$ ; ref.<sup>85</sup>) than observed in particle-free water ( $\Omega > 10$ ; ref.<sup>86</sup>). One study<sup>56</sup> assumes a fixed exponential calcification rate above saturation states ( $\Omega > 7$ ) consistent with presence of particulate nucleation sites, but does not vary this rate with respect to particle size or concentration.

**Dissolution Products** refers to any chemically or biologically active tracer that is released as alkalinity is explicitly or implicitly dissolved. In some models without an explicit source of alkalinity, this is accomplished by forcing the model with nutrient additions (Si, Fe) in stoichiometric proportion to the assumed alkalinity source, typically olivine<sup>36,47–49</sup>. Only two models include chemically active mineral dissolution products beyond Si, Fe, or DIC<sup>45,50</sup>.

No models consider *Trace Metals (Ni, Mn, Cr, Co)* other than Fe as chemically or biologically reactive dissolution products. The most important omission may be *Ni*, which can act as both a nutrient and/or toxin depending on phytoplankton species (see **Biological Pump**).

### (3) Biogeochemical Feedbacks

**Biogeochemical Feedbacks** refer to mechanistic pathways through which anthropogenic alkalinity can alter a baseline biogeochemical property that is relevant to the CDR potential of the intervention (i.e. additionality<sup>87</sup>). If there is no pathway for OAE to alter a given process, then it is not considered a feedback.

**Biogenic  $\text{CaCO}_3$  Cycle** refers to the pelagic removal, transfer, and/or addition of alkalinity in the water column through the precipitation, sinking, and/or dissolution of inorganic minerals. It is agnostic to the fate of those minerals at the seafloor or in the sediments (see **Sediment Fluxes**) but is inclusive of corals where included<sup>53,54</sup>.  $\text{CaCO}_3$  *Dissolution(DIC)* and  $\text{CaCO}_3$  *Calcification(DIC)* refer to whether the dissolution and precipitation, respectively, of biogenic  $\text{CaCO}_3$  is functionally dependent on carbonate chemistry. Importantly, this only refers to biogenic  $\text{CaCO}_3$  associated with explicit or implicit calcifying organisms (plankton or corals), not anthropogenic particles (see **Alkalinity Source**) or abiotic authigenic precipitation (see **Authigenic Precipitation**). In all cases,  $\text{CaCO}_3$  *Dissolution(DIC)* varies specifically with  $\Omega_{\text{Calcite}}$ . In most cases  $\text{CaCO}_3$  *Calcification(DIC)* also varies with  $\Omega_{\text{Calcite}}$ ; however, a more complex empirical dependency on  $\text{HCO}_3^-$ ,  $\text{CO}_2$ , and pH is employed in Seifert et al., (ref.<sup>52</sup>). Some models additionally include nutrient or temperature dependencies on calcification<sup>32,52</sup>.

Dynamic calcification rates are often included implicitly through a variable PIC:POC ratio in the marine phytoplankton community, reflective of implicit changes in both community composition (calcifiers:non-calcifiers) and physiology (PIC:POC)<sup>32,42,51</sup>. However, three models include an explicit calcifying class of phytoplankton<sup>52</sup> or coral<sup>53,54</sup>, and are noted as having an *Explicit Calcifier*. None include a calcifying zooplankton class<sup>88</sup>. Note, Anderson et al. (ref.<sup>72</sup>) uses the same core biogeochemical model as Mongin et al., (ref.<sup>54</sup>), but in temperate Tasmanian waters with no coral (or calcifying plankton), and thus no feedback on calcification is included in the model. Other OAE studies include dynamic calcification rates implicitly dependent on, e.g., temperature or nutrients, but without any dependency on carbonate chemistry, and thus no feedback from OAE (e.g., refs.<sup>62,67</sup>). Several other biogeochemical models include an explicit calcifying phytoplankton<sup>89–91</sup> or even zooplankton<sup>92</sup>, but these configurations were not been used in the surveyed OAE perturbations.

*Authigenic Precipitation* refers to the abiotic, spontaneous precipitation of solid-phase minerals from the aqueous phase, inclusive of secondary precipitation and reverse weathering reactions. It is agnostic to the mineral being precipitated but refers here exclusively to precipitation in the water column, rather than sediments (see **Sediment Fluxes**). *Authigenic Precipitation* has been parameterized empirically<sup>45</sup>, thermodynamically<sup>42,55</sup>, and as a fixed exponential rate above a prescribed threshold saturation state<sup>56</sup>. The empirical parameterization is implicitly inclusive of more complex kinetics than implied by simple thermodynamic, equilibrium parameterizations. Note:  $\text{CaCO}_3$  precipitation in Fakhraee et al. (ref.<sup>42</sup>) is included only in the 1D water column forcing model used to quantify anthropogenic alkaline particle dissolution and implicitly represents both biotic and abiotic (i.e., authigenic) precipitation, but does not distinguish between the two.

No models resolve responsive *Corrosive Micro-environments* or *Calcification(Mg:Ca)*. *Corrosive Micro-environments* refer to pelagic micro-environments such as zooplankton guts<sup>93,94</sup> or marine snow<sup>95</sup> with highly reduced pH relative to the surrounding seawater. Simulating such environments could be necessary to understand the suppression of  $\text{CaCO}_3$  dissolution, and thus natural alkalinity addition, in the upper water column. While some models may implicitly include corrosive zooplankton guts<sup>48,96,97</sup>, they are not influenced by the carbonate chemistry of ambient seawater and thus have no feedback pathway. *Calcification(Mg:Ca)* refers to whether the addition of reactive  $\text{Mg}^{2+}$  (from olivine dissolution) or  $\text{Ca}^{2+}$  (from  $\text{CaCO}_3$  dissolution) can influence the direct rate of calcite precipitation<sup>98</sup> or the relative rate of calcite vs aragonite polymorphs, which could alter the depth of the lysocline<sup>99</sup>.

**Biological Pump** refers to the biologically-mediated transfer of carbon from the surface ocean to depth. *NPP(Nutrients)* refers to whether net primary production (NPP) is influenced by OAE through the addition of anthropogenic nutrient dissolution products derived from the alkalinity source, agnostic to the type of nutrient. 4 of 54 models simulate the implicit dissolution of mafic minerals (e.g., olivine) through simultaneous addition of Si, Fe, or both in stoichiometric proportions to the assumed source mineral and alkalinity addition<sup>36,47–49</sup>. Many additional models include Si and/or Fe growth dependencies on phytoplankton but do not add nutrients during the OAE perturbation, thus there is no feedback

pathway. *NPP(DIC)* refers to whether net primary production is influenced directly or indirectly by carbonate chemistry. While CO<sub>2</sub> limitation is well understood to occur at low concentrations (<100 µatm) (refs.<sup>100,101</sup>) that could occur during the transient period prior to equilibration<sup>102</sup>, only two models have included the direct effect of carbonate chemistry on growth rates<sup>52</sup>. Seifert et al. (ref.<sup>52</sup>) employ an empirical dependency on pH, HCO<sub>3</sub><sup>-</sup>, and CO<sub>2</sub> parameterized against a global data set of laboratory experiments<sup>103</sup>. Schartau et al. (ref.<sup>56</sup>) use an empirical dependency on pH<sup>100</sup>. They also include an indirect effect of carbonate chemistry on NPP through a pH dependency on bacterial growth rates, prescribed as a bell-shaped optima<sup>104</sup>. One other study<sup>47</sup> includes an indirect effect of carbonate chemistry on NPP through pH dependency on bioavailable iron<sup>96</sup>. However, this has been shown to have limited influence in the OAE context<sup>105</sup> and was not the focus of the study<sup>47</sup>.

*Mineral Ballast* refers to changes in the sinking rate and/or degradation of organic matter triggered by an increase in CaCO<sub>3</sub> ballast material<sup>106,107</sup>. While several models assume that organic material associated with an implicit calcifying group sinks faster<sup>43,47,67</sup>, only one also allows the rate of implicit calcification to be influenced by carbonate chemistry, thus establishing a feedback pathway<sup>51</sup>. Mineral ballast through increased growth of a siliceous diatom class is not included here. *GSP(Particles)* refers to whether the presence of anthropogenic particles can influence Gross Secondary Production (GSP), i.e. zooplankton growth, either by direct toxicity (e.g., clogged mesh in gelatinous filter feeders<sup>108</sup>) or indirectly as a poor-quality prey option capable of reducing grazing<sup>42</sup>. Only one model includes any such effects and does so only with implicit zooplankton in an offline forcing model used to quantify anthropogenic particle dissolution — not the downstream ecological impacts of reduced grazing through increased inorganic particle ingestion<sup>42</sup>.

No models resolve *GSP(DIC)*, *NPP(Ni)*, or *PAR(Particles)*. *GSP(DIC)* refers to the direct influence of carbonate chemistry on zooplankton growth rates, which have primarily been investigated only in response to acidification<sup>106</sup>. *NPP(Ni)* refers to the direct effect of nickel on NPP. Nickel can act as both a nutrient and/or toxin depending on phytoplankton species and concentration<sup>109,110</sup>, with nitrogen-fixing bacteria most likely to benefit from elevated concentrations<sup>111</sup>. *PAR(Particles)* refers to the reduction in photosynthetically available radiation via increased turbidity and light attenuation associated with anthropogenic particles, which could reduce NPP<sup>112</sup>.

**Sediment Fluxes** refer to the consumption or generation of natural alkalinity in the sediments, differing from the processes resolved in the water column (see **Biogenic CaCO<sub>3</sub> Cycle**) and dissolution of anthropogenically sourced particles (see **Alkalinity Source**). *CaCO<sub>3</sub> Dissolution (DIC)* refers to whether natural CaCO<sub>3</sub> in the sediments dissolves as a function of carbonate chemistry. CaCO<sub>3</sub> in the sediments can be derived prognostically from the sinking detrital material of calcifying plankton or can be a prescribed (and fixed) characteristic of the sediments<sup>54,72</sup> or both<sup>53</sup>. *Reactive Porewater* refers to models in which coupled porewater chemistry is responsive to the precipitation and dissolution of CaCO<sub>3</sub> and degradation of organic material. In models without reactive porewater, CaCO<sub>3</sub> dissolution rates are computed based on the carbonate chemistry of the overlying ocean grid cell. In models with *Reactive Porewater* the decomposition of organic matter or dissolution of alkaline minerals can create more or less corrosive conditions, respectively, relative to overlying waters. No models resolve *Authigenic Precipitation* in the sediments.

## Supplementary Note 2. Interactions between Models and Observations in Assessment of OAE

It is widely recognized that precise carbon budgeting for OAE will require models and that these models must be informed by observations<sup>83,113,114</sup>. However, it is not always obvious how exactly to leverage diverse types of observations (e.g. measurement, monitoring and experimentation) during model development. Here, we consider 4 pathways through which site-specific observations could help build reliable, trustworthy models, fit-for-purpose for OAE-based “monitoring, reporting, and verification”.

**1. Model Forcing and Design.** Observations can be used to describe the non-prognostic physical and biogeochemical conditions that will be used either to force the model or design its fundamental architecture. From the forcing perspective this includes any prescribed information used to drive the model, such as seasonal variability in boundary conditions, river discharge rates, wind stress, wave activity and incoming solar radiation. From the design perspective this includes information about what is - and is not - represented explicitly or implicitly in the model, such as sediment characteristics (e.g. porosity, composition), plankton functional groups (e.g. calcifying phytoplankton/zooplankton), and macro (Si) and micro (Fe) nutrient tracers. Importantly, any decision to prescribe or omit a given aspect of the model (rather than represent it explicitly/prognostically) based on observations or knowledge of the baseline should be well understood as insensitive to the OAE perturbation.

**2. Model Parameterization.** Process-focused experimentation can be used to parameterize key processes in the model via in-situ/ex-situ experimentation that directly reflects specific local conditions. This requires conducting targeted, process-based studies to empirically derive mathematical relationships that can then be mechanistically incorporated into the model to describe, for instance, the functional response of grazers to prey abundance<sup>115</sup> of coccolithophores to carbonate chemistry<sup>103</sup>. Locally calibrated empirical parameterizations could extend to any number of biogeochemical processes resolved in the model, but should prioritize those that:

- 1) *Determine the sourcing of alkalinity*, such as temperature and grain size dependent dissolution and sinking rates of solid-source alkalinity.
- 2) *Exhibit feedbacks on the additionality of alkalinity*, such as carbonate chemistry dependencies on the precipitation/dissolution of phases that consume alkalinity.
- 3) *Are likely to be regionally specific* compared to known relationships derived at either the global scale or unrepresentative sites, such as air-sea exchange rates and local nutrient and/or alkalinity sources. These also include processes that are poorly constrained in global models even if they are only indirectly related to the perturbation, such as the rate parameters governing phytoplankton<sup>116</sup> and zooplankton<sup>117</sup> growth.

**3. Model Assimilation and Tuning.** Field observing can be used to nudge or tune model output towards baseline observations. From the physical perspective, if the OAE deployment is unlikely to alter physical properties (e.g. temperature, salinity, bottom-water velocities, surface mixing), then the physical ocean model could potentially be nudged towards observations in a data assimilating framework<sup>83,113</sup>. Since OAE deployments are designed to alter seawater chemistry, the biogeochemical properties of the model should evolve in a free-running biogeochemical model in order to best differentiate perturbed from baseline scenarios. However, biogeochemical parameters (e.g. rate terms, nutrient affinities, temperature dependencies) should be tuned towards baseline observations during model development. This requires minimizing a cost function between emergent model properties and adequately resolved (in space and time) local measurements. Importantly, if any biogeochemical parameters included in the tuning algorithm were originally derived from region-specific empirical experiments (see **Point 2**, above), then tuning should be performed within the range of uncertainty quantified in the empirical experimentation. Biogeochemical models should be tuned to recreate climatologic spatio-temporal variability in e.g. carbonate system parameters, chlorophyll, nutrient, and POC fields, most relevant to a region-specific OAE deployment.

**4. Model Validation.** Direct field measurements must be used to validate the model both in baseline (counterfactual) and perturbation scenarios. Validation of baseline scenarios can be accomplished largely through the tuning process. However, validation of perturbation scenarios should be distinct from the tuning process and evaluated against the model's emergent response to OAE perturbation (i.e. the training dataset and the test dataset should be distinct). Here, direct field measurements should be quantitatively assessed against synthetic sampling of the model output at analogous locations, i.e. sampling sites, following analogous perturbations. However, it is not yet obvious which parameters and what degree of validation should be considered satisfactory. The expectations range from the replication of precise near-field measurements of alkalinity following a perturbation (which may require very high resolution models) to the validation of simulated alkalinity transport, dilution and mixing against observations of natural or artificial tracers<sup>113,118</sup>, to the recreation of more general relationships between the size of the perturbation and the size of the response measured at variable times and distances from the injection site. Ultimately, if a model can adequately replicate the observed OAE response *and* includes all relevant processes that could influence it (See **Supporting Note 1**), then it may be a trustworthy tool for carbon accounting. This can help disentangle rapidly diluting alkalinity from processes that reduce the total amount of additional alkalinity added to the system.

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