Supplement to: "Efficiency metrics for ocean alkalinity enhancement under responsive and prescribed atmosphere conditions"

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1 Analytical equality of $\eta(t)$ and direct atmosphere removal

Consider a simple box model comprising an atmosphere box of finite volume, and an ocean mixed layer box, as shown in Figure S1. The intent is not to create a model with detailed realism but to demonstrate the consequences of modelling the atmosphere to be finite, and therefore change its pCO_2 in response to fluxes of CO_2 across the air-sea interface.

Firstly, the flux of CO_2 across the air-water interface is typically modelled as proportional to the partial pressure difference

$$F_{CO_2} = k_w\alpha(pCO_2^{atm} - pCO_2^{ocn}) = k_w\alpha\Delta pCO_2, \tag{1}$$

where α is the solubility of CO_2 in seawater [≈ 34 mol/m^3/atm] and k_w is the gas transfer velocity [m/s]. Thus, the total amount of dissolved inorganic carbon (DIC) in the mixed layer and the atmosphere respectively change according to

$$\frac{dC^{ocn}}{dt} = -\frac{dC^{atm}}{dt} = F_{CO_2}A = k_w\alpha A\Delta pCO_2 = \Psi\Delta pCO_2 \tag{2}$$

where A is the surface area over which gas exchange occurs. For later convenience, Ψ is defined here as $k_w\alpha A$. The atmospheric partial pressure pCO_2^{atm} is directly proportional to the total amount of CO_2 in the atmosphere C^{atm} in [mol]

$$pCO_2^{atm} = V_m p\frac{C^{atm}}{V^{atm}}, \tag{3}$$

where V_m is the molar volume of gas (0.024 m^3/mol) and p is the atmospheric pressure. Or as a differential:

$$\frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}C^{atm}} = \frac{V_m p}{V_{atm}} \tag{4}$$

In the ocean (specifically in the mixed layer, "ml") the differential relationship between pCO_2 and C, for small changes in either, is

$$\frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}} = \frac{1}{\alpha V_{ml}}\frac{\partial[CO_2]}{\partial[DIC]} \tag{5}$$

Prescribed atmosphere

V_{atm} is **infinite**,

pCO_2 is **constant**

V_{ml} is finite,
pCO_2 is variable

$\Delta pCO_2(t) = \Delta pCO_2(0)\exp(-t\tau_c^{-1})$

$\Delta C(t) = \Delta C(0)\exp(-t\tau_c^{-1})$

Responsive atmosphere

V_{atm} is finite,

pCO_2 is variable

V_{ml} is finite,
pCO_2 is variable

$\Delta pCO_2(t) = \Delta pCO_2(0)exp(-t\tau^{-1})$

$\Delta C(t) = \Delta C(0)\left(\dfrac{\gamma}{1+\gamma} + \dfrac{1}{1+\gamma}\exp(-t\tau^{-1})\right)$
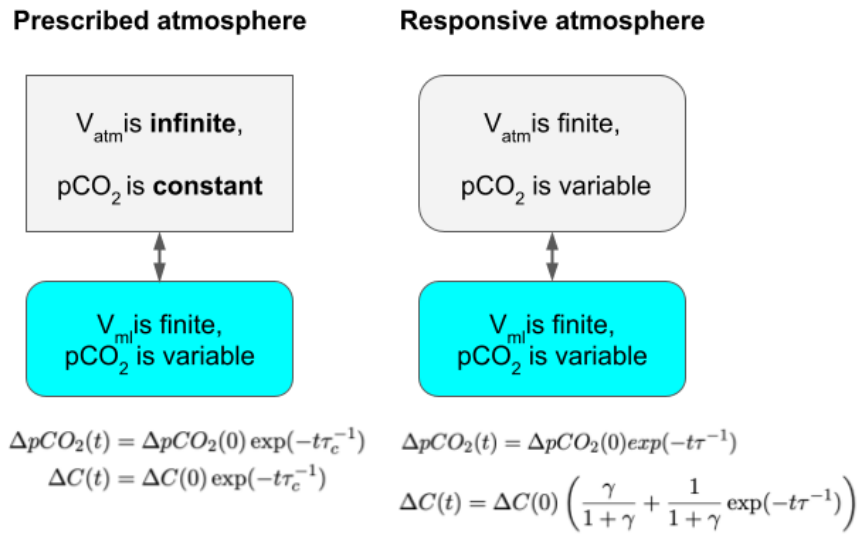
Figure S1: Two simple box models, one with a static, prescribed atmosphere and one with a reactive finite-volume atmosphere. The resulting decay laws for an initial pulse of CO_2 added to to ocean ($\Delta C(0)$) and and initial pulse of $pCO_2(0)$ are given below.

where [DIC] is the concentration of total dissolved carbon and α is the solubility of CO_2 in seawater (Zeebe and Wolf-Gladrow, 2001). This relation follows from the following differential relationships $\mathrm{d}[DIC] = \mathrm{d}C/V_{ml}$ and $\mathrm{d}pCO_2 = \frac{1}{\alpha}\mathrm{d}[CO_2]$.

Now the changes in partial pressure in the atmosphere and ocean mixed layer can be written using the chain rule ($\frac{\mathrm{d}pCO_2}{\mathrm{d}t} = \frac{\mathrm{d}C}{\mathrm{d}t} \cdot \frac{\mathrm{d}pCO_2}{\mathrm{d}C}$) as:

$$\frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}t} = -\Psi \Delta pCO_2 \frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}C^{atm}} \tag{6}$$

$$\frac{\mathrm{d}pCO_2^{oce}}{\mathrm{d}t} = \Psi \Delta pCO_2 \frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}} \tag{7}$$

The partial pressure difference between the two reservoirs thus evolves as

$$\frac{\mathrm{d}\Delta pCO_2}{\mathrm{d}t} = \frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}t} - \frac{\mathrm{d}pCO_2^{oce}}{\mathrm{d}t} = -\Psi \left[\frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}C^{atm}} + \frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}} \right] \Delta pCO_2 \tag{8}$$

$$= -\tau^{-1}\Delta pCO_2 \tag{9}$$

This ODE in ΔpCO_2 is readily solved - any partial pressure differences evolve in an exponential fashion

$$\Delta pCO_2(t) = \Delta pCO_2(0) exp(-t\tau^{-1}) \tag{10}$$

with a characteristic timescale

$$\tau = \Psi^{-1} \left(\frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}C^{atm}} + \frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}} \right)^{-1} \tag{11}$$

Note that the frequently taken assumption that atmospheric pCO_2 is constant, corresponds to the assumption that $V_{atm} = \infty$, in which case $\frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}C^{atm}} = 0$, i.e. it assumes an atmosphere which can accommodate arbitrary movement of CO_2 in or out of it, without a change in pCO_2^{atm}. In this case the e-folding time simplifies to

$$\tau_c = \Psi^{-1} \left(\frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}} \right)^{-1} \tag{12}$$

denoted here as τ_c for "constant". It is worth noting that the timescale obtained under constant atmosphere assumptions τ_c is always a little larger than τ due to the missing $\frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}C^{atm}}$ term. The ODE for the pCO_2 difference simplifies to the often used gas-exchange expression (Zeebe and Wolf-Gladrow, 2001) after substitution of Ψ and $\frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}}$:

$$\frac{\mathrm{d}\Delta pCO_2}{\mathrm{d}t} = -\frac{k_w}{z_{ml}} \frac{\partial[CO_2]}{\partial[DIC]} \Delta pCO_2 = -\tau_c^{-1}\Delta pCO_2 \tag{13}$$

where the depth of the mixed layer $z_{ml} = V_{ml}/A$.

1.1 Differences between reference and perturbed simulations

In general, the state of the mixed layer will not be in equilibrium with the atmosphere. Some areas may be taking in CO_2, some may be outgassing. The pCO_2^{atm} may be changing. However the effects of a small perturbation can be studied by taking the difference between a reference simulation and an otherwise identical simulation that has a perturbation applied to it.

Since the ODEs describing changes in ΔpCO_2 in both simulations (let's say A and B) are linear, the same kinetics also describe any differences between two simulations, say, ΔpCO_2^A and ΔpCO_2^B:

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\Delta pCO_2^A - \Delta pCO_2^B\right) = \frac{\mathrm{d}\Delta pCO_2^A}{\mathrm{d}t} - \frac{\mathrm{d}\Delta pCO_2^B}{\mathrm{d}t} = -\tau^{-1}(\Delta pCO_2^A - \Delta pCO_2^B) \qquad (14)$$

Therefore, from here on I assume that ΔpCO_2 and ΔC refer to the difference between a perturbed simulation and a reference simulation, for example:

$$\Delta pCO_2 = (\Delta pCO_2^A - \Delta pCO_2^B) = (\Delta pCO_2^{atm,A} - \Delta pCO_2^{ml,A}) - (\Delta pCO_2^{atm,B} - \Delta pCO_2^{ml,B}) \quad (15)$$

1.2 Equilibration of CO_2 inventories under reactive atmospheres

How does a small CO_2 deficit of size $\Delta C(0)$ induced in the ocean reservoir evolve in time under finite, responsive atmospheres ? If the perturbation is small then an initial perturbation $\Delta C(0)$ causes a initial change in pCO_2^{ocn} of

$$\Delta pCO_2^{ocn}(0) = \frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}}\Delta C(0) \qquad (16)$$

This initial partial pressure difference will then cause transfer of CO_2 from the atmosphere to the ocean. The ΔpCO_2 will equilibrate with the exponential law given by equation 10. The total rate of transfer of CO_2 between the atmosphere and the ocean (Equation 2) is proportional to ΔpCO_2, given by Equation 10:

$$\begin{aligned}
\frac{\mathrm{d}C}{\mathrm{d}t} &= \Psi\Delta pCO_2(t) \\
&= \Psi\Delta pCO_2^{ocn}(0)\exp(-t\tau^{-1}) \\
&= \Delta C(0)\Psi\frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}}\exp(-t\tau^{-1})
\end{aligned} \qquad (17)$$

The cumulative transfer of CO_2 is then expressed by the integral of the above rate. Subtracting this integral from the initial perturbation $\Delta C(0)$, the time evolution of the ocean inventory perturbation in the ocean is obtained:

$$\Delta C(t) = \Delta C(0) - \int_0^t \frac{\mathrm{d}C}{\mathrm{d}t}\mathrm{d}t \qquad (18)$$

Evaluating the integral

$$\begin{aligned}
\Delta C(t) &= \Delta C(0) - \Delta C(0)\Psi\frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}}\tau\left[1 - \exp(-t\tau^{-1})\right] \\
&= \Delta C(0) - \Delta C(0)\frac{\frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}}}{\frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}C^{atm}} + \frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}}}\left[1 - \exp(-t\tau^{-1})\right]
\end{aligned} \qquad (19)$$

where Ψ conveniently cancels. Simplifying yields:

$$\Delta C(t) = \Delta C(0) \left(\frac{\gamma}{1+\gamma} + \frac{1}{1+\gamma} \exp(-t\tau^{-1}) \right) \tag{20}$$

where γ expresses the ratio of the capacities of the two interacting reservoirs.

$$\gamma = \frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}C^{atm}} \bigg/ \frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}} = \frac{V_m p \alpha V_{ml}}{V_{atm}} \frac{\partial [DIC]}{\partial [CO_2]} \tag{21}$$

As expected, under finite, responsive atmosphere conditions, some fraction of the initial pulse will always remain in the reservoir it was induced in, even once the pCO_2 values have reached equilibrium. The fraction that will move to the other reservoir is $\frac{1}{1+\gamma}$, while the fraction that will remain in the ocean is $\frac{\gamma}{1+\gamma}$.

1.3 Equilibration of CO_2 inventories under prescribed atmospheres

The non-reactive atmosphere conditions, where the atmosphere is modelled with a constant or prescribed pCO_2^{atm}, correspond to the assumption that $V_{atm} = \infty$. In that case, $\gamma \to 0$ and $(1+\gamma)^{-1} \to 1$. Under these circumstances, the evolution of a $\Delta C(0)$ pulse is a simple exponential decay with the simpler e-folding time τ_c given by Equation **??** and that will decay all the way to zero.

$$\begin{aligned} \Delta pCO_2(t) &= \Delta pCO_2(0) \exp(-t\tau_c^{-1}) \\ \Delta C(t) &= \Delta C(0) \exp(-t\tau_c^{-1}) \\ \frac{\Delta \mathrm{d}C(t)}{\mathrm{d}t} &= -\Delta C(0)\tau_c^{-1} \exp(-t\tau_c^{-1}) \end{aligned} \tag{22}$$

This is basis of the $\eta(t)$ curve obtained in OAE efficiency simulations(Tyka et al., 2022; He and Tyka, 2023; Zhou et al., 2024; Suselj et al., 2024).

$$\eta(t) = 1 - \frac{\Delta C(t)}{\Delta C(0)} = 1 - \exp(-t\tau_c^{-1}) \tag{23}$$

1.4 Equivalence

We have seen that the function $\eta(t)$, determined under constant pCO_2^{atm} conditions, does not properly describe the actual movement of CO_2 from the atmosphere into the ocean under realistic conditions (where feedback of the atmosphere is taken into account). This raises the question, what does $\eta(t)$ represent, if anything ? To illuminate this question, consider the following scenario. How would the total inventory of CO_2 in the atmosphere evolve, under realistic reactive-atmosphere conditions, if CO_2 was directly removed from the atmosphere at a rate proportional to $d\eta(t)/dt$, using a terrestrial technology like DAC, instead of indirectly by applying an alkalinity pulse to the ocean?

Specifically, the rate of carbon removal out of the atmosphere would be

$$\frac{\mathrm{d}\Delta C}{\mathrm{d}t} = \Delta C(0) \frac{\mathrm{d}\eta(t)}{\mathrm{d}t} = \Delta C(0)\tau_c^{-1} \exp(-t\tau_c^{-1}) \tag{24}$$

where $\Delta C(0)$ is a scaling factor that sets the total, eventual amount of CO_2 removed. This steady removal of CO_2 from the atmosphere lowers the pCO_2 of the atmosphere at a rate of

$$\frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}t} = \frac{dpCO_2^{atm}}{dC}\Delta C(0)\,\tau_c^{-1}\exp(-t\tau_c^{-1}) \tag{25}$$

Since the atmosphere now develops a ΔpCO_2 deficit, relative to the ocean, the ocean would react and degas into to the atmosphere (remember all Δs are differences between a reference and a perturbed simulation). The rate of ΔpCO_2 change therefore would be given by the regular gas exchange ODE (Equation 9) which describes the ongoing ocean-atmosphere equilibration plus the bias induced by the CO_2 removal process (Equation 25).

$$\frac{\mathrm{d}\Delta pCO_2}{\mathrm{d}t} = -\tau^{-1}\Delta pCO_2 + \frac{dpCO_2^{atm}}{dC}\Delta C(0)\tau_c^{-1}\exp(-t\tau_c^{-1}) \tag{26}$$

This ODE is of the form $y'(t) = gy(t) - mh\exp(th)$ with the solution $y(t) = c_1\exp(gt) + \frac{hm}{g-h}\exp(ht)$. The initial condition for this linear ODE is $\Delta pCO_2(0) = 0$ since at the moment the removal starts, the reference and perturbed states are equal. This implies in the generalized form above $y(t) = 0$ and therefore the constant $c_1 = -\frac{hm}{g-h}$. Hence $y(t) = -\frac{hm}{g-h}\exp(gt) + \frac{hm}{g-h}\exp(ht) = \frac{hm}{g-h}(\exp(ht) - \exp(gt))$

Substituting the placeholders g, h and m:

$$\begin{aligned}
\Delta pCO_2^{atm}(t) &= \Delta C(0)\left(\frac{dpCO_2^{atm}}{dC^{atm}}\right)\frac{\tau_c^{-1}}{\tau^{-1} - \tau_c^{-1}}[\exp(-t\tau_c^{-1}) - \exp(-t\tau^{-1})] \\
&= \Delta C(0)\frac{\tau_c^{-1}}{\Psi}[\exp(-t\tau_c^{-1}) - \exp(-t\tau^{-1})]
\end{aligned} \tag{27}$$

Given this trajectory of ΔpCO_2, what is the combined effect on total atmospheric CO_2 inventory, i.e. the sum of direct removal and ocean reactance ? The total rate of change is the sum of the the direct CO_2 removal rate (Equation 24) and the rate due to gas exchange between ocean and atmosphere (due to the imbalance of ΔpCO_2):

$$\frac{\mathrm{d}\Delta C^{atm}}{\mathrm{d}t} = \Delta C(0)\tau_c^{-1}\exp(-t\tau_c^{-1}) - \Psi\Delta pCO_2(t) \tag{28}$$

Substituting the expression for $\Delta pCO_2^{atm}(t)$ from Equation 27:

$$\begin{aligned}
\frac{\mathrm{d}\Delta C^{atm}}{\mathrm{d}t} &= \Delta C(0)\tau_c^{-1}\exp(-t\tau_c^{-1}) - \Delta C(0)\tau_c^{-1}[\exp(-t\tau_c^{-1}) - \exp(-t\tau^{-1})] \\
&= \Delta C(0)\tau_c^{-1}\exp(-t\tau^{-1}) \\
&= \Delta C(0)\Psi\frac{dpCO_2^{ocn}}{dC^{ocn}}\exp(-t\tau^{-1})
\end{aligned} \tag{29}$$

This net rate of change in atmospheric carbon inventory equals precisely that obtained earlier when considering a a pulsed DIC deficit in the ocean, Equation (17). This result clarifies the exact meaning of the $\eta(t)$ OAE efficiency curve: The OAE curve of an *instantaneously* induced ocean-based DIC deficit (obtained under artificial, non-reactive conditions), gives the progression of the equivalent *gradual* removal of CO_2 directly from the atmosphere (under real, responsive atmosphere conditions), which would result in the same reduction in net atmospheric carbon inventory.

1.5 Calculating $\eta(t)$ from a coupled simulation

When simulating OAE under responsive atmospheres or earth system models, it is still useful to calculate $\eta(t)$, especially or purposes of comparing different negative emissions technologies and calculating negative emission credits. The directly measured change in the ocean inventory unfortunately conflates the effect of the OAE intervention and the subsequent feedback. Therefore a correction is necessary which can be used to factor out the effect of reservoir feedback. Schwinger et al. (2024) proposed a simple simulation setup to calculate this correction, which allows the conversion between the two carbon flows under the two different simulation conditions: First one simulates the fully coupled system with the OAE intervention. Then one conducts an additional reference simulation in which the atmosphere pCO_2 is prescribed at the exact trajectory obtained in the fully-coupled simulation with the OAE intervention. However, in the new reference simulation no OAE is performed - only the atmospheric pCO_2 is prescribed.

We can show below, using the the two-box model, that the difference in ocean inventory between these two simulations indeed equals the $\eta(t)$ value, had the OAE been simulated under prescribed atmosphere in the first place.

The rate of CO_2 transfer due to an OAE pulse under responsive atmosphere is Equation 17

$$\frac{\mathrm{d}C^{ocn}}{\mathrm{d}t} = \Delta C(0)\Psi \frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}} \exp(-t\tau^{-1}), \tag{30}$$

where $\Delta C(0)$ gives the initial DIC deficit. But under prescribed conditions the rate is given by Equation 22

$$\frac{\mathrm{d}\Delta C^{ocn}}{\mathrm{d}t} = -\Delta C(0)\tau_c^{-1}\exp(-t\tau_c^{-1}) \tag{31}$$

The correction required is equal to the difference of the two rates, i.e. the purely reactionary part of the carbon flux caused by the atmosphere feedback equals:

$$\frac{\mathrm{d}C^{Feedback}}{\mathrm{d}t} = \Delta C(0)\Psi \frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}} \left[\exp(-t\tau^{-1}) - \exp(-t\tau_c^{-1})\right] \tag{32}$$

Now consider the reference simulation with the pCO_2 prescribed at the trajectory from the OAE-perturbed coupled-simulation. In this new reference simulation, the difference in atmospheric pCO_2 compared to what it would have been without the prescription is equal to the change in pCO_2 caused by OAE in the fully-coupled simulation. That change is equal to the rate of CO_2 transfer caused by OAE (from Equation 20) times $\mathrm{d}pCO_2^{atm}/\mathrm{d}C^{atm}$, i.e.

$$\frac{\mathrm{d}\Delta pCO_2^{atm}}{\mathrm{d}t} = -\frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}C^{atm}}\frac{\mathrm{d}C^{ocn}}{\mathrm{d}t} = \Delta C(0)\Psi \frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}C^{atm}}\frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}}\exp(-t\tau^{-1}) \tag{33}$$

In this new reference simulation there will be offgassing of CO_2 from the ocean to the atmosphere (relative to a completely unperturbed simulation), because the prescribed atmospheric pCO_2 is lower than it would have been otherwise. The total air-sea difference ΔpCO_2 will then evolve according to the difference between rates of change of the atmosphere and the ocean:

$$\frac{\mathrm{d}\Delta pCO_2}{\mathrm{d}t} = -\tau_c^{-1}\Delta pCO_2 - \Delta C(0)\Psi \frac{\mathrm{d}pCO_2^{atm}}{\mathrm{d}C^{atm}}\frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}}\exp(-t\tau^{-1}) \tag{34}$$

Where the first term is the regular ocean gas-exchange law under prescribed conditions (Equation 13) and the second term is the additional atmospheric pCO_2 deviation due to the prescribed pCO_2

trajectory (Equation 33). This, once again, is a linear ODE of the form $y'(t) = gy(t) - m\exp(th)$ with the solution $y(t) = c_1 \exp(gt) + \frac{m}{g-h}\exp(ht)$. Again, the initial condition for this linear ODE is $\Delta pCO_2(0) = 0$ since up to the point where the atmospheric pCO_2 is prescribed, the new reference state is identical to the original one.

The solution is (after simplification):

$$\Delta pCO_2(t) = \Delta C(0)\frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}}\left[\exp(-t\tau^{-1}) - \exp(-t\tau_c^{-1})\right] \tag{35}$$

Which describes the time evolution of the pCO_2 difference across the air-sea interface in this new reference simulation. The rate of CO_2 transfer caused by this $\Delta pCO_2(t)$ is then obtained from the gas transfer law (Equation 2), i.e.

$$\frac{\mathrm{d}C^{ocn}}{\mathrm{d}t} = \Delta C(0)\Psi\frac{\mathrm{d}pCO_2^{ocn}}{\mathrm{d}C^{ocn}}\left[\exp(-t\tau^{-1}) - \exp(-t\tau_c^{-1})\right] \tag{36}$$

This matches exactly the sought after $\frac{\mathrm{d}C^{Feedback}}{\mathrm{d}t}$, given by Equation 32. In other words, as suggested by Schwinger et al. (2024), the difference in CO_2 ocean inventory between a simulation with OAE under a responsive atmosphere and one where the pCO_2^{atm} trajectory is prescribed to that of the former simulation but with no OAE applied yields the same metric (after normalization by ΔAlk) as given by $\eta(t)$.

1.6 Supplementary figures
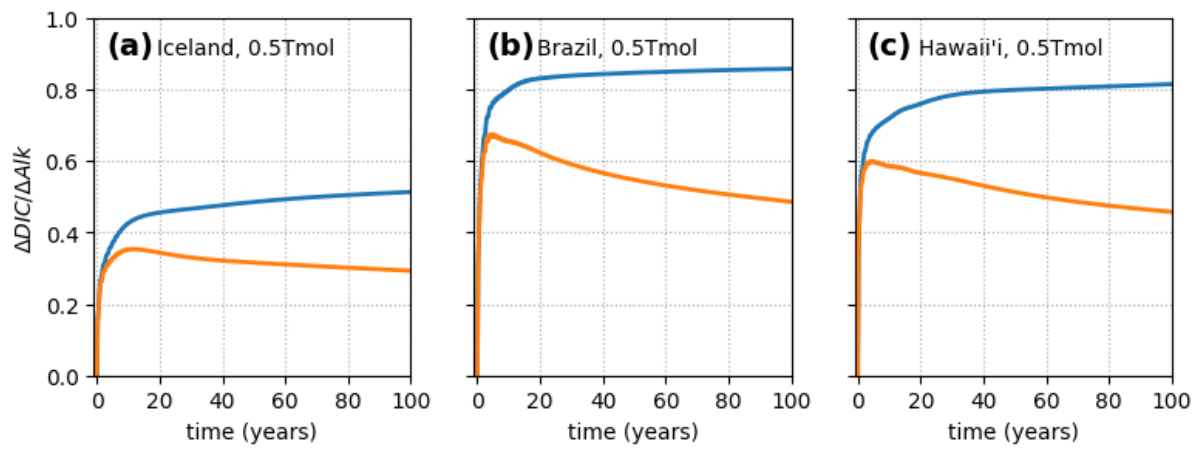
Figure S2: Increase in total ocean DIC following a 1-month pulse of alkalinity (ΔAlk) applied to the surface layer at a specific location, under a prescribed (non-reactive) atmosphere (blue) and a coupled (reactive) atmosphere respectively. The three different point-injection locations were off the coast of Brazil (2.50S,37.50W), near Iceland (61.50N,19.50W) and at Hawaii'i (19.02N,155.49E).

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