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***Dear Editorial Board of Global Biogeochemical Cycles,***

Please consider the manuscript entitled “*Quantifying Regional Efficiency of Marine Carbon Dioxide Removal (mCDR) via Alkalinity Enhancement using the ECCO-Darwin Ocean Biogeochemistry State Estimate and an Idealized Vertical 1-D Model*” for publication in *Global Biogeochemical Cycles*.

In this study we use data-assimilative numerical model simulations to address a crucial question regarding the estimation of efficiencies associated with ocean alkalinity enhancement (OAE) deployments in the open ocean across diverse regions. Recognizing the potential scalability of OAE to climate-relevant scales within a decade and its significant contribution to addressing climate change, our research is timely and impactful.

Our numerical simulations primarily use the global-ocean biogeochemistry state estimate, ECCO-Darwin. ECCO-Darwin incorporates both physical and biogeochemical observations to generate a data-constrained baseline ocean state, which is then subsequently perturbed by OAE deployments. We emphasize the advantage of ECCO-Darwin over traditional forward-only models for simulating OAE. We also introduce a reduced complexity model, *rapid-mCDR*, which capitalizes on computational efficiency to scale up our results while reasonably reproducing the ECCO-Darwin OAE experiments.

A critical aspect of our study involves the differentiation of OAE efficiency into two factors: CO<sub>2</sub> solubility and dynamical efficiency. We present a global map of the former and underscore the significant influence of ocean dynamics on the latter, demonstrating substantial variability among OAE injection location sites and seasons. This insight is imperative for the design of effective real-world OAE deployments. Furthermore, our research extends beyond theoretical frameworks, as we showcase the applicability of *rapid-mCDR* for numerous OAE deployments across the global ocean. Through this, we unravel the impact of different dynamical processes on the efficiencies of OAE, providing a comprehensive understanding of the underlying mechanisms.

In conclusion, our manuscript offers not only novel insights into the efficiency of OAE across the global ocean but also presents numerical tools crucial for addressing climate change that be readily employed by industry stakeholders and the broader community. We believe that our contribution will play a vital role in advancing real-world OAE deployments and contribute to the ongoing efforts to combat anthropogenic climate change.

Thank you for considering our manuscript. We look forward to the opportunity for publication in *Global Biogeochemical Cycles* and contributing to the scholarly discourse in the field.

Sincerely,  
Kay Suselj  
Jet Propulsion Laboratory

1           **Quantifying Regional Efficiency of Marine Carbon**  
2           **Dioxide Removal (mCDR) via Alkalinity Enhancement**  
3           **using the ECCO-Darwin Ocean Biogeochemistry State**  
4           **Estimate and an Idealized Vertical 1-D Model**

5           **Kay Suselj<sup>1</sup>, Dustin Carroll<sup>2,1</sup>, Dimitris Menemenlis<sup>1</sup>, Hong Zhang<sup>1</sup>, Nate**  
6           **Beatty<sup>3</sup>, Anna Savage<sup>3</sup>, Daniel Whitt<sup>4</sup>,**

7                   <sup>1</sup>Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, USA

8                   <sup>2</sup>Moss Landing Marine Laboratories, San José State University, Moss Landing, CA, USA

9                   <sup>3</sup>Running Tide, Inc., Portland, ME, USA

10                   <sup>4</sup>NASA Ames Research Center, Moffett Field, CA, USA

11           **Key Points:**

- 12           • We use a data-constrained ocean biogeochemistry model (ECCO-Darwin) to sim-  
13           ulate regional Ocean Alkalinity Enhancement (OAE).
- 14           • OAE efficiency hinges on intricate ocean dynamics, air-sea gas exchange, and sea-  
15           sonality — components to consider for OAE deployment design.
- 16           • For computationally-efficient OAE optimization and quantification, we introduce  
17           and evaluate the performance of a reduced-complexity model.

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Corresponding author: Kay Suselj, [kay.suselj@jpl.nasa.gov](mailto:kay.suselj@jpl.nasa.gov)

**Abstract**

As a marine Carbon Dioxide Removal (mCDR) approach, Ocean Alkalinity Enhancement (OAE) is emerging as a viable method for removing anthropogenic CO<sub>2</sub> emissions from the atmosphere to mitigate climate change. To achieve substantial carbon reduction using this method, OAE would need to be widespread and scaled-up across the global ocean. However, the efficiency of OAE varies substantially across a range of space-time scales and as such field deployments must be carefully planned to maximize efficiency and minimize logistical costs and risks. Here we develop a mCDR efficiency framework based on the data-assimilative ECCO-Darwin ocean biogeochemistry model, which examines two key factors over seasonal to multi-decadal timescales: 1) mCDR potential, which quantifies the CO<sub>2</sub> solubility of the upper ocean; and 2) dynamical mCDR efficiency, representing the full-depth impact of ocean advection, mixing, and air-sea CO<sub>2</sub> exchange. To isolate and quantify the factors that determine dynamical efficiency, we develop a reduced complexity 1-D model, rapid-mCDR, as a computationally-efficient tool for evaluation of mCDR efficiency. Combining the rapid-mCDR model with ECCO-Darwin allows for rapid characterization of OAE efficiency at any location globally. This research contributes to our understanding and optimization of OAE deployments (i.e., deploying experiments in the real-world ocean) as an effective mCDR strategy and elucidates the regional differences and mechanistic processes that impact mCDR efficiency. The modeling tools developed in this study can be readily employed by research teams and industry to plan and complement future field deployments and provide essential Monitoring, Reporting, and Verification (MRV).

**Plain Language Summary**

In an effort to counteract ongoing climate warming, engineering methods have been proposed to add materials to the ocean that increase the buffering capacity of seawater to sequester atmospheric carbon dioxide into the ocean — this is called Ocean Alkalinity Enhancement or OAE. However, expanding these pilot efforts to the scale where they would have a substantial impact on anthropogenic carbon emissions is a costly and challenging human endeavor. In order to simulate OAE deployments and guide future experiments and field trials, we used a state-of-the-art ocean model (ECCO-Darwin), which includes ocean carbon and biogeochemistry and is adjusted to be consistent with observations. We use both ECCO-Darwin and a reduced complexity model, which we develop

51 in this work, to show how and why the efficiency of OAE varies across ocean basins. Our  
52 modeling tools can be used by researchers and companies to better guide future OAE  
53 experiments in the real ocean.

## 54 **1 Introduction**

55 The major aim of the Paris Agreement is to reduce emissions and enhance carbon  
56 sinks that will keep the global temperature increase well below 2 degrees in this century  
57 (Rogelj et al., 2018; Schimel & Carroll, 2024). This limit requires a 50% reduction in an-  
58 thropogenic carbon dioxide ( $\text{CO}_2$ ) emissions by 2030, with emissions nearly eliminated  
59 by 2050 (i.e., net zero). This will require almost complete decarbonization of the world's  
60 energy supply (Friedlingstein et al., 2022; Palter et al., 2023). Furthermore, the IPCC's  
61 6th assessment report has emphasized that atmospheric  $\text{CO}_2$  removal on the gigaton scale  
62 will be necessary to reach net zero emissions (IPCC, 2022).

63 Due to the vast carbon reservoir size of the global ocean and its well-understood  
64 sink of anthropogenic  $\text{CO}_2$  emissions (Gruber et al., 2019; Friedlingstein et al., 2022),  
65 various methods for marine carbon dioxide removal (mCDR) strategies have been pro-  
66 posed (National Academies of Sciences, Engineering, and Medicine, 2022). Ocean Al-  
67 kalinity Enhancement (OAE; Renforth & Henderson, 2017) is a method designed to bol-  
68 ster the natural  $\text{CO}_2$  absorption of the ocean. Examples of particular OAE approaches  
69 include: 1) removal of acidity through electrochemical processes (House et al., 2007), 2)  
70 deliberate deployment of alkaline substances on the surface ocean, and 3) enhanced weath-  
71 ering of alkaline minerals in the terrestrial environment or coastal zone (Taylor et al.,  
72 2016; Montserrat et al., 2017). Eisaman et al. (2023) provides a detailed technical re-  
73 view of various OAE approaches.

74 The core principle of OAE leverages tight coupling between ocean alkalinity (Alk)  
75 and the nonlinear marine carbonate chemistry system (Middelburg et al., 2020). OAE  
76 is generally focused on the deployment of near-surface Alk, which transforms aqueous  
77 carbon dioxide ( $\text{CO}_2$ ) into bicarbonate ( $\text{HCO}_3^{2-}$ ) and carbonate ions ( $\text{CO}_3^{2-}$ ) ions through  
78 a series of rapid acid-base reactions (Zeebe & Wolf-Gladrow, 2001). This chemical ad-  
79 justment leads to a reduction in aqueous  $\text{CO}_2$  and thus lowers the partial pressure of car-  
80 bon dioxide ( $p\text{CO}_2$ ) in seawater. If the  $p\text{CO}_2$  reduction occurs in the near-surface ocean,  
81 it can induce disequilibrium with atmospheric  $p\text{CO}_2$  and drive ocean uptake of  $\text{CO}_2$  from

82 the atmosphere. This uptake acts to restore the ocean-atmosphere  $p\text{CO}_2$  gradient (i.e.,  
83 the disequilibrium which resulted from OAE deployment) back towards an equilibrium  
84 state.

85 OAE is well established as a conceptual mechanism in marine geoengineering. Fur-  
86 thermore, National Academies of Sciences, Engineering, and Medicine (2022) ocean-based  
87 CDR research strategy plan states that OAE efficacy is rated as “high confidence”, with  
88 durability and scalability as “medium-high”, yet the knowledge base remains “low-to-  
89 medium”. To permit widespread gigaton-scale OAE, it is critical to conduct field trials  
90 as these include aspects of ocean-atmosphere exchange, ocean-sediment exchange, bio-  
91 geochemical side effects and feedbacks, and ecological dynamics that cannot be replicated  
92 with lab experiments (Iglesias-Rodríguez et al., 2023). With the rapid growth of start-  
93 up companies involved in mCDR, which are starting to market  $\text{CO}_2$  removal to buyers  
94 interested in offsetting carbon emissions, there is an urgent need to develop numerical  
95 tools to 1) simulate, optimize, and quantify efficiency of mCDR approaches at various  
96 ocean locations before expensive and labor-intensive field tests are conducted, 2) per-  
97 mit standardized third-party Monitoring, Reporting, and Verification (MRV) of the as-  
98 sociated carbon capture, and 3) assess potential harmful impacts to aquatic and ocean  
99 ecosystems.

100 For typical ocean conditions, OAE has a potential to remove between 0.75 to 1 mole  
101 of  $\text{CO}_2$  from the atmosphere per mole of deployed Alk (Renforth & Henderson, 2017;  
102 Tyka et al., 2022). The actual amount, however, hinges on the complex interplay of ocean  
103 physics, thermodynamics, and biogeochemistry (Sarmiento & Gruber, 2006). While car-  
104 bonate chemistry reactions can be assumed to occur nearly instantaneously, reequilibra-  
105 tion of ocean  $p\text{CO}_2$  perturbations occur over annual-to-decadal timescales (Jones et al.,  
106 2014; He & Tyka, 2023). This reequilibration process takes place against the backdrop  
107 of multi-scale ocean dynamics, which influence the marine carbonate system state and  
108 can sequester disequilibrated waters away from the air-sea interface and deep into the  
109 ocean interior. Hence, as corroborated by prior numerical investigations (Ilyina et al.,  
110 2013; González et al., 2018; Burt et al., 2021; He & Tyka, 2023), the effectiveness of OAE  
111 is subject to considerable regional and temporal variability across the global ocean. How-  
112 ever, the details of how the three-dimensional ocean circulation, sea-ice, carbonate, and  
113 ecological state impact regional OAE experiments through space and time have yet to  
114 be quantified using data-constrained ocean simulations.

115 In this study, we assess the effectiveness of OAE as a mCDR approach across var-  
116 ious ocean regions representative of distinct dynamical and biogeochemical regimes. To  
117 achieve this, we utilize a new version of the ECCO-Darwin biogeochemistry model over  
118 the period from January 1995 to December 2017. The ECCO-Darwin model is partic-  
119 ularly well-suited for simulating OAE as it assimilates a suite of in-situ and remotely-  
120 sensed physical and biogeochemical observations (Carroll et al., 2020, 2022) and thus pro-  
121 vides the realistic background ocean state required for quantifying mCDR efficiency in  
122 the context of MRV (Köhler et al., 2013). As such, the ECCO-Darwin model accurately  
123 depicts the spatiotemporal evolution of historical ocean conditions, which typical climate  
124 models do not as they are not constrained by observations. Furthermore, unlike conven-  
125 tional assimilation techniques, the ECCO-Darwin data assimilation method avoids in-  
126 troducing non-physical source and sink terms (Carroll et al., 2022), making it an ideal  
127 tool for attributing OAE impacts on the time-dependent, three-dimensional ocean car-  
128 bon, biogeochemical, and ecological state.

129 To achieve a computationally-efficient assessment of mCDR CO<sub>2</sub> uptake efficiency,  
130 we develop a reduced-complexity, vertically-resolved 1-D model termed *rapid-mCDR*. Rapid-  
131 mCDR simulates the vertical transport of dissolved inorganic carbon (DIC) and Alk per-  
132 turbations and OAE additionality (i.e. net CO<sub>2</sub> uptake due to OAE). We demonstrate  
133 that rapid-mCDR can emulate key processes that affect mCDR additionality found in  
134 the higher-complexity 3-D ECCO-Darwin model. We use rapid-mCDR to expand our  
135 regional ECCO-Darwin analysis to basin-wide scales. Unlike ECCO-Darwin, which at  
136 present time needs to be run on a high-performance computing platform, rapid-mCDR  
137 can be easily run on a personal computer. We therefore propose that rapid-mCDR can  
138 serve as an alternative method for efficiently planning, characterizing, and optimizing  
139 field deployments, and thus can be used as the numerical foundation for MRV of mCDR  
140 deployments.

## 141 2 Methods

### 142 2.1 Quantification of OAE-driven Atmospheric CO<sub>2</sub> Removal

143 In this section, we summarize our approach for separating the impacts of dynam-  
144 ical and biogeochemical processes on OAE additionality (defined as the net CO<sub>2</sub> removed

Term/Symbol	Brief Explanation/Reference
Deployment site	Location of surface-ocean Alk injection.
OAE additionality	Net CO <sub>2</sub> removed from the atmosphere due to OAE (Section 2.1).
$mCDR_{pot}$	mCDR potential, i.e. maximum OAE additionality per unit of deployed Alk (Section 2.1).
$mCDR_{eff}$	Dynamical mCDR efficiency (Section 2.1).
$mCDR_{eff}^{cont}$	Dynamical mCDR efficiency for continuous OAE experiments (Equation 3).
$mCDR_{exch}$	Normalized net CO <sub>2</sub> flux (Supporting Information Text S2, Section 1).
$mCDR_{eff}^{sol}$	Solubility component of $mCDR_{eff}$ (Supporting Information Text S2, Section 3).
$mCDR_{equil}$	CO <sub>2</sub> Equilibration coefficient (Supporting Information Text S2, Section 4)
rapid-mCDR (Deploy)	Reduced-complexity rapid-mCDR model with input ocean conditions horizontally averaged over the deployment site (Section 4).
rapid-mCDR (HorAdv)	Rapid-mCDR model with input ocean conditions accounting for horizontal advection at the ocean surface (Section 4).

**Table 1.** List of key quantities used in this work.

145 from the atmosphere). Supporting Information Text S2–S4 describes these methods in  
 146 further detail and Table 1 provide a list of key terms and symbols used in this work.

147 When Alk is deployed into the surface ocean at location  $a_0$  with a time-dependent  
 148 prescribed rate (i.e., flux integrated over  $a_0$ ) of  $f_{Alk}(t)$ , the OAE-attributed CO<sub>2</sub> uptake  
 149 can be written as the integral function:

$$\Delta F_{CO_2}(\tau) = \int_{t_s}^{\tau} f_{Alk}(t) \times mCDR_{pot}(t, a_0) \times mCDR_{eff}(\tau - t, t, a_0) dt, \quad (1)$$

150 where  $\Delta F_{CO_2}$  represents the total OAE additionality by time  $\tau$  (in units of mol C), and  
 151  $mCDR_{pot}$  and  $mCDR_{eff}$  are mCDR potential and dynamic efficiency averaged over de-

152 deployment site area  $a_0$ . Times  $t_s$  and  $\tau$  denote, respectively, the start time of OAE de-  
 153 ployment and time when the OAE additionality is evaluated. The integration period spans  
 154 time  $t = t_s$  to  $t = \tau$ .

155 The mCDR potential,  $mCDR_{pot}(t, a_0)$ , represents the maximum possible ocean CO<sub>2</sub>  
 156 uptake per unit of Alk addition at time  $t$  and averaged over surface area  $a_0$ . It is com-  
 157 puted assuming complete reequilibration of ocean  $p\text{CO}_2$  and neglects any other feedbacks  
 158 except disassociation of aqueous CO<sub>2</sub> bicarbonate and carbonate ions. The dynamical  
 159 efficiency,  $mCDR_{eff}(\tau-t, t, a_0)$ , characterizes the fraction of mCDR potential that has  
 160 been realized by time  $\tau - t$  (for Alk deployed at time  $t$  over area  $a_0$ ) and is primarily  
 161 controlled by 3-D ocean dynamics and air-sea gas exchange.

162 Separation of OAE additionality into potential- and dynamical-efficiency compo-  
 163 nents provides a meaningful separation into drivers relating to CO<sub>2</sub> solubility and ocean  
 164 dynamics, respectively. This separation is accurate only when OAE additionality is lin-  
 165 ear with regards to the deployed Alk flux (i.e., the total amount of added Alk is small  
 166 enough that it does not substantially impact  $mCDR_{pot}$  and  $mCDR_{eff}$ ) and the deploy-  
 167 ment site area  $a_0$  is small enough that both  $mCDR_{pot}$  and  $mCDR_{eff}$  do not significantly  
 168 vary across it.

169 As shown in previous work,  $mCDR_{pot}$  values typically range between 0.75–1 mol  
 170 C per mol Alk.  $mCDR_{eff}$  is approximately an exponential function of elapsed time af-  
 171 ter OAE deployment with a characteristic multi-annual relaxation time, where over the  
 172 multi-decadal time scales  $mCDR_{eff}$  can reach values of up to one (e.g., He & Tyka, 2023).

173 For simplicity, in Equation 1  $mCDR_{pot}$  is defined as the average value over deploy-  
 174 ment site  $a_0$  at deployment time  $t$ , whereas its value over the area and time where ac-  
 175 tual net OAE CO<sub>2</sub> exchange occurs is the relevant mCDR potential. In our framework,  
 176 the difference between the two  $mCDR_{pot}$  values is reflected in a modification of  $mCDR_{eff}$ .  
 177 In Section 3.1, we show that  $mCDR_{pot}$  values vary gradually with time and distance from  
 178 the deployment site. Therefore, the impact of varying  $mCDR_{pot}$  on  $mCDR_{eff}$  are small.

179 Sections 2.1.1 and 2.1.2 describe methods for estimating  $mCDR_{pot}$  and  $mCDR_{eff}$   
 180 respectively, as they are the key factors that impact OAE additionality. If their global  
 181 values are known, Equation 1 provides a framework for characterizing the OAE addi-  
 182 tionality of any deployment in the global ocean with a known Alk rate  $f_{Alk}$ .

183 **2.1.1 Estimation of  $mCDR$  Potential,  $mCDR_{pot}$**

184  $mCDR_{pot}$  can be estimated diagnostically from surface-ocean thermodynamic and  
 185 carbonate conditions using a marine carbonate chemistry solver (Follows et al., 2006).  
 186 The results of the baseline/unperturbed ECCO-Darwin simulation, which are described  
 187 in Section 2.2.1, provide all of the necessary inputs and we use the *PyCO2SYS* (Humphreys  
 188 et al., 2022) Python toolbox for solving the marine carbonate chemistry system, as de-  
 189 tailed in Supporting Information Text S3.

190 To estimate the reliability of  $mCDR_{pot}$  from ECCO-Darwin, we also compute its  
 191 values using the OceanSODA-ETHZ product (Gregor & Gruber, 2021). While we do not  
 192 argue that  $mCDR_{pot}$  from any of the two datasets is superior, our comparison shows broad-  
 193 scale consistency, therefore lending support for the use of ECCO-Darwin to estimate  $mCDR_{pot}$ .

194 **2.1.2 Estimation of  $mCDR$  Dynamical Efficiency ( $mCDR_{eff}$ ): Pulse and**  
 195 **Continuous OAE Experiments**

196 At present time, the most obvious way of quantifying  $mCDR_{eff}$  is by running nu-  
 197 merical OAE experiments for locations and times of interest using global-ocean biogeo-  
 198 chemistry models, such as ECCO-Darwin. An approach discussed in He and Tyka (2023)  
 199 considers pulse experiments, where Alk is deployed over a short period (i.e., one month,  
 200 which is short compared to CO<sub>2</sub> reequilibration timescales). For a pulse experiment, in  
 201 which OAE is applied at time  $t$  and location  $a_0$ , the cumulative CO<sub>2</sub> flux at time  $\tau$  can  
 202 be expressed as:

$$\Delta F_{CO_2}(\tau) = \Delta Alk \times mCDR_{pot}(t, a_0) \times mCDR_{eff}(\tau - t, t, a_0) \quad (2)$$

203 Equation 2 can be derived from Equation 1, assuming that the Alk flux  $f_{Alk}$  fol-  
 204 lows a Dirac delta function with a spike at time  $t$  and a total Alk addition  $\Delta Alk$ .

205 For the pulse experiments,  $mCDR_{eff}$  is evaluated using Equation 2, where the cu-  
 206 mulative OAE additionality is estimated from the difference in simulated CO<sub>2</sub> flux be-  
 207 tween OAE and baseline/unperturbed simulations. This approach can be computationally  
 208 expensive if substantial regions of the global ocean over multiple deployment times  
 209 (e.g., different seasons) are considered, as for each location ( $a_0$ ) and deployment time  
 210 ( $t$ ) a new numerical experiment needs to be run. In fact, a single OAE simulation itself  
 211 is computationally expensive because it needs to represent the large ocean volume (e.g.,

212 global ocean) that is potentially affected by OAE over the duration of a decade or longer  
 213 — this is required to fully characterize the spread of OAE perturbations and long timescales  
 214 associated with  $mCDR_{eff}$ .

215 While we find pulse experiments useful, here we propose a modified approach us-  
 216 ing a continuous OAE deployment as our primary method for characterization of OAE  
 217 additionality. For a continuous OAE experiment starting at time  $t_s$  and with a constant  
 218 OAE flux  $f_{Alk}$ , the instantaneous flux of  $CO_2$ ,  $\Delta f_{CO_2}$  at time  $\tau$  ( $\tau > t_s$ ), can be ex-  
 219 pressed as (see Supporting Information Text S4 for details):

$$\Delta f_{CO_2}(\tau) = f_{Alk} \times \overline{mCDR_{pot}(a_0, t)}|_{t_s}^{\tau} \times mCDR_{eff}^{cont}(\tau - t_s, t_s, a_0), \quad (3)$$

220 where  $\overline{mCDR_{pot}(a_0, t)}|_{t_s}^{\tau}$  represents the time-mean value of  $mCDR_{pot}(a_0, t)$  from  $t_s$  to  
 221  $\tau$ .

222 Equation 3 provides a definition for  $mCDR_{eff}^{cont}$ , which is closely related to  $mCDR_{eff}$   
 223 as discussed below. Numerically,  $mCDR_{eff}^{cont}$  is estimated by differencing  $CO_2$  flux be-  
 224 tween the continuous OAE and baseline/unperturbed simulations, similar to how it is  
 225 done for pulse experiments except that we compare the instantaneous instead of cumu-  
 226 lative  $CO_2$  flux.

227 The dynamical efficiencies for pulse and continuous OAE experiments are closely  
 228 related (Supporting Information Text S4).  $mCDR_{eff}^{cont}$ , filtered with a high-frequency fil-  
 229 ter (for example, with an annual running-mean filter), represents time-mean values of  
 230  $mCDR_{eff}$ . Therefore, a single continuous OAE experiment can be used to characterize  
 231 the time-mean mCDR efficiency for a certain location. The continuous OAE experiments  
 232 also better characterizes overall OAE additionality of deployment sites.  $mCDR_{eff}^{cont}$  also  
 233 indicates variability of  $CO_2$  uptake on a shorter timescales (e.g., seasonal). Note that  
 234 the OAE-attributed  $CO_2$  flux is proportional to  $mCDR_{eff}^{cont}$ . If such variability is iden-  
 235 tified to be considerable, it is likely that  $mCDR_{eff}$  will be highly dependent on the time/season  
 236 of OAE deployment, which can be further investigated using targeted pulse experiments.

237 In summary, our numerical studies rely primarily on continuous OAE experiments,  
 238 which help us assess overall dynamical mCDR efficiency and pinpoint geographical ar-  
 239 eas with considerable short-term variability (e.g., seasonal). We find that mCDR effi-  
 240 ciency for regions with significant seasonal variability depends on the deployment time

and therefore requires supplementary pulse experiments to identify the dependence of  $mCDR_{eff}$  on the deployment time/season.

## 2.2 ECCO-Darwin Experiments for Quantification of mCDR Potential and Dynamical Efficiency

### 2.2.1 ECCO-Darwin Description

The ECCO-Darwin model and data assimilation methods have been extensively described in the literature (e.g., Brix et al., 2015; Manizza et al., 2019, 2023; Carroll et al., 2020, 2022; Bertin et al., 2023). In particular, a technical description of the ECCO-Darwin model set-up, observational constraints, and optimization methodology is presented in Carroll et al. (2020). In this study, we use a coarser-resolution ( $1^\circ$  vs  $1/3^\circ$  horizontal grid spacing) version of the Carroll et al. (2020) solution. Below, we provide a brief introduction to ECCO-Darwin and highlight the unique features of this model that are essential for our OAE studies.

The Lat-Lon-Cap-90 (LLC90) version of ECCO-Darwin used in this paper has  $1^\circ$  nominal horizontal grid spacing, spans 1992–2017, and is based on ocean circulation and physical tracers (i.e., temperature, salinity, and sea ice) from the Estimating the Circulation and Climate of the Ocean (ECCO) Version 4 release 4 solution (V4r4; ECCO Consortium, 2021; Forget et al., 2015). Horizontal grid spacing varies from 110 km at mid-latitudes to roughly 42 km at high latitudes. The vertical grid spacing increases from 10 m near the surface to 457 m near the seafloor. Since the horizontal discretization is insufficient to resolve mesoscale eddies, their impact on large-scale ocean circulation is parameterized using the Redi (1982) and Gent and McWilliams (1990) schemes; vertical mixing is parameterized with the Gaspar et al. (1990) scheme.

The ECCO V4r4 circulation estimate is used at each time step to drive an online biogeochemistry and ecosystem model developed by the Massachusetts Institute of Technology Darwin Project (Follows et al., 2007; Dutkiewicz et al., 2015, 2020). The Darwin model includes the cycling of organic and inorganic carbon, phosphorus, iron, silica, oxygen, and Alk. Carbonate chemistry is based on the efficient solver of Follows et al. (2006). Air-sea  $\text{CO}_2$  flux is computed using the parameterization of Wanninkhof (1992) and forced with atmospheric partial pressure of  $\text{CO}_2$  from the zonally-averaged National Oceanic and Atmospheric Administration Marine Boundary Layer Reference (NOAA MBL)

272 product (Andrews et al., 2014). The Darwin ecology includes five large-to-small phyto-  
273 plankton functional types (diatoms, other large eukaryotes, *Synechococcus*, and low- and  
274 high-light adapted *Prochlorococcus*), along with two zooplankton types that graze pref-  
275 erentially on either large eukaryotes or small picoplankton.

276 Physical observations are assimilated using the adjoint method (i.e., 4-D-Var; Wun-  
277 sch et al., 2009; Wunsch & Heimbach, 2013), which minimizes a weighted least squares  
278 sum of model-data misfit (i.e., a cost function) to optimize initial conditions, time-varying  
279 surface-ocean boundary conditions, and time-invariant, three-dimensional mixing coef-  
280 ficients for along-isopycnal, cross-isopycnal, and isopycnal thickness diffusivity. Because  
281 the initial conditions, surface boundary conditions, and mixing coefficients are estimated  
282 as part of the adjoint-method optimization, the ECCO ocean circulation estimate has  
283 negligible drift and therefore does not require spin-up. The biogeochemical model is op-  
284 timized in an additional step from the circulation using a low-dimensional Green’s Func-  
285 tions approach (Menemenlis et al., 2005) to assimilate a variety of biogeochemical ob-  
286 servations and adjust Darwin initial conditions and ecological parameters. We neglect  
287 the first 3 years of model simulation due to biogeochemical spin-up. The LLC90 ECCO-  
288 Darwin version closely matches the previously-published solution (Supporting Informa-  
289 tion Figure S2).

### 290 ***2.2.2 ECCO-Darwin OAE Experiments***

291 We first ran an unperturbed *baseline* simulation that represents the natural state  
292 of the ocean in the absence of any OAE perturbations. The time period used in this anal-  
293 ysis spans from January 1, 1995 to December 31, 2017. In Supporting Information Text  
294 S1, we show that the results of the *baseline* ECCO-Darwin simulation agree well with  
295 observations over the global surface ocean and in the various OAE deployment sites.

296 A suite of perturbed continuous and pulse OAE simulations were simulated, with  
297 Alk deployed at one of five deployment locations that are representative of diverse dy-  
298 namical and biogeochemical open-ocean regions. The deployment locations are discussed  
299 in Section 2.2.5 and summarized in Table 2. The perturbed simulations were compared  
300 against the baseline simulation to evaluate the impact of OAE on the biogeochemical ocean  
301 state and OAE additionality. For the perturbed simulations, we do not consider a par-

302 ticular OAE approach in terms of materials used and its dissolution rate, etc., but sim-  
 303 ply assume deployment of Alk at a prescribed rate.

### 304 **2.2.3 Continuous OAE Experiments**

305 For *continuous OAE experiments*, a constant Alk flux is applied to the ECCO-Darwin  
 306 surface-ocean layer (which is 10-m thick) over a regionally-defined deployment site from  
 307 January 1995 to December 2017. Continuous experiments are performed for all five de-  
 308 ployment locations described in Section 2.2.5. This allow us to estimate time-mean mCDR  
 309 efficiency and its variability, which might be the most relevant factors characterizing field  
 310 deployment locations.

311 For each of these experiments, an Alk rate (i.e., surface-integrated flux) of  $3.33 \times$   
 312  $10^7$  mol eq.  $s^{-1}$  is applied over a horizontal area of roughly  $270 \times 10^3$  km<sup>2</sup>. The amount  
 313 of deployed Alk is such that each experiment has a potential to remove  $10^{-2}$  Pg C yr<sup>-1</sup>  
 314 from the atmosphere, assuming 0.8 mol of CO<sub>2</sub> is removed for each mole of deployed Alk.  
 315 The molar ratio between removed CO<sub>2</sub> and added Alk is approximately valid for the global  
 316 ocean and assumes complete reequilibration of ocean  $p\text{CO}_2$

317 citeHe2023. Its exact value varies regionally as discussed in Section 3.1; we use the  
 318 ratio of 0.8 only as a rough estimate to contextualize the potential CO<sub>2</sub> uptake due to  
 319 OAE. We expect that the results will be fairly insensitive to the horizontal deployment  
 320 area, as long as it is large enough to avoid inorganic mineral precipitation.

321 We note that the magnitude of ocean CO<sub>2</sub> uptake, pH perturbations, and other  
 322 possible environmental impacts, which will be specific to the particular OAE approach  
 323 used (and might include inorganic mineral precipitation and impact on marine food web  
 324 via the introduction of micro-nutrients and trace metals), are expected to be strongly  
 325 correlated with the magnitude of the OAE Alk flux. In this work, we do not explore these  
 326 environmental impacts in depth as they are specific to the particular OAE approach. As  
 327 described in Section 2.1, our characterized OAE efficiency is normalized by Alk flux and  
 328 is not sensitive to its exact value.

### 329 **2.2.4 Pulse OAE Experiments**

330 For two deployment sites associated with strong seasonality in  $m\text{CDR}_{\text{eff}}^{\text{cont}}$  (North  
 331 Atlantic Subduction (NAS) and Antarctic Circumpolar Current (ACC), see Section 2.2.5

332 and Table 2 for more details), we performed three additional types of pulse experiments  
 333 with shorter Alk deployments. These experiments are used to further elucidate depen-  
 334 dence of mCDR efficiency to deployment season.

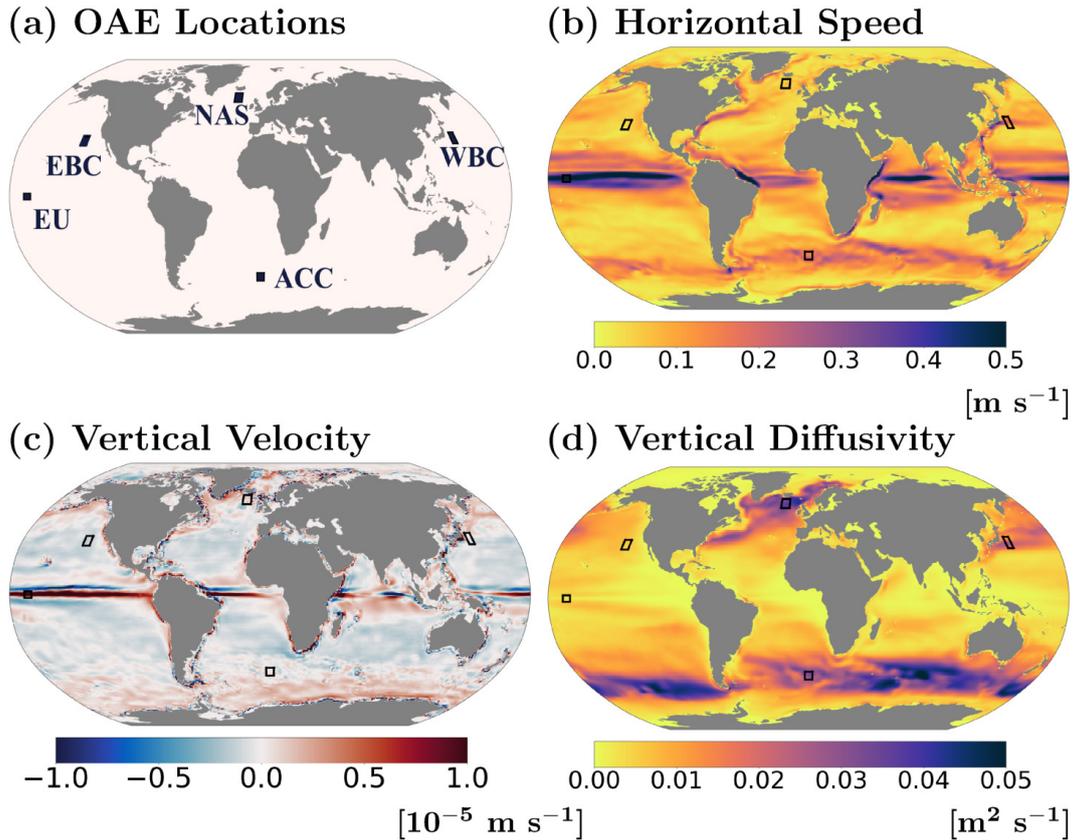
335 For two *monthly-pulse* experiments, OAE is applied for a duration of 31 days, start-  
 336 ing on January 1, 1995 and July 1, 1995; these monthly-pulse experiments are termed  
 337 *Jan1995* and *Jul1995* experiments, respectively. The monthly-pulse experiments start  
 338 during the months associated with nearly minimum and maximum  $mCDR_{eff}^{cont}$ . For these  
 339 experiments, the magnitude of the Alk flux is such that each pulse experiments has a  
 340 potential to remove  $10^{-2}$  Pg C from the atmosphere (with the same assumption of mo-  
 341 lar ratio between added Alk and removed CO<sub>2</sub> as before).

342 For the *yearly-pulse* OAE experiments, Alk is deployed during a single year (from  
 343 January 1st 1995 to December 31st 1995), where the magnitude of the Alk flux is equal  
 344 to that of the monthly pulse experiments and therefore the potential removed CO<sub>2</sub> from  
 345 the atmosphere is roughly 12 times larger compared to the monthly-pulse experiments.  
 346 We refer to these experiments as *Yr1995* experiments. While the duration of the Alk pulse  
 347 in the three pulse OAE experiments may appear long, we stress that its timescale is short  
 348 compared to typical multi-year to decadal CO<sub>2</sub> reequilibration timescales (He & Tyka,  
 349 2023).

### 350 **2.2.5 OAE Experiment Locations**

351 Figure 1 and Table 2 describe the chosen OAE experiment locations and associ-  
 352 ated surface-ocean conditions that are expected to impact mCDR efficiency in these re-  
 353 gions. The five experiments are representative of diverse dynamical and biogeochemi-  
 354 cal open-ocean conditions and include the following locations:

- 355 • *The North Atlantic Subduction (NAS)* experiment represents unique conditions  
 356 found in subpolar regions associated with subduction driven by sea-ice formation  
 357 and brine rejection, strong seasonally-driven vertical mixing, and seasonal biolog-  
 358 ical CO<sub>2</sub> uptake.
- 359 • *The Western Boundary Current (WBC)* experiment is representative of mid-latitude  
 360 conditions with strong horizontal currents and shear, along with intense vertical  
 361 mixing.



**Figure 1.** OAE experiments and time-mean values from January 1995 to December 2017 of ocean circulation fields from the baseline simulation. (a) Location of OAE experiments (black boxes show deployment sites), (b) magnitude of surface-ocean horizontal velocity, (c) mean vertical velocity over the upper-100 m, and (d) mean vertical diffusivity over the upper-100 m.

- 362
- *The Antarctic Circumpolar Current (ACC)* experiment represents conditions found

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    - in the Southern Ocean, which are associated with strong zonal currents, seasonal

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      - sea-ice cover, large-scale upwelling fronts, and seasonal biological uptake.

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    - *The Equatorial Upwelling (EU)* experiment is centered over the narrow upwelling

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      - zone of the Tropical Pacific Ocean and is characterized by strong CO<sub>2</sub> outgassing

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        - and seasonal biological uptake; its interannual variability tends to be dominated

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          - by El Niño–Southern Oscillation events (ENSO).

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      - *The Eastern Boundary Current (EBC)* experiment is centered over a region dom-

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        - inated by relatively-slow surface-ocean currents, coastal upwelling, and weaker pri-

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          - mary production.

Experiment Name	Abbreviation	Spatial Bounds	Inject/Impact Area $\times 10^3$ km <sup>2</sup>	Experiment Type	Potential Carbon Removed	$mCDR_{pot}$ : Mean/Range mol C/mol Alk
North Atlantic	NAS	16°W–23°W	270	continuous	$10^{-2}$ PgC yr <sup>-1</sup>	0.854
Subduction		56°N–62°N	7,734	monthly-pulse	$10^{-2}$ PgC	0.834–0.876
				yearly-pulse	$12 \times 10^{-2}$ PgC	
Western Boundary	WBC	144°E–148°E	267	continuous	$10^{-2}$ PgC yr <sup>-1</sup>	0.807
Current		31°N–38°N	14,155			0.792–0.826
Antarctic Circumpolar	ACC	3°E–3°W	268	continuous	$10^{-2}$ PgC yr <sup>-1</sup>	0.876
Current		45°S–50°S	13,033	monthly-pulse	$10^{-2}$ PgC	0.866–0.899
				yearly-pulse	$12 \times 10^{-2}$ PgC	
Equatorial Upwelling	EU	165°W–170°W	267	continuous	$10^{-2}$ PgC yr <sup>-1</sup>	0.799
		2°S–2°N	38,353			0.783–0.811
Eastern Boundary	EBC	130°W–135°W	266	continuous	$10^{-2}$ PgC yr <sup>-1</sup>	0.834
Current		30°N–35°N	7,811			0.822–0.844

**Table 2.** List of OAE experiments: experiment names and their abbreviations used in this work; regional bounds of OAE deployment site; surface area of deployment site (deployment area) and OAE-impacted area (impact area) for continuous experiments characterized as an area where OAE-attributed CO<sub>2</sub> flux exceeds 0.1 mol C m<sup>2</sup> by the end of simulation; experiment type with respect to OAE duration; potential of removed CO<sub>2</sub> assuming typical ocean conditions; and mean  $mCDR_{pot}$  over the deployment site from ECCO-Darwin (time-mean value over the simulation period and minimum and maximum monthly values, see Section 3.1 and Table 1 for definition of  $mCDR_{pot}$ ). OAE deployment site locations are shown in Figure 1.

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### 2.3 Rapid-mCDR: One-Dimensional Model for mCDR Simulation

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At present time, the ECCO-Darwin simulations discussed in the previous section might be computationally too expensive for simulating and quantifying  $mCDR_{eff}$  and therefore OAE additionality especially if a large number of OAE deployment sites and seasons are considered. Operational needs for quantification of real-world OAE might also require a more rapidly-deployable modeling framework.

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For these reasons, and as an alternative numerically-efficient approach, we develop a one-dimensional model *rapid-mCDR* consisting of the following three equations:

$$\frac{\partial}{\partial t} \Delta \widehat{Alk} = -\frac{\partial}{\partial z} \overline{w}^* \Delta \widehat{Alk} + \frac{\partial}{\partial z} (\overline{K}^* \frac{\partial}{\partial z} \Delta \widehat{Alk}) + \frac{\delta_{z_k,0}}{\Delta z_1} f_{Alk}, \quad (4)$$

$$\frac{\partial}{\partial t} \Delta \widehat{DIC} = -\frac{\partial}{\partial z} (\overline{w}^* \Delta \widehat{DIC}) + \frac{\partial}{\partial z} (\overline{K}^* \frac{\partial}{\partial z} \Delta \widehat{DIC}) - \frac{\delta_{z_k,0}}{\Delta z_1} \Delta f_{CO_2}, \text{ and} \quad (5)$$

$$\Delta f_{CO_2} = \overline{\kappa}^* (1 - \overline{a}_{ice}^*) \left( \frac{\partial \overline{pCO_2}^*}{\partial Alk} \Delta \widehat{Alk} + \frac{\partial \overline{pCO_2}^*}{\partial DIC} \Delta \widehat{DIC} \right), \quad (6)$$

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where variables  $w$  and  $K$  are 3-D vertical velocity and diffusivity,  $\kappa$  is the surface-ocean piston velocity, and  $a_{ice}$  is sea-ice cover. The  $\widehat{\varphi}$  and  $\overline{\varphi}^*$  represent horizontally-integrated values of  $\varphi$  over the global ocean and horizontal-mean values of rapid-mCDR forcing  $\varphi$  over the OAE-impacted area, respectively. The value of  $\delta_{z_k,0}$  is 1 for the uppermost rapid-mCDR vertical level and zero otherwise, and  $\Delta z_1$  represents the ocean depth represented by that layer.

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Equations 4 and 5 relate the time derivative of  $\Delta \widehat{Alk}$  and  $\Delta \widehat{DIC}$  (terms on the left hand side of these two equations) to its vertical advection and diffusion terms (first and second term on the right hand side of these equations) and prescribed Alk deployment rate or OAE-attributed  $CO_2$  uptake (the last terms in Equations 4 and 5, respectively). These two equations are derived by simplifying budget equations (Supporting Information Equations 14 and 15), guided by analysis of these budget terms for the five representative OAE experiments (Supporting Information Text S4). The following two approximations are used: 1) the biological source term was neglected and 2) we linearized the products of vertical velocity and Alk perturbations as:  $w \Delta \widehat{Alk} \approx \overline{w}^* \Delta \widehat{Alk}$  — this approximation neglects correlation between the vertical velocity and  $\Delta \widehat{Alk}$  over the OAE-

396 impacted regions. A similar approximation is made for DIC and diffusion terms in the  
 397 conservation equation.

398 Equation 6 represents the horizontally-integrated net CO<sub>2</sub> flux due to OAE (i.e.  
 399 OAE additionality) which is a function of ocean-surface perturbations  $\Delta\widehat{Alk}$  and  $\Delta\widehat{DIC}$ .

400 The rapid-mCDR equations are solved for 50 vertical levels (that coincide with the  
 401 ECCO-Darwin vertical levels) using a 1-day time step. The numerical finite difference  
 402 scheme uses an implicit Euler method for time derivatives, which ensures numerical sta-  
 403 bility. A simple numerical stability analysis and sensitivity study indicates that the daily  
 404 time step is sufficient (not shown). We assume that within the simulation time Alk and  
 405 DIC perturbations will not reach the lowest model level and therefore bottom-level  $\Delta\widehat{DIC}$   
 406 and  $\Delta\widehat{Alk}$  is set to zero throughout the simulation. We initialize rapid-mCDR at Jan-  
 407 uary 1, 1995 (before the start of OAE deployments), at which time the Alk and DIC per-  
 408 turbations are set to zero. The model is then integrated through the ECCO-Darwin pe-  
 409 riod (January 1st, 1995 to December 31st, 2017).

410 We note that there is some level of uncertainty in the method for horizontally av-  
 411 eraging the required inputs to rapid-mCDR (i.e. for computation of  $\bar{w}^*$ ,  $\bar{K}^*$ , and par-  
 412 tial derivatives of  $p\text{CO}_2$  in Equation 6). To address this uncertainty, we use two differ-  
 413 ent horizontal-averaging methods and quantify how they impact simulated  $m\text{CDR}_{eff}$ :

- 414 • *Deploy*: is the simplest approach where ocean conditions are horizontal means over  
 415 the deployment site. This approach neglects the horizontal advection of OAE-impacted  
 416 waters as they are transported away from the deployment site and into remote lo-  
 417 cations. Therefore, we expect the Deploy approach to be appropriate for OAE de-  
 418 ployment sites that are associated with relatively weak horizontal advection or ho-  
 419 mogeneous ocean regions. Rapid-mCDR results obtained with this averaging method  
 420 are referred as rapid-mCDR (Deploy).
- 421 • *HorAdv*: Improving on the Deploy approach, we also consider horizontal advec-  
 422 tion of OAE-impacted waters in a simplified manner. Ocean conditions are com-  
 423 puted as weighted means over the region where OAE modifies surface  $p\text{CO}_2$ ; here  
 424 the weights are proportional to the  $p\text{CO}_2$  perturbation  $\Delta p\text{CO}_2$ . We expect this ap-  
 425 proach to capture the impact of horizontal transport on Alk and DIC cycling and  
 426 CO<sub>2</sub> uptake, which the Deploy approach neglects. However, the HorAdv approach  
 427 assumes that surface-ocean perturbations are indicative of horizontal transport

through the entire water column. The results of rapid-mCDR using this method are termed rapid-mCDR (HorAdv).

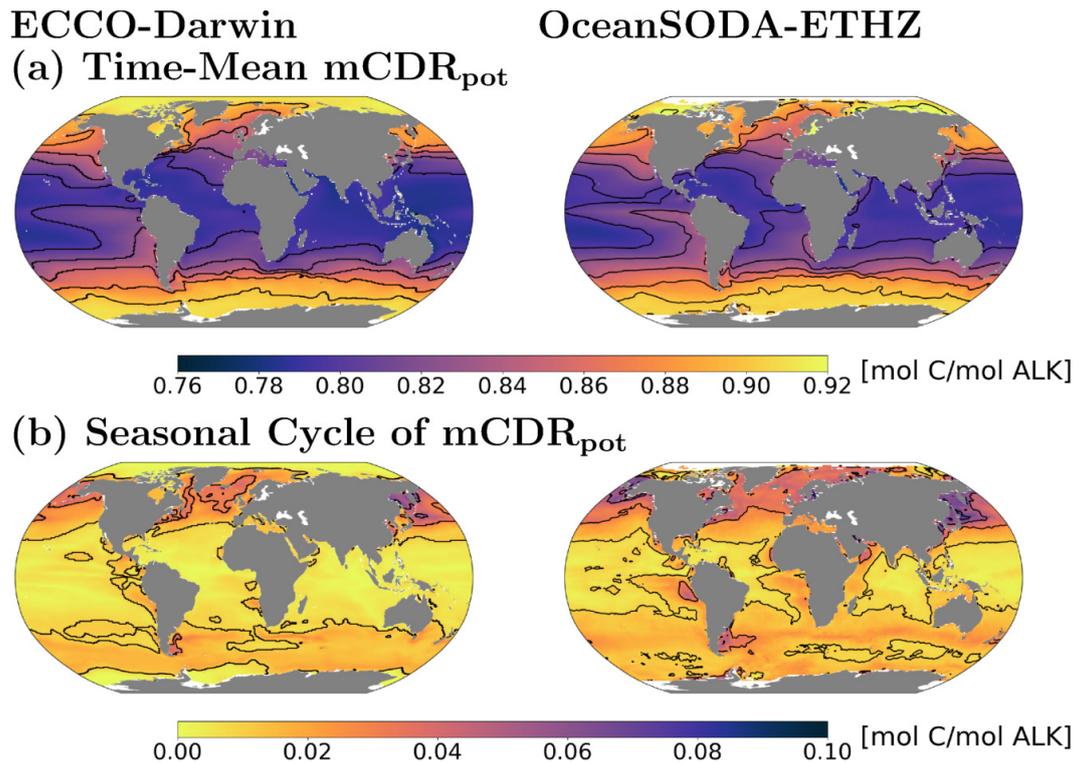
### 3 Results: ECCO-Darwin OAE Potential and Dynamical Efficiency

#### 3.1 OAE Potential

As described in Section 2.1.1 and Supporting Information Text S3, surface-ocean  $mCDR_{pot}$  is estimated diagnostically using *PyCO2SYS* from monthly-mean fields from 1) the baseline ECCO-Darwin simulation and 2) OceanSODA-ETHZ dataset (Gregor & Gruber, 2021), both covering the same time period from January 1995 to December 2017. The comparison of  $mCDR_{pot}$  from the two datasets provides a qualitative measure of its uncertainty. Figure 2 shows time-mean  $mCDR_{pot}$  for the global ocean and its climatological seasonal cycle from both datasets; Table 2 lists the ECCO-Darwin values for the 5 OAE deployment sites.

Time-mean  $mCDR_{pot}$  reveals a pronounced meridional gradient, with the lowest values (approximately 0.75 mol C/mol Alk located in the tropics, progressively increasing poleward and eventually exceeding 0.9 mol C/mol Alk). For the same latitudinal range, values in regions dominated by western boundary currents tend to be lower than eastern boundary currents, exhibiting an anti-correlation with SST (Supporting Information Figure S1). This structure is consistent with the expected increase in CO<sub>2</sub> solubility at lower SST. Time-mean  $mCDR_{pot}$  computed from both datasets exhibits similar features, with model-data differences generally not exceeding 0.025 mol C/mol Alk. One notable distinction is that OceanSODA-ETHZ values are marginally lower in eastern subtropical basins.

The climatological seasonal cycle of  $mCDR_{pot}$  remains modest, not surpassing 0.05 mol C/mol Alk in either ECCO-Darwin or OceanSODA-ETHZ (Figure 2b). The most pronounced seasonal cycle occurs in northern mid-latitudes and polar regions. Peak values are found in western boundary currents regions and are likely associated with elevated seasonality in SST (Jo et al., 2022). Although both ECCO-Darwin and OceanSODA-ETHZ exhibit similar seasonal patterns of  $mCDR_{pot}$ , the values from OceanSODA-ETHZ tend to have larger magnitudes. Across the five deployment sites, mean  $mCDR_{pot}$  ranges from 0.799 mol C/mol Alk (EU) to 0.854 mol C/mol Alk (NAS), while climatological seasonal cycle values do not exceed 0.05 mol C/mol Alk (Table 2). In summary, these



**Figure 2.**  $mCDR_{pot}$  from the baseline ECCO-Darwin simulation (left) and OceanSODA-ETHZ dataset (right) showing (a) time-mean values over the ECCO-Darwin period (January 1995 to December 2017) and (b) magnitude of climatological seasonal cycle.

459 findings suggest that, in the current climate,  $mCDR_{pot}$  is adequately represented by time-  
 460 mean values, except in localized regions that experience pronounced seasonality. Both  
 461 ECCO-Darwin and OceanSODA-ETHZ demonstrate similar spatial structure; hence in  
 462 the subsequent section we use only ECCO-Darwin .

463 In summary, as depicted in Figure 2, the most effective OAE deployments, solely  
 464 from a CO<sub>2</sub> solubility perspective, would be over polar oceans — regions characterized  
 465 by the highest potential for CO<sub>2</sub> removal. However, in the next sections, when we con-  
 466 sider ocean sea-ice and dynamical effects that are captured by the mCDR efficiency fac-  
 467 tor, this narrative will substantially change.

### 468 **3.2 OAE Impact on Ocean State for Continuous OAE experiments**

469 Before quantifying OAE efficiency, we first investigate the impact of OAE on the  
 470 ocean state for the five continuous experiments. Because of the larger Alk perturbation,  
 471 our continuous experiments serve as a upper limit of ocean state perturbations. These  
 472 results serve as an illustration of expected OAE impacts, because the environmental im-  
 473 pacts are expected to scale with the magnitude of injected Alk. We examine the spatial  
 474 patterns of atmospheric CO<sub>2</sub> uptake and alteration of surface-ocean pH. We also char-  
 475 acterize the vertical transport of OAE-impacted waters.

476 For the five continuous OAE experiments, Figure 3 shows a map of time integrated  
 477 net CO<sub>2</sub> flux due to OAE from the atmosphere into the ocean by the end of the ECCO-  
 478 Darwin simulation. The OAE additionality by the end of simulation can be obtained by  
 479 integrating these values over the surface ocean. For all OAE deployments, integrated net  
 480 CO<sub>2</sub> flux is largest close to the deployment site and its footprint is indicative of near-  
 481 surface horizontal advection, with the following key features:

- 482 • For NAS, the North Atlantic and the Norwegian Currents transport OAE-modified  
 483 waters towards high-latitude regions, with the flow bifurcating near Iceland. As  
 484 a result, the largest values of net CO<sub>2</sub> flux are found at, or north of the deploy-  
 485 ment site.
- 486 • For WBC and ACC, predominant zonal transport result in the largest net CO<sub>2</sub>  
 487 flux values located primarily east of the deployment site. In particular, the strong  
 488 Antarctic Circumpolar Current in ACC spreads net CO<sub>2</sub> flux eastward over a large  
 489 region of the Southern Ocean.

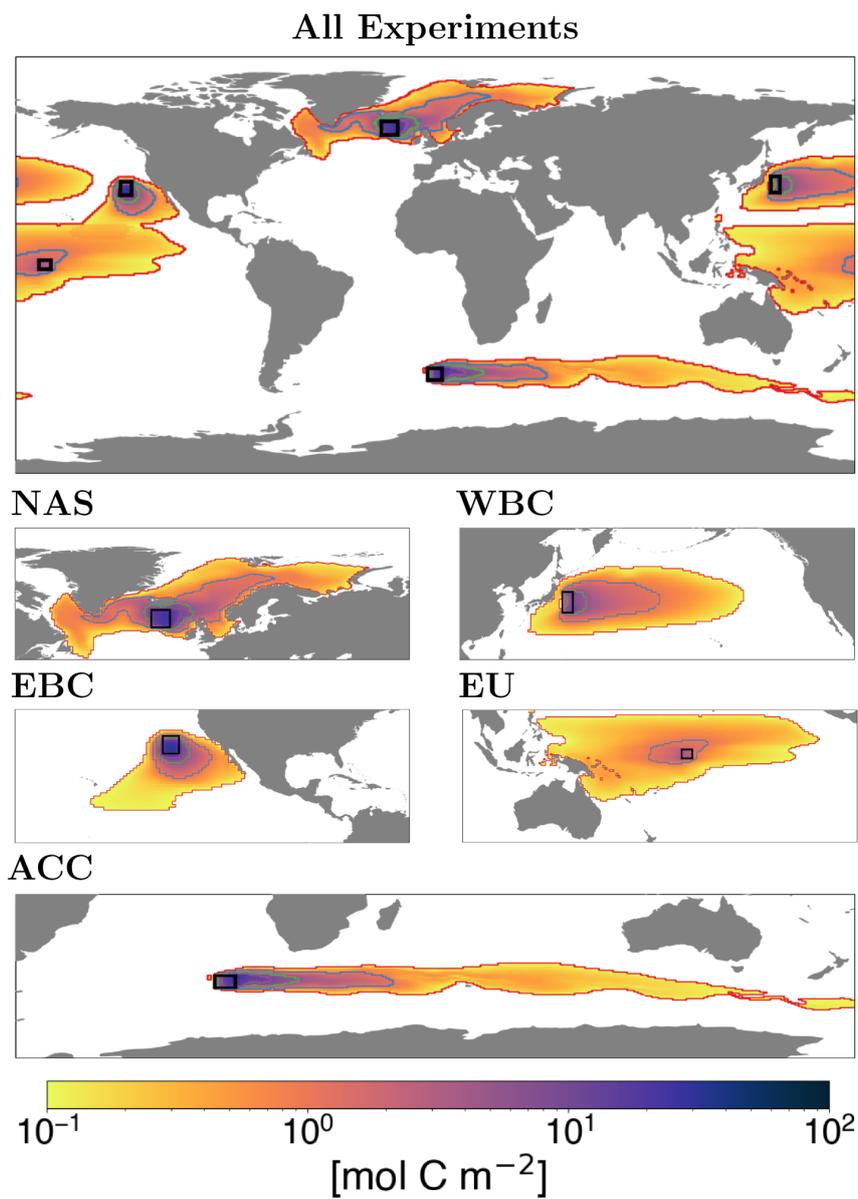
- 490 • For EU, equatorial upwelling and upper-ocean zonal flow both north and south  
491 of the equator spreads the net CO<sub>2</sub> flux footprint over most of the tropical/subtropical  
492 Pacific Ocean. The signature of cumulative net CO<sub>2</sub> flux for EU covers the largest  
493 horizontal area (see Table 2), while the maximum magnitude is the lowest of all  
494 5 experiments.
- 495 • For EBC, the anticyclonic circulation associated with the subtropical gyre advects  
496 OAE-impacted waters towards the southwest, spreading the net CO<sub>2</sub> flux foot-  
497 print west of Southern California and Baja Mexico.

498 One important concern regarding OAE is modification of ocean  $pH$ , which might  
499 inadvertently harm marine ecosystem health (e.g., Bach et al., 2019). Figure 4 shows the  
500 maximum  $pH$  modification due to OAE for all five continuous experiments. These spa-  
501 tial patterns are approximately correlated with spatial patterns of high net CO<sub>2</sub> flux.  
502 We note that none of the continuous experiments change OAE-attributed  $pH$  more than  
503 0.05 with respect to the background state, which is far less than what was considered  
504 a safe limit in He and Tyka (2023).

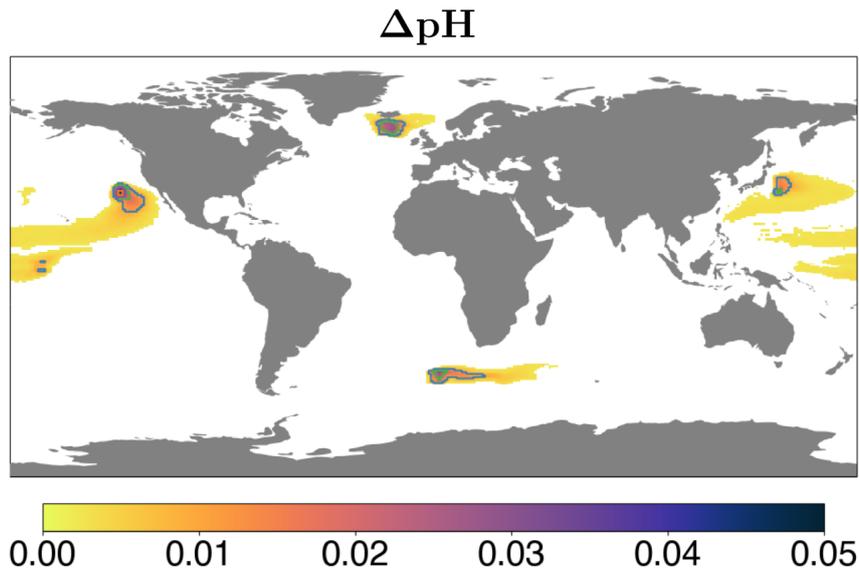
505 To illustrate the depth and temporal change in the disequilibrium of OAE-impacted  
506 waters across the five continuous OAE experiments, Figure 5a shows the temporal evo-  
507 lution of the depth above which 95% of the deployed Alk remains. We adopt this depth  
508 threshold as a metric to demarcate OAE-modified waters from those unaffected by OAE.  
509 Figure 5b shows time series of the horizontally-averaged equilibration coefficient  $mCDR_{equil}$   
510 for each of the experiments (mean over upper 100 m shown) and Figure 5c shows the  
511 spatial distribution of  $mCDR_{equil}$  at the end of the simulation.  $mCDR_{equil}$  (described in  
512 detail in Supporting Information Text S4, Section 3), is similar to  $mCDR_{eff}$  but is a lo-  
513 cal value computed for a particular volume of seawater in ECCO-Darwin.

514 For all OAE experiments, the OAE perturbation spreads to deeper waters with elapsed  
515 time after deployment, with large differences occurring in the depth of the OAE impact  
516 across all experiments. By the end of the simulation, the Alk perturbation reaches the  
517 depth in excess of 2000 m for NAS and roughly 1000 m for ACC. For the other three ex-  
518 periments, Alk perturbations remains much closer to the ocean surface and the ocean  
519 waters below 500 m remain largely unaffected.

520 The time evolution of  $mCDR_{equil}$  for all experiments exhibits an approximately ex-  
521 ponential increase, overlaid by strong seasonality (Figure 5b). The exponential increase



**Figure 3.** Time-integrated net CO<sub>2</sub> flux due to OAE from January 1995 to December 2017 for the 5 continuous OAE experiments. The color scale is logarithmic, highlighting variations in CO<sub>2</sub> uptake intensity and isolines represent DIC increases of 0.1, 1, 5, 10, and 50 mol C m<sup>-2</sup>. Black boxes show OAE deployment sites. Upper panel shows all 5 OAE experiments across the global ocean; lower panels show individual OAE experiments.

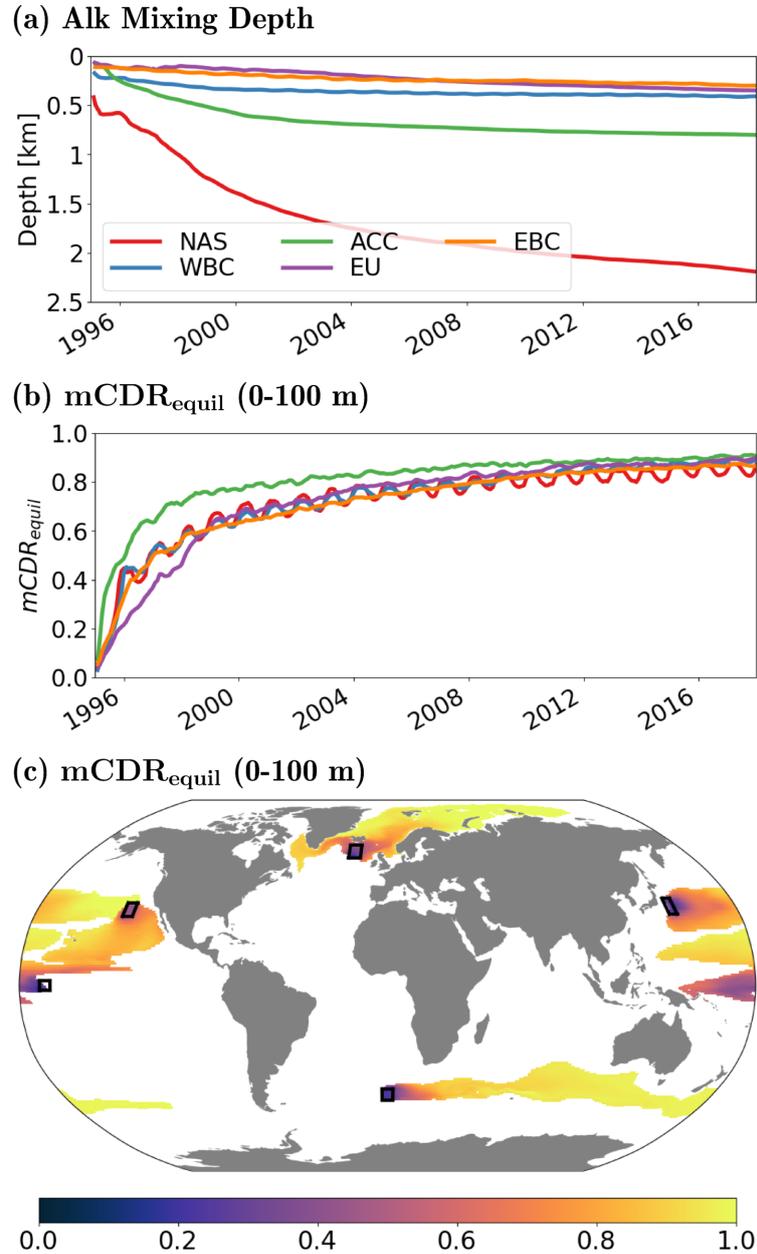


**Figure 4.** Maximum  $pH$  perturbation across time and depth (maximum  $\Delta pH$ ) due to OAE for the 5 continuous experiments. Only values above 0.0025 are shown. Isolines represent maximum  $\Delta pH$  values of 0.01, 0.02, 0.03, and 0.04.

522 is associated with the long time scale of  $CO_2$  reequilibration and the overlaid seasonal-  
 523 ity is associated with the strong seasonality of the mixed layer depth (MLD) and asso-  
 524 ciated transport of Alk-modified water below the MLD as the MLD shoals (detrainment),  
 525 and its re-entrainment as the MLD deepens.

526  $mCDR_{equil}$  tends to be highest for ACC and is associated with the shortest reequi-  
 527 libration time scale, despite the fact that the deployed Alk mixes relatively deep at this  
 528 site (Figure 5b). The short reequilibration time scale is consistent with the high  $CO_2$   
 529 piston velocity over this region, associated with strong zonal winds (e.g., Jones et al.,  
 530 2014). The seasonality of  $mCDR_{equil}$  is strongest at NAS and WBC; in particular, the  
 531 former is consistent with strong seasonality in MLD and vertical transport of OAE-attributed  
 532 Alk and DIC (Supporting Information Figures S3 and S4). Figure 5c shows that the low-  
 533 est values of  $mCDR_{equil}$  are found 1) close to the deployment sites where ocean-atmosphere  
 534  $CO_2$  reequilibration has not been fully realized and 2) increases as OAE-impacted wa-  
 535 ters spread into remote locations.

536 In Supporting Information Text S5, we discuss horizontally-integrated budgets for  
 537 DIC and Alk perturbations for the continuous OAE experiments. These budgets sep-  
 538 arate and quantify the contributions of key processes that modify DIC and Alk pertur-



**Figure 5.** Continuous OAE experiments: (a) time series of the depth that separates OAE-impacted waters from unmodified waters, (b) time series of  $mCDR_{equil}$  in the upper ocean (mean value over the upper 100 m), and (c) spatial pattern of  $mCDR_{equil}$  in the upper ocean (mean value over the upper 100 m) at the end of the simulation.

539 perturbations. With the exception of the air-sea interface, where the DIC and Alk perturba-  
 540 tions increase due to additional CO<sub>2</sub> flux from the atmosphere and the prescribed OAE  
 541 Alk flux, respectively, changes to DIC and Alk perturbations are predominantly dom-  
 542 inated by vertical ocean dynamics.

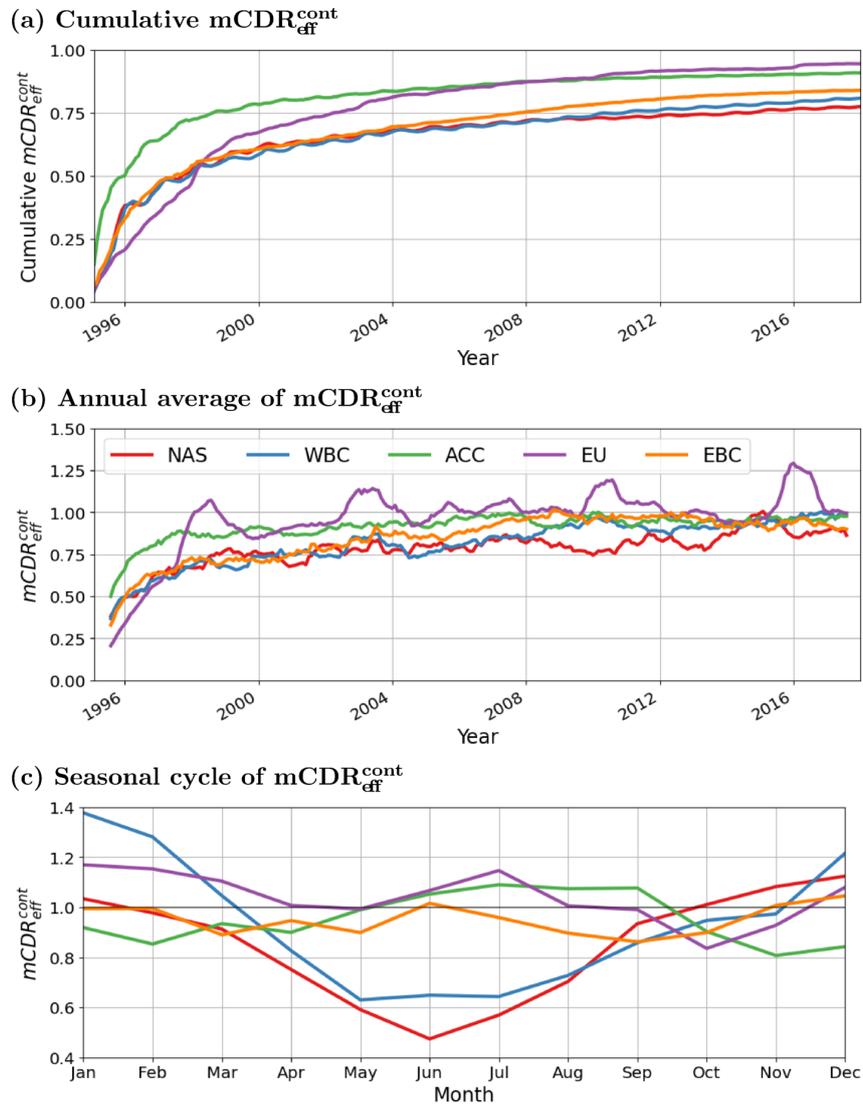
543 Across all deployment sites, excluding EU, a pronounced seasonality characterizes  
 544 the strength of vertical mixing and advection, coinciding with variations in MLD. As the  
 545 MLD deepens, DIC and Alk perturbations are transported into deeper waters, poten-  
 546 tially sequestering them from the atmosphere (which inhibits or delays OAE addition-  
 547 ality) until the MLD shoals again. Notably, the relative influence of vertical advection  
 548 and mixing varies significantly by deployment site. The depth of the MLD and its sea-  
 549 sonal variability differs substantially among sites, with NAS having the deepest seasonal  
 550 MLD.

### 551 3.3 Efficiency of Alk Enhancement

#### 552 3.3.1 Continuous OAE Experiments

553 For the five continuous OAE experiments, Figure 6 shows three key aspects of mCDR  
 554 efficiency  $mCDR_{eff}^{cont}$ . In panel (a), we show time-mean  $mCDR_{eff}^{cont}$  from the start of the  
 555 OAE deployment (i.e., January 1, 1995) to the time shown on the x-axis. These curves  
 556 represent the fraction of OAE potential realized by the time indicated on the x-axis; we  
 557 refer to this quantity as *realized mCDR potential*. The realized mCDR potential is thus  
 558 an important metric for quantifying the OAE additionality of continuous deployments.

559 Figure 6b shows  $mCDR_{eff}^{cont}$  filtered with a centered 1-year running mean. These  
 560 curves relate instantaneous Alk deployment to instantaneous CO<sub>2</sub> uptake, expressed us-  
 561 ing efficiency terms and with seasonal cycle removed. This is because  $mCDR_{eff}^{cont}$  and CO<sub>2</sub>  
 562 uptake are basically proportional to each other (Equation 3). For deployments associ-  
 563 ated with weak interannual variability in  $mCDR_{eff}^{cont}$ , we expect that these curves also  
 564 closely represent the mean efficiency of pulse deployments over a seasonal cycle. Figure  
 565 6c shows the seasonal cycle of  $mCDR_{eff}^{cont}$  over the last ten years of simulation. The de-  
 566 ployment locations associated with significant seasonal cycle indicate significant season-  
 567 ality of CO<sub>2</sub> uptake. For instantaneous OAE deployments, this is likely related to mCDR  
 568 efficiency being dependent on deployment season, which we further investigate in Sec-  
 569 tion 3.3.2.

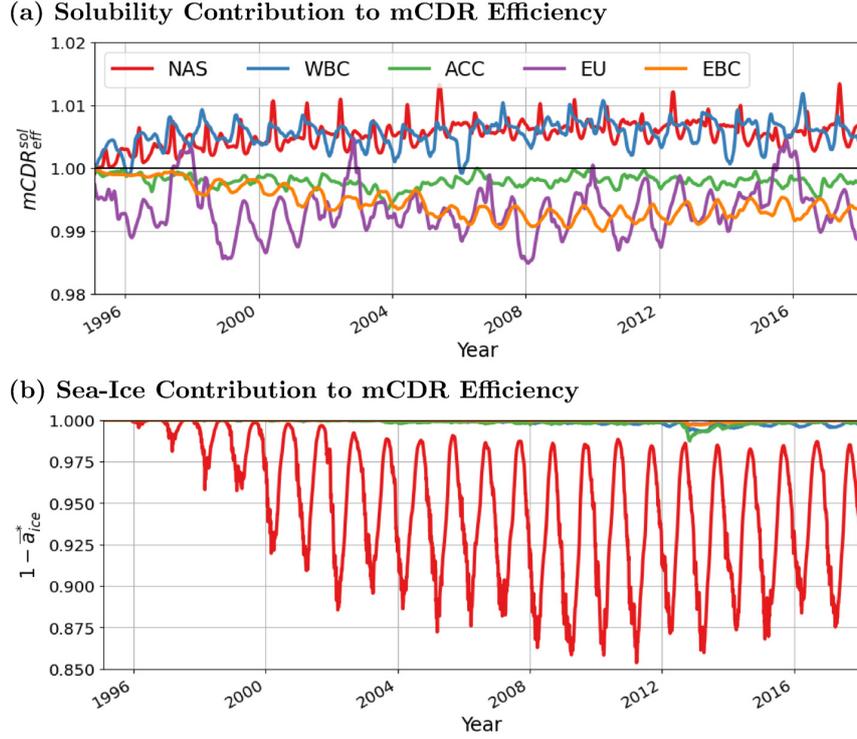


**Figure 6.** Three different key aspects of  $mCDR_{eff}^{cont}$  efficiency: (a) time-mean values from the start of the OAE deployment to time indicated on the x-axis; (b) centered 1-year running-mean values; and (c) seasonal cycle over the last simulation decade calculated as monthly-mean values over the last 10 years of simulation. The colors represent the five different OAE deployments, as indicated by the legend.

570 For all continuous experiments, the realized mCDR potential curves increase in an  
 571 approximately exponential manner with time after start of deployment (Fig. 6a), with  
 572 the following key characteristics:

- 573 • ACC is associated with the most rapid increase of realized mCDR potential, where  
 574 values exceed 0.75 within 5 years after the start of OAE and also reach one of the  
 575 highest values by the end of the simulation (0.91). This is consistent with the most  
 576 rapid increase of  $mCDR_{equil}$  in the upper ocean as shown in Figure 5.
- 577 • For all deployments, EU is associated with the slowest initial increase of realized  
 578 potential, consistent with the slowest increase of  $mCDR_{equil}$ . However, after roughly  
 579 13 years after deployment its values reach that of the ACC and by the end of the  
 580 simulation, EU reaches the highest value of all simulations, 0.95. The realized mCDR  
 581 potential for this location diverges from the exponential shape due to strong in-  
 582 terannual variability of  $mCDR_{eff}^{cont}$ , which we discuss below.
- 583 • For the other three experiments (EBC, WBC, and NAS), the realized mCDR po-  
 584 tential behaves remarkably similar over the first decade after the start of deploy-  
 585 ments and by the end of 2017 their values differ by only a few percent each (0.84,  
 586 0.81, and 0.77 for EBC, WBC, and NAS, respectively).

587 Figure 6b shows similar exponential behavior of  $mCDR_{eff}^{cont}$  as discussed for the re-  
 588 alized mCDR potential above, but it also reveals interannual variability in  $mCDR_{eff}^{cont}$ .  
 589 Note that for most of the experiments,  $mCDR_{eff}^{cont}$  is also associated with a strong sea-  
 590 sonal cycle which is filtered from the plots shown on Figure 6b. EU is associated with  
 591 the largest interannual variability of  $mCDR_{eff}^{cont}$ , superimposed on its exponential and sub-  
 592 sequent tapered increase. This interannual variability is positively correlated with the  
 593 multivariate El-Niño/Southern Oscillation (ENSO) index (Wolter & Timlin, 2011) (not  
 594 shown), demonstrating that ENSO has a substantial impact on mCDR efficiency in the  
 595 Tropical Pacific Ocean. We note that other locations also exhibit interannual variabil-  
 596 ity in  $mCDR_{eff}^{cont}$ , although it is lower than EU. The values of running mean  $mCDR_{eff}^{cont}$   
 597 can exceed one for a limited time period, which is most evident for the EU experiment.  
 598 This does not mean that OAE efficiency exceeds 100%, as the overall efficiency is related  
 599 to the time-mean  $mCDR_{eff}^{cont}$  values in Figure 6a and which do not exceed the value of  
 600 one.



**Figure 7.** Impact of CO<sub>2</sub> solubility and sea-ice cover on  $mCDR_{eff}^{cont}$ . The five colors represent the different OAE experiments, as shown in the legend.

601 Figure 6c shows that  $mCDR_{eff}^{cont}$  for NAS and WBC are associated with strong sea-  
 602 seasonality, yielding maximum values (and thus strongest ocean CO<sub>2</sub> uptake) in winter and  
 603 minimum values in summer. Peak monthly values are up to 40% lower/higher compared  
 604 to annual-mean values. In ACC, the magnitude of the seasonal cycle of  $mCDR_{eff}^{cont}$  is roughly  
 605 half that of NAS and is shifted in phase roughly 6 months due to its location in the south-  
 606 ern hemisphere. We emphasize that the seasonal cycle of  $mCDR_{eff}^{cont}$  is not directly re-  
 607 lated to the seasonal cycle of mCDR efficiency for the pulsed experiments. We discuss  
 608 the relations between the two seasonal cycles in the next section.

609 Next we discuss the impact of one of the key simplifications when estimating  $mCDR_{eff}^{cont}$ .  
 610 As described in Section 2.1.2, we defined  $mCDR_{eff}^{cont}$  with respect to  $mCDR_{pot}$  at the de-  
 611 ployment site and time (Equation 3); however, its values at all locations and times of  
 612 OAE-driven air-sea CO<sub>2</sub> exchange should be used. Figure 7a shows  $mCDR_{eff}^{sol}$ , which rep-  
 613 represents the multiplicative factor with which the  $mCDR_{eff}^{cont}$  should be corrected to account  
 614 for this simplification (Supporting Information Text S4).  $mCDR_{eff}^{sol}$  values greater/smaller  
 615 than one represent fractional increase/decrease of  $mCDR_{eff}^{cont}$  due to a change in  $mCDR_{pot}$

616 as the perturbation spreads from the deployment site to remote locations. This primar-  
 617 ily results from changes in CO<sub>2</sub> solubility due to cooling or warming of OAE-impacted  
 618 waters.

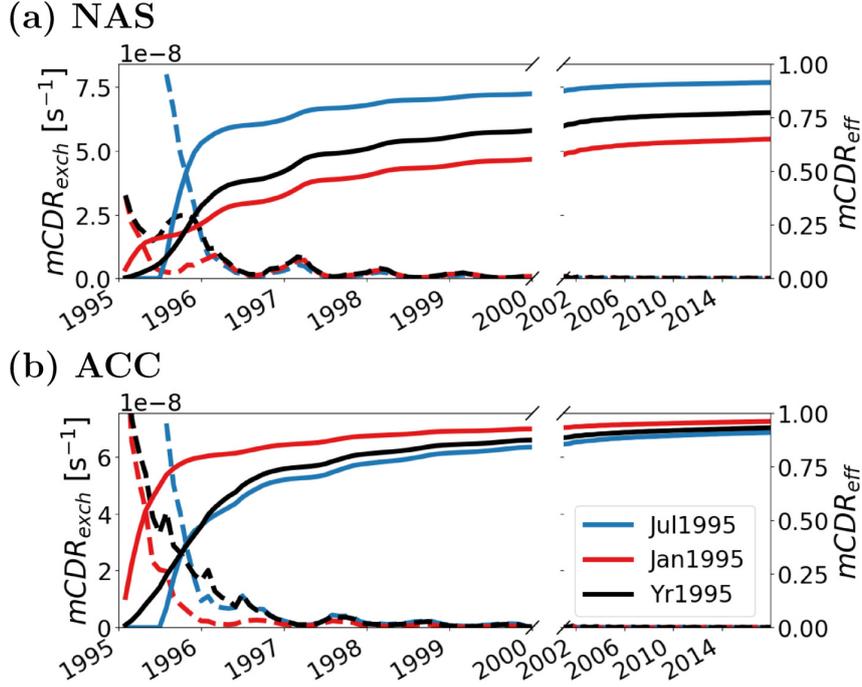
619 Figure 7a shows that the impact of the above-discussed simplification is small and  
 620 does not modify  $mCDR_{eff}^{cont}$  by more than 2% and rarely more than 1% for any of the  
 621 experiments. In NAS and WBC, northward and eastward flows, respectively, transport  
 622 Alk-enhanced waters to regions with lower SSTs and higher  $mCDR_{pot}$ , therefore the  $mCDR_{eff}^{sol}$   
 623 exceeds values of one. For the other three locations, horizontal transport results in a de-  
 624 crease of  $mCDR_{eff}^{cont}$ . In summary, we show that using the deployment site value of  $mCDR_{pot}$   
 625 is appropriate for the experiments described here. Figure 7b shows the impact of sea-  
 626 ice on  $mCDR_{eff}^{cont}$ , as sea-ice cover inhibits air-sea CO<sub>2</sub> exchange and limits mCDR ef-  
 627 ficiency. The impact of sea-ice cover is substantial only in NAS during winter, as Alk-  
 628 impacted waters are transported polarwards into seasonally ice-covered regions.

### 629 **3.3.2 Pulse OAE Experiments**

630 As described in Section 2.2.4, we use three targeted pulse OAE experiments for the  
 631 NAS and ACC deployments. Each uses a different OAE deployment strategy, Yr1995,  
 632 Jan1995, and Jul1995, to further understand how  $mCDR_{eff}$  depends on the season of de-  
 633 ployment. Figure 8 shows time series of  $mCDR_{eff}$  and  $mCDR_{exch}$  (which is essentially  
 634 a normalized net CO<sub>2</sub> flux and is defined in Supporting Information Text S2).

635 For the three NAS experiments (Figure 8a), the time evolution of  $mCDR_{eff}$  is highly  
 636 dependent on the month of Alk deployment. By the end of simulation, Alk deployed in  
 637 summer (Jul1995) reaches an efficiency of roughly 0.9 while winter deployment (Jan1995)  
 638 is only slightly above 0.6; the efficiency of the annual deployment (Yr1995) lies between  
 639 these two extreme values. In addition to seasonally-dependent efficiency for NAS, all three  
 640 experiments show strong seasonality in  $mCDR_{exch}$ , with the highest values occurring dur-  
 641 ing spring; this coincides with the deepest MLD in the region surrounding NAS (see Sup-  
 642 porting Information Figure S3).

643 For the NAS experiments, seasonality of  $mCDR_{eff}^{cont}$  and  $mCDR_{exch}$  (Figures 6c and  
 644 8a, respectively) are generally in phase, and track the seasonality of CO<sub>2</sub> flux. However,  
 645  $mCDR_{eff}$  is not the highest for the deployment months associated with the highest CO<sub>2</sub>  
 646 flux. Instead OAE deployment a few months prior to the maximum value of  $mCDR_{exch}$



**Figure 8.** Monthly-pulse and yearly-pulse experiments for (a) NAS and (b) ACC. Dashed lines show  $mCDR_{exch}$  ( $y$ -axis shown on left-hand-side of figure); solid lines show  $mCDR_{eff}$  ( $y$ -axis shown on right-hand-side of figure).

647 is associated with the highest overall efficiency. In ACC, Jul1995 is associated with lower  
 648  $mCDR_{eff}$  compared to Jan1995, which is consistent with results in NAS, considering that  
 649 these two locations are located in different hemispheres. The difference between these  
 650 two experiments is only a few percent at the beginning of 2017, which also indicates a  
 651 weaker seasonal cycle of  $mCDR_{eff}^{cont}$  and  $mCDR_{exch}$ .

#### 652 **4 Results: $mCDR_{eff}$ from Rapid-mCDR**

653 In this section, we first compare rapid-mCDR against ECCO-Darwin and discuss  
 654 the difference in results when using the two horizontal-averaging methods described in  
 655 Section 2.3. Then as an example use case, we use rapid-mCDR to understand how and  
 656 why mCDR efficiency varies latitudinally across a section in the central Pacific Ocean.

#### 4.1 Evaluation of rapid-mCDR against ECCO-Darwin for the 5 OAE Deployments

Figure 9 shows a comparison of continuous mCDR efficiency,  $mCDR_{eff}^{cont}$ , from rapid-mCDR against ECCO-Darwin for all five continuous OAE experiments. While the main output from rapid-mCDR is the OAE additionality, we prefer to show  $mCDR_{eff}^{cont}$  as it disentangles the effect of  $mCDR_{pot}$ . For each OAE experiment, both rapid-mCDR (Deploy) and rapid-mCDR (HorAdv) simulations are performed. The compared quantities on Figure 9 include:

1. Scatter plots of monthly-mean  $mCDR_{eff}^{cont}$  between ECCO-Darwin and two rapid-mCDR versions. This provides a measure of the overall skill of rapid-mCDR in emulating CO<sub>2</sub> uptake efficiency (left panel).
2. Time series of  $mCDR_{eff}^{cont}$  using a centered-running mean, showing possible bias in rapid-mCDR simulations (middle panel).
3. Seasonal cycle of  $mCDR_{eff}^{cont}$ , further indicating the skill of rapid-mCDR and possible seasonal bias (right panel).

In addition, Supporting Information Figure S5 shows the agreement of  $mCDR_{equil}$  between the two rapid-mCDR versions and ECCO-Darwin. This comparison demonstrates in a compact form the agreement in vertical transport of both Alk and DIC between rapid-mCDR and ECCO-Darwin.

For NAS, Figure 9a shows that  $mCDR_{eff}^{cont}$  from rapid-mCDR (HorAdv) agrees extremely well with ECCO-Darwin for the entire duration of the OAE deployment. The coefficient of determination for the monthly-mean values is  $R^2 = 0.9$ . The rapid-mCDR (HorAdv) slightly underestimates annual-mean values of  $mCDR_{eff}^{cont}$  during the first part of the period, but this underestimation is less than 0.05. The seasonal cycle of  $mCDR_{eff}^{cont}$  is extremely well simulated by this version of rapid-mCDR. As expected, rapid-mCDR (Deploy) does not agree as well with ECCO-Darwin and generally overestimates  $mCDR_{eff}^{cont}$  during most of the simulation period, except for the first five years after start of the OAE deployment. While this overestimation is primarily due to winter period, as revealed by the comparison of the seasonal cycle, the overestimation remains present to some degree throughout the year. The winter overestimation is likely dominated by the absence of sea-ice for the rapid-mCDR (Deploy), which the ECCO-Darwin and rapid-mCDR (Ho-

688 rAdv) are impacted by. As discussed above, Figure 7b indicates that the spatial mean  
 689 of sea-ice area taken over the deployment region can reach up to 0.15 in the winter pe-  
 690 riod, which is expected to arrest winter CO<sub>2</sub> uptake (and therefore  $mCDR_{eff}^{cont}$ ) by roughly  
 691 that fraction.

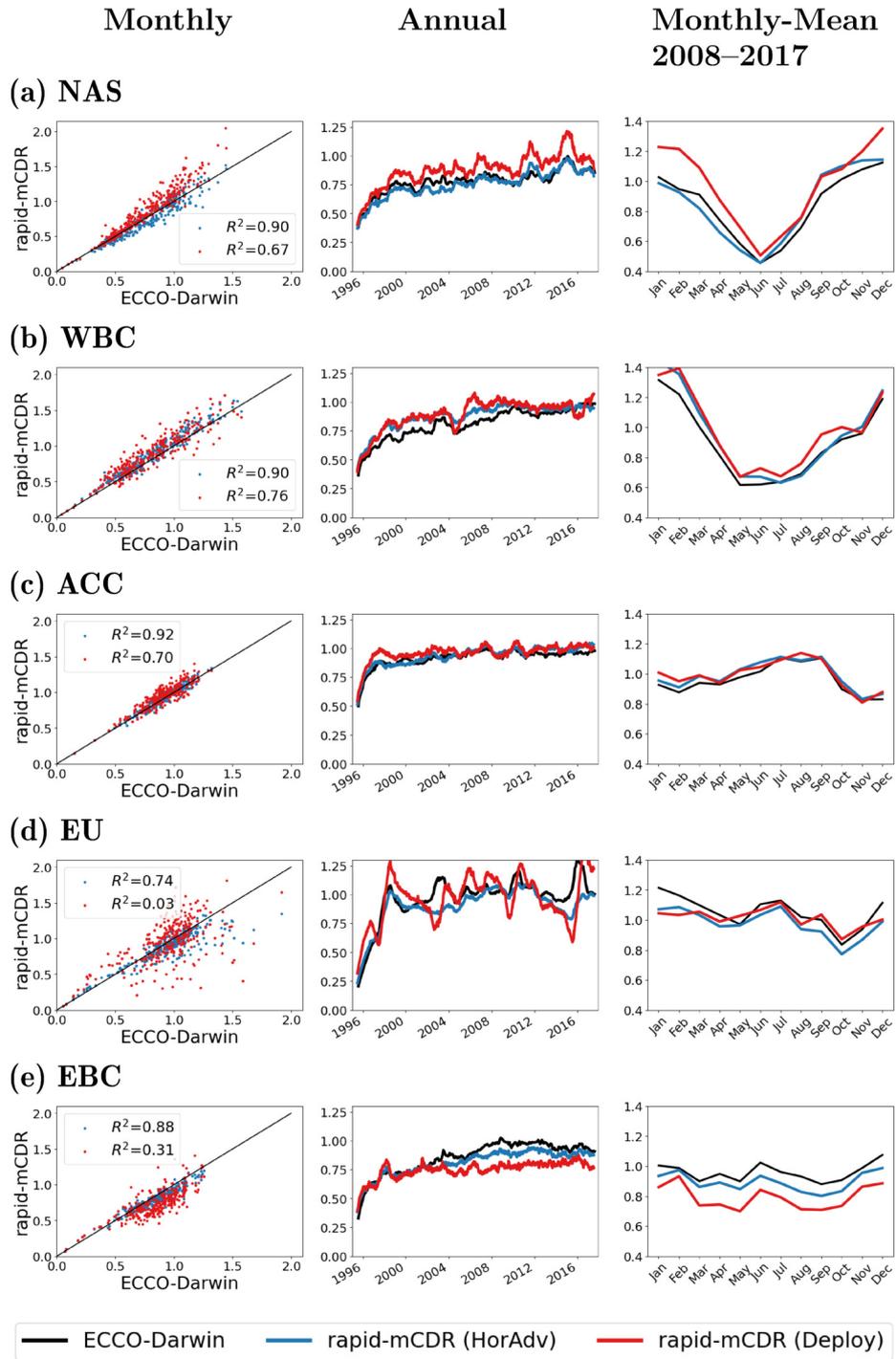
692 For WBC (Figure 9b), both version of rapid-mCDR somewhat overestimate annual-  
 693 mean  $mCDR_{eff}^{cont}$ , where as expected, rapid-mCDR (HorAdv) better represents ECCO-  
 694 Darwin. This is the case for time series of annual-mean values, as well as the coefficients  
 695 of determination which are 0.90 and 0.76 for rapid-mCDR (HorAdv) and rapid-mCDR  
 696 (Deploy), respectively; the seasonal cycle from both versions of rapid-mCDR well repro-  
 697 duces ECCO-Darwin. For ACC (Figure 9c), both version of rapid-mCDR well reproduce  
 698 all aspects of  $mCDR_{eff}^{cont}$ , with rapid-mCDR (HorAdv) performing better than rapid-mCDR  
 699 (Deploy). The difference from the two rapid-mCDR versions is small, despite the OAE  
 700 perturbation extending over a large meridional distance (Figure 5).

701 For EU, the agreement of  $mCDR_{eff}^{cont}$  between the two rapid-mCDR simulations and  
 702 ECCO-Darwin is the poorest of all five OAE experiments, with a  $R^2$  of 0.74 for rapid-  
 703 mCDR (HorAdv) and only 0.03 for rapid-mCDR (Deploy). The annual-mean compar-  
 704 ison shows that rapid-mCDR (Deploy) is unable to well represent multi-annual variabil-  
 705 ity, which is strong in this deployment location. rapid-mCDR (HorAdv) represents this  
 706 multi-annual variability better likely due to capturing horizontal advection.

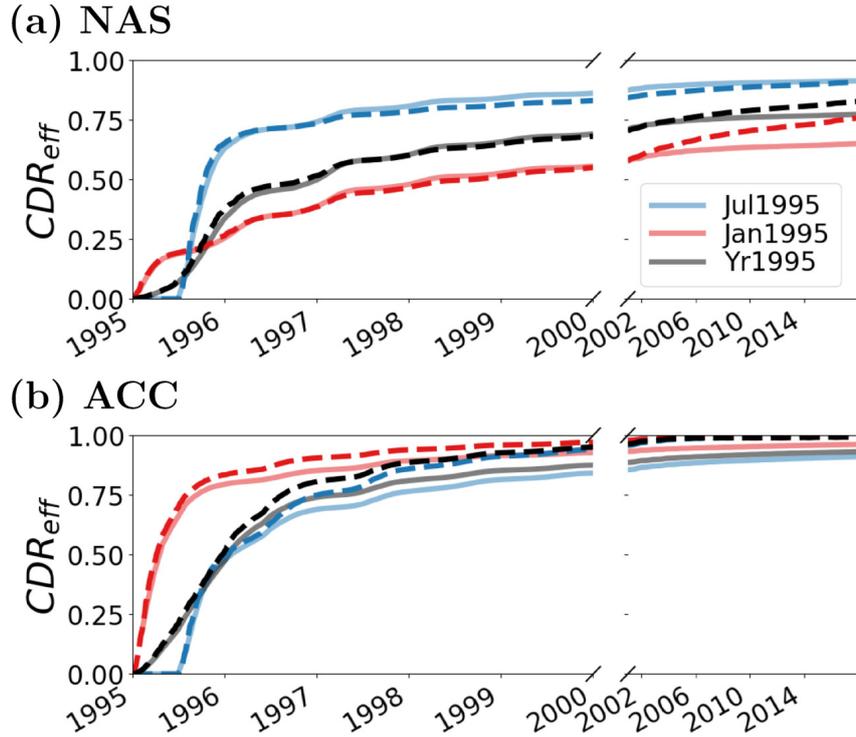
707 In EBC, rapid-mCDR (Deploy) significantly underestimates  $mCDR_{eff}^{cont}$ , especially  
 708 after approximately a decade after the start of Alk deployment, while rapid-mCDR (Ho-  
 709 rAdv) agrees somewhat better with ECCO-Darwin. The cause of this underestimation  
 710 is likely similar as for the EU deployment, where after a number of years the OAE-impacted  
 711 waters enter the tropical Pacific, which is strongly impacted by the ENSO variability.

712 In Figure 10 we show the ability of rapid-mCDR to represent mCDR efficiency for  
 713 the three pulse experiments for NAS and ACC, which were shown to strongly vary with  
 714 deployment season.

715 For NAS, rapid-mCDR agrees very well with ECCO-Darwin over the first five years  
 716 after deployment and shows skill in representing seasonally-varying  $mCDR_{eff}$  as discussed  
 717 above. By the end of the simulation period, rapid-mCDR somewhat overestimates  $mCDR_{eff}$ ;  
 718 this overestimation is consistent for all three experiments. For ACC,  $mCDR_{eff}$  is over-



**Figure 9.** Continuous OAE experiments. Comparison of  $mCDR_{eff}^{cont}$  from both versions of rapid-mCDR against ECCO-Darwin. Left panels (a–e) show monthly-mean rapid-mCDR vs. ECCO-Darwin and associated  $R^2$  values. Middle panels show time series using a 12-month centered-running mean; right panels show monthly-mean values for a zoom in period during the last 10 years of simulation. Blue and red lines represent rapid-mCDR (HorAdv) and rapid-mCDR (Deploy), respectively. Black line in middle and right panels shows ECCO-Darwin results.



**Figure 10.**  $mCDR_{eff}$  for monthly-pulse and yearly-pulse experiments in (a) NAS and (b) ACC. Solid lines show ECCO-Darwin, dashed lines show rapid-mCDR (HorAdv).

719 estimated by rapid-mCDR — a result that is consistent with the continuous OAE ex-  
 720 periments in this region. Rapid-mCDR predicts that by the end of the simulation pe-  
 721 riod  $mCDR_{eff}$  approaches a value of one, which is roughly 0.1 larger than ECCO-Darwin.

## 722 4.2 Expanding Rapid-mCDR to Ocean-basin Scales

723 In this section, we provide an example use case of rapid-mCDR to characterize  $mCDR_{eff}$   
 724 across spatial and temporal scales that might be prohibitively expensive to examine with  
 725 ECCO-Darwin. We also use rapid-mCDR to identify physical processes that adversely  
 726 impact  $mCDR_{eff}$ .

727 We simulate Alk deployment across the meridional extent of the Pacific Ocean, cen-  
 728 tered on 165°W. The deployment sites are spaced 1° apart in latitude from 77°S to 55°N.  
 729 This latitudinal range is chosen so that deployment sites represent open-ocean conditions.  
 730 Each site covers a rectangular area of 10° wide in longitude and 3° wide in latitude; the  
 731 central deployment site locations are shown on Fig. 11a. For all of these sites, we use rapid-

732 mCDR (Deploy) version of the model and where the inputs, which are taken from the  
 733 baseline ECCO-Darwin simulation. At each deployment site, we performed three exper-  
 734 iments using rapid-mCDR — these are equivalent to the three ECCO-Darwin pulse ex-  
 735 periments discussed in Section 2.2.4: *Yr1995*, *Jul1995*, and *Yr1995*. As with ECCO-Darwin,  
 736 these experiments are run until December 31, 2017 and the values of  $mCDR_{eff}$  are eval-  
 737 uated at the end of simulation.

738 Furthermore, to isolate the role of physical processes (vertical advection, vertical  
 739 diffusivity, and sea-ice cover) on  $mCDR_{eff}$ , we performed three additional sets of rapid-  
 740 mCDR sensitivity experiments to separate and quantify the impact of each of these fac-  
 741 tors. These three sensitivity experiments are based on the Yr1995 experiment described  
 742 above with the following modifications: 1) *Yr1995-w0* is an experiment with vertical ve-  
 743 locity set to zero, 2) *Yr1995-k0* is an experiment with vertical diffusivity set to zero, and  
 744 3) *Yr1995-ice0* is an experiment with no sea-ice cover (i.e., open-water conditions).

745 Figure 11b shows  $mCDR_{eff}$  for the Yr1995 experiment at the end of the simula-  
 746 tion, plotted against central latitude of deployment site, along with profiles of time-mean  
 747 vertical velocity and vertical diffusivity. Figure 11c shows profiles of normalized Alk per-  
 748 turbation (i.e.,  $\widehat{\Delta Alk}$  normalized by the maximum value of all experiments) at the end  
 749 of simulation time to show the vertical extent of the OAE perturbation.

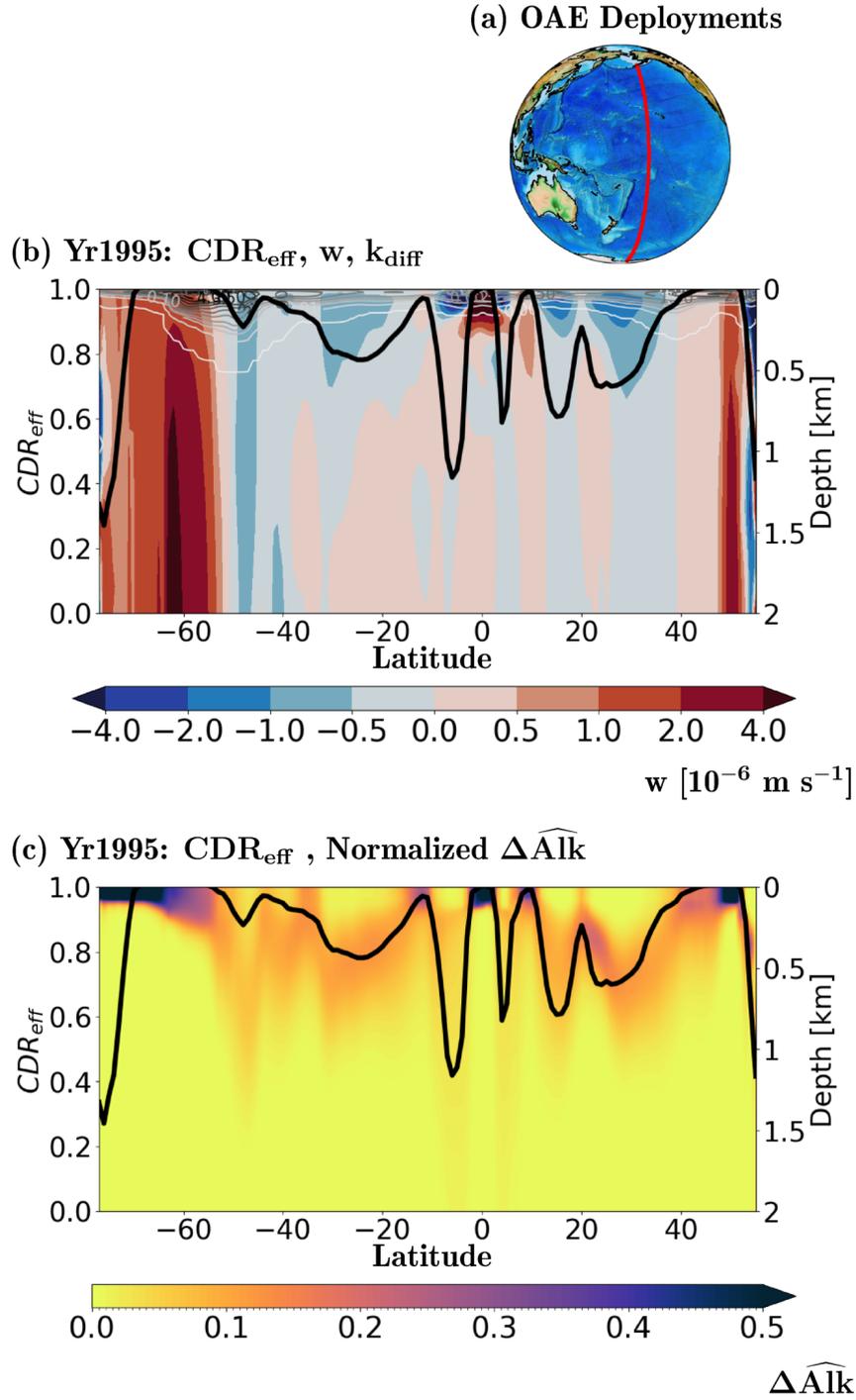
750 We find that  $mCDR_{eff}$  is strongly dependent on deployment site location, with the  
 751 largest values found in tropical regions, mid-latitude and polar regions in the southern  
 752 hemisphere (between approximately 60–50°S) and mid-latitudes in the northern hemi-  
 753 sphere (between approximately 40–50°N). The lowest values (less than 0.5) are gener-  
 754 ally found in subtropical regions and in the northern extent of polar regions. Except for  
 755 polar regions in the southern hemisphere, high  $mCDR_{eff}$  values are found at locations  
 756 associated with significant ocean upwelling and outcropping and the lowest values are  
 757 located in downwelling regions. Figure 11c shows that high values of  $mCDR_{eff}$  are as-  
 758 sociated with Alk perturbations that remain close to the surface; for locations associ-  
 759 ated with low values, Alk perturbations are either transported to depth or spread over  
 760 a substantial vertical extent.

761 Figure 12 shows  $mCDR_{eff}$  for the three deployment seasons (Figure 12a) and for  
 762 the three sensitivity experiments (Figure 12b); these quantities are compared against the  
 763 Yr1995 experiment (Figure 12a,b, black lines) and CDRpot and sea-ice cover (Figure 12c).

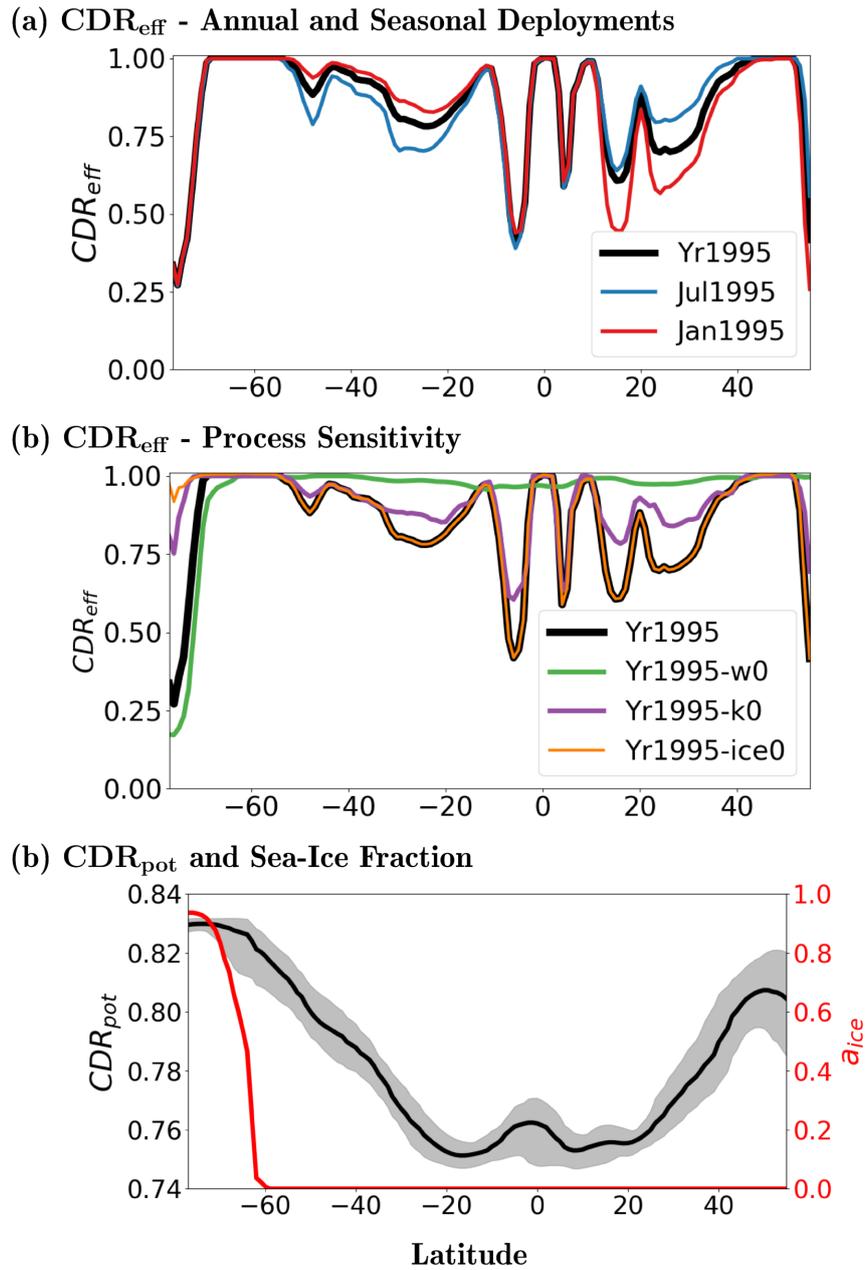
764 All quantities are plotted as a function of deployment site latitude. For locations in mid-  
765 latitudes and subtropical regions, there is strong dependence of  $mCDR_{eff}$  on the deploy-  
766 ment season. Summer months are generally associated with higher efficiency compared  
767 to winter, which is consistent with the pulse experiments results for NAS and ACC (see  
768 Section 3.3). The difference of  $mCDR_{eff}$  between the deployments in summer season reach  
769 up to 0.3 higher values than the deployments in the winter season.

770 The experiments shown in Figure 12b demonstrate that for polar OAE deployment  
771 sites in the southern hemisphere, which are under the influence of seasonal sea ice, the  
772 ice cover efficiently prevents  $\text{CO}_2$  uptake and therefore these regions are associated with  
773 low values of  $mCDR_{eff}$ . Removing sea-ice cover in rapid-mCDR (Figure 12b, orange line)  
774 increases  $mCDR_{eff}$  to values close to one below roughly  $50^\circ\text{S}$ . Therefore, our simulations  
775 suggest that mCDR efforts will be much less effective in this, and other ice-covered re-  
776 gions.

777 From the two dominant ocean circulation processes, vertical velocity and diffusiv-  
778 ity, we find that vertical velocity dominates low-efficiency regions (i.e., the role of ver-  
779 tical diffusivity here is second order). There is only a small increase of  $mCDR_{eff}$  with  
780 respect to the Yr1995 experiment if the vertical diffusivity is set to zero (Figure 12b, pur-  
781 ple line). However, if the vertical velocity is set to zero  $mCDR_{eff}$  becomes close to one  
782 for most of the deployment sites (Figure 12b, green line), except for ice-covered regions  
783 in the southern hemisphere. Figure 12c shows  $mCDR_{pot}$  vs. latitude; the lowest values  
784 are found in the tropical/subtropical regions of the Pacific Ocean, with increasing val-  
785 ues towards the poles. The variability of  $mCDR_{pot}$  over each deployment site is small  
786 (Figure 12c, gray shaded envelope) compared to the latitudinal variability.



**Figure 11.** (a) Location of rapid-mCDR deployment sites across the Pacific Ocean; (b)  $mCDR_{eff}$  at the end of 2017 for Yr1995 experiment (solid black line), profile of mean vertical velocity (colored contours), and vertical diffusivity (grayscale contour lines with units of  $10^{-2} \text{ m}^2 \text{ s}^{-1}$ ); (c)  $mCDR_{eff}$  at the end of 2017 for Yr1995 experiment (solid black line) and normalized  $\widehat{\Delta Alk}$  (colored contours).



**Figure 12.** Pacific Ocean vertical sections of  $mCDR_{eff}$  at the end of 2017 for (a) 3 different pulse deployment seasons (Jul1995, Jan1995, and Yr1995). (b) Experiment with vertical velocity and diffusivity set to zero (Yr1995-w0 and Yr1995-k0, respectively) and simulation without sea ice forcing (Yr1995-ice0). (c) Mean  $mCDR_{pot}$  and variability over the deployment site (solid black lines and gray shading, respectively); these are computed from daily-mean values and time-mean sea-ice cover. All values are shown at the central latitude of the deployment site.

## 5 Discussion

In recent years, mCDR efforts via OAE have gathered considerable attention as a potential method for removing anthropogenic CO<sub>2</sub> from the atmosphere. The OAE approach mimics natural processes (Subhas et al., 2023) and has a potential to be scaled-up to significantly mitigate climate change (Renforth & Henderson, 2017). As the efficiency of OAE-based mCDR varies across different spatial and temporal scales, field deployments must be carefully planned to achieve maximum efficiency while minimizing cost and logistical risk. While a number of field methods, technical approaches, and experiment designs have been proposed (Eisaman et al., 2023), the use of numerical ocean models to simulate and quantify OAE impacts before expensive field trials occur, and provide much-needed MRV quantification (Ho et al., 2023), still remains in its infancy.

While experiments and observations (Boyd et al., 2023) will be invaluable to inform these efforts, environmentally- and societally-responsible OAE perturbations (Fakhraee et al., 2022; Nawaz et al., 2023) should modify the natural ocean carbonate and ecological state (Ferderer et al., 2022) only slightly compared to its natural variability. Our work shows that the impacts of OAE are spatially dispersed across ocean basins before the full CO<sub>2</sub> potential is realized. Due to these two factors, it will be difficult if not impossible to observe and separate actual OAE-deployment effects from natural ocean variability. Therefore, the optimization of deployment strategies and their MRV will have to heavily rely on numerical models. Numerical models are also the ideal tool for exploring and quantifying efficiencies of potential mCDR deployment strategies before significant investments in deployment infrastructure occur.

In this paper, we use a state-of-the-art ocean biogeochemistry state estimate (ECCO-Darwin) constrained by a suite of in-situ and remotely-sensed observations, to quantify OAE additionality and to characterize the resultant 3-D ocean carbonate state perturbation attributed to regional-scale, multi-decadal 1) continuous and 2) month- and year-long surface-ocean Alk deployment. To our knowledge, ECCO-Darwin is the only open-source model at present time that is ideally suited for attribution of the ocean physical-sea-ice-biogeochemical state and OAE additionality. This is because of its unique data assimilation approach, which is a combination of adjoint-based (Wunsch et al., 2009; Wunsch & Heimbach, 2013) and Green's function (Menemenlis et al., 2005) approaches that constrains the dynamical, carbonate, and biogeochemical state with a suite of observa-

819 tions (Carroll et al., 2022). Thus ECCO-Darwin provides an accurate background state,  
820 and in particular, 3-D ocean physics, for studying the impact of mCDR over multi-decadal  
821 timescales. Furthermore, the data assimilation in ECCO-Darwin does not introduce non-  
822 physical observation-based nudging or increments which can conceal the impact of OAE.

823 The alternative approaches include forward-only ocean-biogeochemistry models un-  
824 constrained by observations. These models typically exhibit larger biases in terms of ocean  
825 dynamics and carbonate cycle (e.g., Séférian et al., 2020; Fu et al., 2022) compared to  
826 the ECCO-Darwin solution described in Carroll et al. (2020, 2022). These model biases  
827 are expected to contribute to additional uncertainty and biases in ocean CO<sub>2</sub> solubil-  
828 ity and dynamics, which are both important considerations for mCDR studies. Model-  
829 ing systems that assimilate either one or all of the components of the ocean system (e.g.,  
830 Perruche, 2018; Turner et al., 2023) are usually based on sequential data assimilation and  
831 correct simulated fields with observational increment. These data assimilation systems  
832 are geared towards the best representation of the ocean state, but conceal relationship  
833 between processes which introduces uncertainties in attribution studies, for example at-  
834 tribution of OAE additionality.

835 Furthermore, we use our numerical ocean model results to motivate and develop  
836 a 1-D model for rapid quantification of OAE additionality (rapid-mCDR). Rapid-mCDR  
837 provides a user friendly and easily-deployable model for mCDR end-users that can be  
838 used across various ocean conditions without the need for supercomputing resources —  
839 which is a key advantage compared to more-complex ECCO-Darwin simulations. Com-  
840 bining the 1-D model approach with output from a numerical ocean biogeochemistry model,  
841 such as ECCO-Darwin, permits rapid characterization of mCDR additionality at any lo-  
842 cation in the global-ocean model grid, which can be a backbone for MRV purposes, as  
843 well as a tool for rapid comparison and optimization of different OAE deployment strate-  
844 gies. All of our experiments represent open-ocean deployments (rather than coastal sites).  
845 We are aware that many planned OAE field deployments will occur in coastal regions  
846 or from the nearshore zone, which might require additional model improvements and fea-  
847 tures.

848 Similar to Wang et al., 2023, we separate mCDR efficiency into two factors con-  
849 trolling: 1) CO<sub>2</sub> solubility (mCDR potential,  $mCDR_{pot}$ ; the maximum amount of CO<sub>2</sub>  
850 that can be sequestered per deployed Alk), and 2) dynamical efficiency ( $mCDR_{eff}$ ; a non-

851 dimensional function that represents a fraction of realized  $mCDR_{pot}$  with the time after  
 852 deployment, which is dominated by ocean dynamics and sea-ice cover). We character-  
 853 ize  $mCDR_{pot}$  globally from two independent data sources, the 1) baseline ECCO-Darwin  
 854 simulation and 2) OceanSODA-ETHZ dataset. The  $mCDR_{pot}$  from both datasets shows  
 855 similar features, including:

- 856 • Meridional dependence dominated by increase of CO<sub>2</sub> solubility with colder SSTs.  
 857 This indicates that in the absence of dynamical effects, near-polar regions would  
 858 be associated with the highest potential for CO<sub>2</sub> removal.
- 859 • Weaker dependence within the ocean basins dominated by meridional transport  
 860 and vertical mixing associated with basin-scale boundary currents, river inflows,  
 861 and sea-ice melt which impact the saturated surface-ocean CO<sub>2</sub> state.
- 862 • Seasonal variability is small for most regions; the highest seasonal variability is  
 863 found in the mid-litudinal regions and particularly in western boundary currents.
- 864 • Despite substantial secular trends in CO<sub>2</sub> uptake (Carroll et al., 2020), the lin-  
 865 ear trend of  $mCDR_{pot}$  remains below 0.01 mol C/mol Alk per decade, with min-  
 866 imal interannual variability. We expect an overall decrease of  $mCDR_{pot}$  in the fu-  
 867 ture climate due to ocean warming and accelerated acidification.

868 Compared to mCDR potential, evaluation of dynamical mCDR efficiency using ECCO-  
 869 Darwin is computationally intensive – for each considered OAE deployment, a multi-decadal  
 870 ECCO-Darwin simulation is run and  $mCDR_{eff}$  is computed from the additionality of CO<sub>2</sub>  
 871 uptake with respect to the baseline simulation.

872 We find that OAE simulations with continuous Alk deployment are well suited for  
 873 a general characterization of regional mCDR efficiency, as these simulations can also pro-  
 874 vide information on the seasonal cycle of OAE-induced CO<sub>2</sub> uptake. Locations associ-  
 875 ated with large seasonality are likely to exhibit sensitivity in  $mCDR_{eff}$  depending on the  
 876 deployment season, which can be further quantified with targeted short-term (pulse) ex-  
 877 periments. The main feature of dynamical mCDR efficiency are:

- 878 • Regional-scale ocean circulation, and in particular vertical transport, exerts a strong  
 879 control on the space-time distribution of  $mCDR_{eff}$  and taken together with  $mCDR_{pot}$   
 880 are the first-order control on sequestration efficiency of OAE-induced atmospheric  
 881 CO<sub>2</sub> to depth.

- 882 • Downwelling/subduction regions are associated with low values of  $mCDR_{eff}$  and  
883 upwelling regions exhibit high  $mCDR_{eff}$  — this is because of the relatively long  
884 timescales of ocean-atmosphere  $CO_2$  equilibration (on the order of years) which  
885 takes place against the backdrop of shorter-scale ocean dynamics which can iso-  
886 late  $CO_2$  from non-equilibrated waters from the atmosphere.
- 887 • In high-latitude regions, sea-ice cover can strongly reduce  $mCDR_{eff}$  due to block-  
888 ing of air-sea gas exchange.
- 889 • For extratropical deployments,  $mCDR_{eff}$  can be heavily dependent on the deploy-  
890 ment season — summer is generally associated with higher values.
- 891 • Multi-annual variability in  $mCDR_{eff}$  is found for all deployment sites, and is par-  
892 ticularly significant in the Tropical and Equatorial Pacific Ocean.
- 893 • For most of the studied deployments, OAE-impacted waters remain above 500 m  
894 depth for the duration of the 27-year long continuous experiments. The exception  
895 to this is NAS, in which the OAE perturbation reaches below 1000 m within roughly  
896 four years and eventually penetrates below 2000 m.

897 We stress that care must be taken when relating the seasonal cycle of dynamical  
898 efficiency from continuous OAE experiments to the most efficient deployment season, as  
899 these are not the same. The seasonal efficiency from continuous OAE experiments in-  
900 dicates the seasonality of OAE additionality with the most-efficient deployment time of-  
901 ten being a few months prior to the season with the highest  $CO_2$  uptake.

902 One of our deployment sites, NAS, is very close to the Iceland pulse experiment  
903 from (He & Tyka, 2023), for which they find a much lower efficiency compared to their  
904 other locations. Our experiments indicate that the time-mean mCDR efficiency of that  
905 location is comparable to the other sites examined in this study. The NAS location is  
906 however associated with large seasonal variability of mCDR efficiency, with very low val-  
907 ues during winter for which the deployment in (He & Tyka, 2023) was performed. There-  
908 fore, our results largely agree with (He & Tyka, 2023) and we further show that their  
909 results are heavily influenced by the seasonal cycle of mCDR efficiency.

910 We find that the 1-D model rapid-mCDR can rapidly and realistically reproduce  
911 OAE simulations performed with a complete 3-D ocean biogeochemistry model. Futher-  
912 more, rapid-mCDR can be used to isolate and compare the individual processes that drive  
913 mCDR efficiency and thus provides additional benefits in terms of improving the phys-

914 ical understanding of mechanisms that control mCDR efficiency. The rapid-mCDR in-  
 915 puts are horizontal mean fields from the baseline ECCO-Darwin simulation, which pro-  
 916 vides a background information on the ocean state that is affected by OAE. We test two  
 917 approaches with respect to the horizontal averaging method: 1) ocean fields are spatial  
 918 means over the deployment site — the results of this approach neglect horizontal advec-  
 919 tion of the OAE perturbation and 2) Surface-ocean advection is included and used to  
 920 advect the OAE perturbation throughout the water column. We refer to the two approaches  
 921 as rapid-mCDR (Deploy) and rapid-mCDR (HorAdv), respectively.

922 The key findings using rapid-mCDR are:

- 923 • We find good agreement with ECCO-Darwin in extratropical regions, especially  
 924 when the horizontal advection is considered.
- 925 • For tropical regions, interannual variability is poorly represented, especially for  
 926 the rapid-mCDR simulation which neglects horizontal advection. We note that  
 927 including horizontal advection improves its representation, although there is still  
 928 room for improvement. We speculate that the horizontal advection of OAE per-  
 929 turbation and strong spatial gradients in ocean dynamics surrounding the EU de-  
 930 ployment site play an important role in controlling  $mCDR_{eff}$ , which can only be  
 931 crudely represented by the 1-D rapid-mCDR model.
- 932 • Ocean vertical velocity dominates over vertical diffusivity in its control on  $mCDR_{eff}$ .  
 933 In high-latitudes, seasonal sea-ice cover can significantly decrease  $mCDR_{eff}$ .

### 934 **5.1 Future Model Improvements**

935 The ECCO-Darwin experiments shown in this work are idealized and we assume  
 936 that surface-ocean Alk rate is known without consideration of a specific deployment method.  
 937 Therefore, we suggest that future work tailoring numerical ocean simulations towards  
 938 more-realistic deployment strategies might include:

- 939 • Improved parameterization of interactions between the OAE material and seawater  
 940 to represent relevant processes for the particular deployment strategy and might  
 941 include mineral dissolution and precipitation (e.g., Fennel et al., 2023).
- 942 • Simulation of deployment via minerals and dissolution products, such as Si and  
 943 Fe, which might interact with ocean biota (Bach et al., 2019). While there is un-

944 certainty in understanding of the response of major phytoplankton types to in-  
945 creased Alk (e.g., Gately et al., 2023), ECCO-Darwin is well suited to account for  
946 dispersion of these products as the Darwin component can simulate their impact  
947 on key plankton functional types.

- 948 • In the current version of ECCO-Darwin, air-sea CO<sub>2</sub> flux in seasonally ice-covered  
949 regions is simplified — with CO<sub>2</sub> flux being scaled by the fraction of open-water  
950 area, i.e., 1 - sea-ice cover. Future work should account for a realistic represen-  
951 tation of air-sea gas exchange through sea-ice cracks and leads (Loose & Schlosser,  
952 2011; Søren et al., 2011).
- 953 • Developing regional downscaled set-ups of ECCO-Darwin on higher-resolution grids,  
954 or incorporation of unstructured grids in nearshore mCDR simulations (Ward et  
955 al., 2020), to improve resolution and representation of the coastal periphery and  
956 topography in the desired region. This will provide a better representation of ocean  
957 dynamics and help resolve small-scale coastal flows, which may be important for  
958 coastally-based deployment strategies.

959 We envision that rapid-mCDR will continue to be a useful tool for quick and ef-  
960 ficient evaluation of potential OAE deployments or as the backbone for MRV. Future im-  
961 provements to rapid-mCDR might include:

- 962 • Improved representation of horizontal advection and dispersion of OAE-impacted  
963 seawater. The rapid-mCDR simulations at all 5 deployment regions demonstrate  
964 improvements in terms of fit to ECCO-Darwin when surface-ocean advection is  
965 considered. We note that this improvement is particularly significant for tropical  
966 regions (EU). We suggest that two possible approaches could be used to improve  
967 rapid-mCDR accuracy in this regard: 1) an Eulerian approach where surface ad-  
968 vection is estimated using an offline calculation, for example with OceanPARCELS  
969 (Lange & van Sebille, 2017; Delandmeter & van Sebille, 2019) and 2) a Lagrangian  
970 approach where rapid-mCDR is coupled to a particle tracking model and solved  
971 at each point along the dispersal trajectory. This could be used to estimate the  
972 transport, sinking, and dissolution of minerals added to the surface ocean and in-  
973 form regional-scale Eulerian ocean model simulations.
- 974 • Improving representation of bathymetry and its impact on the spread of OAE-impacted  
975 waters. The current version of rapid-mCDR is developed for the deep ocean and

976 we assume that the OAE impact does not spread to the seafloor. At the seafloor,  
977 rapid-mCDR could be coupled with a sediment diagenesis model, such as RADI  
978 (Sulpis et al., 2021), to account for sequestration of particulate carbon in sediment  
979 and the resultant fluxes between porewaters and the overlying seawater.

- 980 • Parameterization of biogeochemical processes to tailor rapid-mCDR for other mCDR  
981 approaches, e.g., ocean afforestation/macroalgae growth, iron-fertilization, and  
982 enhanced phytoplankton growth.
- 983 • Implement a module for uncertainty quantification of mCDR impact using a Monte-  
984 Carlo/ensemble approach.

## 985 **6 Summary and Conclusions**

986 Using a data-assimilative ocean biogeochemistry model (ECCO-Darwin), we have  
987 characterized the regional-scale efficiency of OAE additionality over seasonal to multi-  
988 decadal timescales. Using both pulsed and continuous OAE experiments at five distinct  
989 open-ocean deployment sites, this work highlights the strong role of three-dimensional  
990 ocean dynamics in transporting OAE-induced atmospheric carbon across ocean basins  
991 and to depth. We also develop a 1-D model approach (rapid-mCDR) that can be run  
992 on a personal computer to rapidly characterize OAE efficiency at any global-ocean lo-  
993 cation, with a single multi-decadal simulation taking only about 1 CPU minute. Rapid-  
994 mCDR can be readily expanded to other mCDR approaches, such as ocean afforestation  
995 and iron fertilization. Our foundational data-constrained modeling work provides a path  
996 forward for quantifying the global-ocean response to OAE deployments and can be used  
997 to develop high-resolution downscaled OAE simulations to complement and support fu-  
998 ture mCDR field trials.

## 999 **Open Research Section**

1000 ECCO-Darwin model output is available at the ECCO Data Portal: [http://data.nasa](http://data.nasa.gov/ecco/)  
1001 [.nasa.gov/ecco/](http://data.nasa.gov/ecco/)

1002 Model code and platform-independent instructions for running ECCO-Darwin and rapid-  
1003 mCDR simulations are available at: <https://doi.org/10.5281/zenodo.10562714>

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