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Pathways for marine carbon dioxide removal using electrochemical acid-base generation

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Research over the past decade has resulted in various methods for removing CO₂ from the atmosphere using seawater and electrochemically generated acids and bases. This Perspective aims to present a unified framework for comparing these approaches. Specifically, these methods can all be seen as falling into one of two categories: those that result in a net increase in ocean alkalinity and use the “ocean as a sponge” for atmospheric CO₂ (ocean alkalinity enhancement, or OAE) and those that cycle ocean alkalinity and use the “ocean as a pump” for atmospheric CO₂ (ocean alkalinity cycling, or OAC). In this Perspective, approaches for marine carbon dioxide removal (mCDR) using electrochemistry are compared using this framework, and the similarities and differences of these two categories are explored.

KEYWORDS

carbon dioxide removal, negative emissions technologies, marine carbon dioxide removal, ocean carbon dioxide removal, ocean alkalinity enhancement, indirect ocean capture, direct ocean capture, ocean alkalinity cycling

1 Introduction

There is now scientific consensus that in addition to decarbonization, the active removal of around 5 Gt of atmospheric CO₂ per year by 2100 will be necessary to prevent an average global warming of more than 2°C (Calvin et al., 2023). Among the carbon dioxide removal (CDR) technologies being pursued, marine carbon dioxide removal (mCDR) has the potential to reach climate-relevant scales of durable CO₂ removal (NASEM, 2022). mCDR leverages the natural role of the oceans in the carbon cycle to remove CO₂ from the air. An important group of mCDR techniques are those that remove CO₂ using aqueous acid and base that have been generated electrochemically from the seawater itself, with the two most actively pursued approaches being electrochemical ocean alkalinity enhancement (OAE) (Oschlies et al., 2023; Ringham et al., 2024) and what I will refer to here as electrochemical ocean alkalinity cycling (OAC) (de Lannoy et al., 2018). In the case of OAE, CO₂ is removed from the atmosphere and durably stored as dissolved inorganic carbon (DIC) in the ocean, whereas OAC captures CO₂ from the atmosphere in its gaseous form (Eisaman et al., 2023). In past publications electrochemical OAC been variously referred to as: CO₂ extraction from seawater (Eisaman et al., 2012), indirect ocean capture (IOC): acid process (de Lannoy et al., 2018; Eisaman et al., 2018; Eisaman, 2020), direct ocean capture (DOC) (Bui et al., 2023; Kim et al., 2023; Lucas et al., 2023), direct ocean removal (DOR) (NOAA, 2023), CO₂ removal from oceanwater (Kim et al., 2023), and electrochemical direct ocean capture (eDOC) (Aleta

et al., 2023). This Perspective will use the OAC nomenclature, as it succinctly and clearly emphasizes its relation to electrochemical OAE, the approach to which it is most closely related. The purpose of this perspective is to provide an understanding of the fundamental operating principles and relative merits of electrochemical OAE and OAC.

It should be mentioned that there is what appears to be a third category of approaches, which have variously been termed indirect ocean capture (IOC): base process (Eisaman et al., 2017; de Lannoy et al., 2018; Eisaman et al., 2018) and electrolytic seawater mineralization (La Plante et al., 2023). In these methods, the base is used to increase the alkalinity and pH of seawater to induce the removal of DIC from seawater by precipitating solid carbonates and/or hydroxides. The precipitation step removes some, but not all, of the added alkalinity. Therefore, the alkalinity of the seawater remaining after precipitation is still elevated relative to the starting point. When equilibrated with the air, this alkalinity enhanced seawater will result in the removal of CO₂ from the air and storage as DIC in the ocean. These approaches are just a form of OAE where some portion of the added alkalinity goes toward precipitating DIC as solids. While this precipitation may appear inefficient because it removes some of the alkalinity that was just added, these approaches may be motivated by the reuse value or storage stability of solid carbonates or the more gravimetrically and volumetrically dense form of alkalinity in solid hydroxides.

Many methods of alkalinity generation for OAE exist other than the electrochemical generation of aqueous base, such as the addition of terrestrial alkaline minerals to the ocean (Caserini et al., 2022; Eisaman et al., 2023). However, this Perspective will focus specifically on OAE and OAC using electrochemically generated acid and base,

and so will generally drop the “electrochemical” qualifier from this point forward.

1.1 Pathways for mCDR using electrochemically generated acid and base: “ocean as a sponge” and “ocean as a pump”

Figure 1 compares the process flows for OAE, i.e., “ocean as a sponge” (panel A), and OAC, i.e., “ocean as a pump” (panel B). The first step of the OAE process, shown in Figure 1A, is the increase of the total alkalinity (TA) of seawater. For the electrochemical OAE processes we are considering here, this can be accomplished in two ways. One is to generate the acid and base from a separate brine stream (most commonly the acid, base, and salt streams are HCl and NaOH generated from NaCl), and then adding the base mixed with the partially desalted brine to the ocean. Another approach is to generate the acid and base from seawater pumped directly from the ocean, returning the base mixed with the partially desalted seawater back to the ocean. This latter case can be seen as increasing ocean alkalinity through acid removal since the acid that is removed is generated from the seawater itself. As shown on the TA-DIC-pH diagram in Figure 2, the first alkalinity addition step of OAE increases the TA and the pH of the brine or seawater, represented as a vertical arrow on the diagram.

The next, and final, step in the OAE process shown in Figure 1A is the equilibration of the seawater or brine containing elevated TA and pH with the air, resulting in the net removal of CO₂ from the air and storage in the ocean as additional DIC (mostly in the form of bicarbonate ions, HCO₃⁻). As indicated by the red box, Monitoring, Reporting, and Verification (MRV) must be performed on this step to

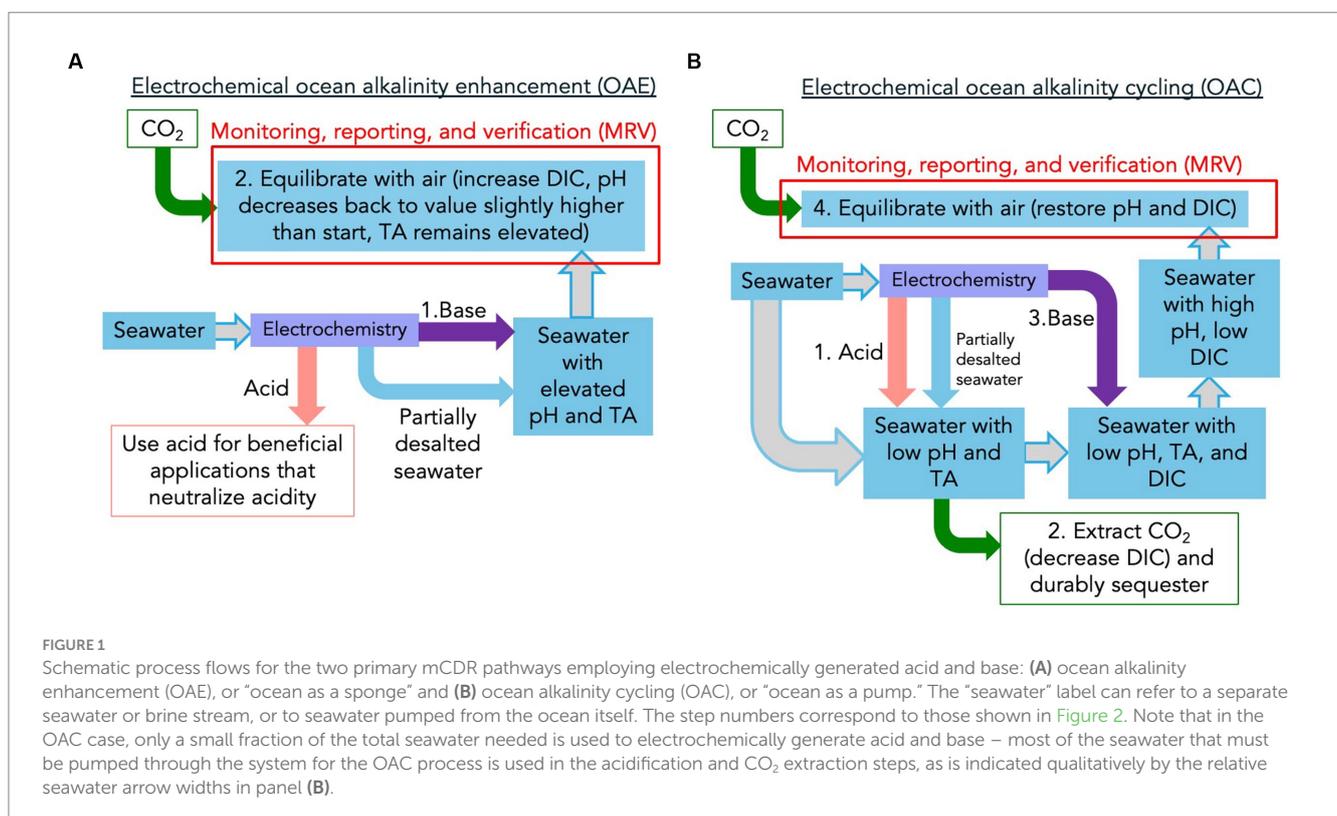


FIGURE 1

Schematic process flows for the two primary mCDR pathways employing electrochemically generated acid and base: (A) ocean alkalinity enhancement (OAE), or “ocean as a sponge” and (B) ocean alkalinity cycling (OAC), or “ocean as a pump.” The “seawater” label can refer to a separate seawater or brine stream, or to seawater pumped from the ocean itself. The step numbers correspond to those shown in Figure 2. Note that in the OAC case, only a small fraction of the total seawater needed is used to electrochemically generate acid and base – most of the seawater that must be pumped through the system for the OAC process is used in the acidification and CO₂ extraction steps, as is indicated qualitatively by the relative seawater arrow widths in panel (B).

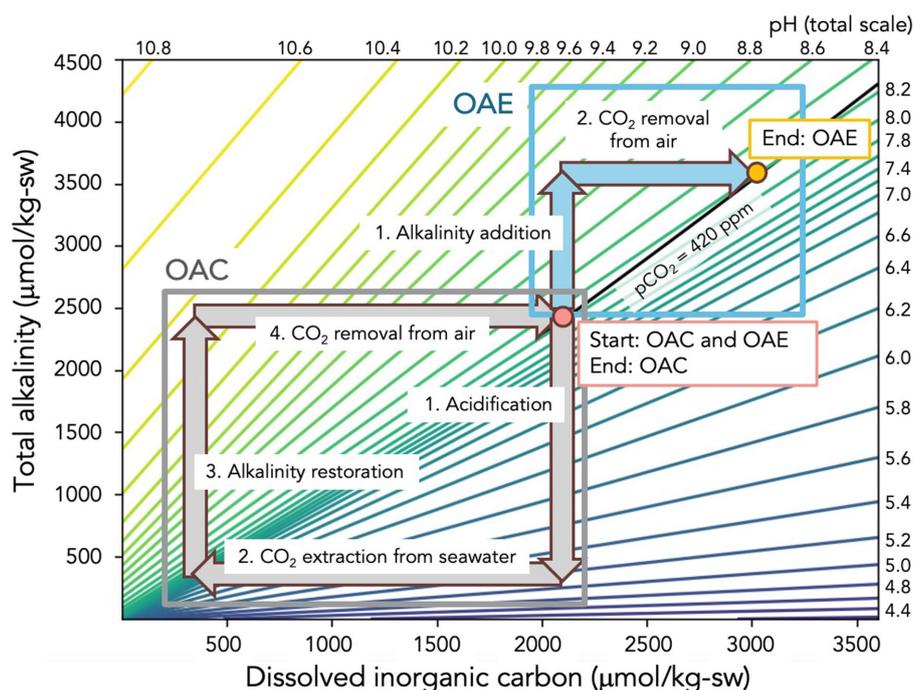


FIGURE 2

The TA-DIC-pH pathway for OAE and OAC. The underlying TA-DIC-pH diagram was created using PyCO2Sys (Humphreys et al., 2022), version 1.8.2 using carbonate equilibrium constants from Lueker et al. (2000), the bisulfate dissociation constant from Dickson (1990), borate: salinity from Lee et al. (2010), and pH on the total scale. The black solid line represents the condition where the surface ocean is in equilibrium with an atmospheric $p\text{CO}_2$ value of 420 ppm. The start and end points of both OAC and OAE lie on this line. Because the slope of this line is slightly steeper than the slope of the constant pH lines, the end point of OAE is at a slightly higher pH value than the start. OAC is a rectangle in TA-DIC space, and OAE is an inverted “L” shape. The lengths of the arrows shown above are just one example, and others are possible. For example, the acidification and alkalinity restoration arrows (or the CO_2 extraction and CO_2 removal arrows) for OAC could be shorter or longer than shown above. Likewise, the alkalinity addition arrow for OAE could be shorter or longer. Finally, for OAE, the length ratio of the CO_2 removal arrow to the alkalinity addition arrow is set to be 0.8, but other values are possible (He and Tyka, 2023).

quantify the amount of CO_2 moved from the air into oceanic DIC as a result of the OAE process (Ho et al., 2023). If the alkalinity is released to the ocean immediately after it is generated, the MRV of CO_2 removal must be performed in the open ocean as the plume of enhanced alkalinity spreads and dilutes as it mixes with the ocean. This is referred to as “open system MRV.” In principle, the alkalinity can be contacted with, and remove CO_2 from, the air in closed containers before releasing it to the ocean. This is referred to as “closed system MRV.” Relative to open systems, closed systems have the advantage of more straightforward MRV, but the disadvantage of greater cost incurred by the equipment and energy needed for controlled air contacting. For a given implementation, these tradeoffs can be optimized by operating in a hybrid mode that partially equilibrates the alkalinity in a closed system, and then releases to the ocean for the remaining CO_2 removal to occur in the open ocean.

The flow diagram and TA-DIC-pH pathway for OAC are shown in Figures 1B, 2. The final two steps of OAC (Steps 3 and 4 – alkalinity restoration and CO_2 removal) are the same as the first and only two steps of OAE. In the case of OAC, however, there are two initial steps not required for OAE: acidification and CO_2 extraction from seawater. The acidification step shifts the carbonate buffer system of the seawater or brine such that all DIC is now in the form of dissolved CO_2 gas. This corresponds to a downward arrow in Figure 2 as TA and pH are decreased. The CO_2 extraction step removes this CO_2 gas from seawater, resulting in the decrease in DIC shown in Figure 2. Note that because the equilibrium partial pressure of seawater DIC converted to

CO_2 gas is much less than atmospheric pressure, vacuum stripping must be used to extract relatively pure CO_2 gas (de Lannoy et al., 2018; Eisaman, 2018; Eisaman et al., 2018). Step 3 for OAC uses the generated base to restore the seawater TA back to its starting value, as shown by the vertical arrow in Figure 2. Finally, Step 4 of OAC is the equilibration of the seawater with air, resulting in CO_2 removal from the air and storage as DIC. As is the case for OAE, the MRV for OAC occurs during this equilibration step, and can be designed as an open, closed, or hybrid system. In the case of OAC, the maximum amount of CO_2 pulled from the air into the ocean is equal to the amount of CO_2 stripped from seawater in Step 2, resulting a closed loop on the TA-DIC-pH plot that returns the seawater back to its starting conditions.

Empirical data exist for specific approaches to OAE (La Plante et al., 2023; Ringham et al., 2024) and OAC (de Lannoy et al., 2018; Eisaman et al., 2018; Kim et al., 2023) that confirm the general process flows shown in Figures 1, 2. In addition, ocean modeling of OAE has also confirmed this behavior (He and Tyka, 2023; Wang et al., 2023).

2 Discussion

From the description above, the “sponge” and “pump” labels for OAE and OAC, respectively, become clear: OAE durably absorbs additional CO_2 from the air into the ocean as added DIC, whereas OAC uses the ocean as a pass-through to pull CO_2 from air.

One important difference between OAE and OAC is the volume of seawater that must be processed per ton of CO₂ removed. For OAC, the amount of CO₂ that can be removed from the air using a given volume of seawater is the amount that exactly replaces the DIC that was extracted in the acidification and extraction steps (Steps 1 and 2 of OAC in Figures 1B, 2). A typical value for DIC in seawater is around 0.0025 M (Butler, 2019), meaning that OAC can remove 0.0025 moles of CO₂ from the air per liter of seawater, corresponding to approximately 9,089 cubic meters of seawater that must pass through the OAC system per ton of CO₂ removed from the air. The amount of seawater that must be processed by OAE for a given amount of CO₂ removal is much less than for OAC. Because OAE generates alkalinity from the salt in seawater, the amount of seawater needed for OAE is governed by its NaCl concentration (approximately 0.5 M) rather than the DIC concentration (0.0025 M). Practically, OAE can generate a maximum of approximately 0.25 moles of alkalinity per liter of seawater, with the factor of two less than the salt concentration due to the need for remaining conductivity in the outgoing salt solution. For OAE, the molar ratio of CO₂ removed to added alkalinity ($\Delta\text{CO}_2/\Delta\text{TA}$) depends on ocean conditions, but a typical range is 0.7–0.8 (He and Tyka, 2023). These values lead to a range of 113 ($\Delta\text{CO}_2/\Delta\text{TA}=0.8$)–129 ($\Delta\text{CO}_2/\Delta\text{TA}=0.7$) cubic meters of seawater that must pass through the OAE system per ton of CO₂ removed from the air. Therefore, OAC requires $9,089/129$ – $9,089/113=69$ –80 times more seawater per ton of CO₂ removed than OAE. It should be noted that both OAE and OAC require the same amount of seawater to generate the acid and base used in their respective processes, but OAC must pump significant additional volumes of seawater into the system as part of the acidification and CO₂ extraction steps of the process. The need for additional seawater pumping will increase both equipment and energy costs (Eisaman et al., 2018; Eisaman, 2020).

Another difference between electrochemical OAE and OAC lies in their byproducts: aqueous acid (HCl) for this specific form of OAE and CO₂ gas for OAC. These products need to be used or stored in a manner that prevents a reversal of the CO₂ removal process, meaning that the CO₂ gas produced by OAC must be durably stored in a way that prevents its leakage to the atmosphere, and the H⁺ ions in the HCl produced by electrochemical OAE must be prevented from leaking back into the ocean. For HCl, this is most easily accomplished by using the acid in processes that result in its neutralization and the storage, disposal, or beneficial use of the resulting salts. In terms of the rate at which these products are generated, OAC produces one molecule of byproduct CO₂ for each molecule removed from the air, so removal of 1 GtCO₂/y using OAC would require the durable storage of 1 Gt/y of byproduct CO₂ gas. For the case of OAE, one molecule of HCl is produced for each molecule of NaOH that is generated, meaning that a typical range for ($\Delta\text{CO}_2/\Delta\text{TA}$) of 0.7–0.8 (He and Tyka, 2023) corresponds to $(1/0.8-1/0.7)=1.25$ –1.43 mol(HCl) generated per mol(CO₂) removed from the air, or 1.04–1.18 tons(HCl) generated per ton(CO₂) on a dry basis. Therefore, the removal of 1 GtCO₂/y using electrochemical OAE would require the neutralization of 1.04–1.18 Gt/y of byproduct HCl, on a dry basis. Electrochemical OAE generates aqueous acid with typical concentrations of around 0.5 M.

For the CO₂ generated by OAC, the sequestration of CO₂, either underground or in long-lived materials, will be required (NASEM, 2019). For underground storage, the mineralization of CO₂ into solid carbonates via reaction with silicate-containing rocks has been shown to be a promising avenue that reduces concerns about leakage and

durability (Matter et al., 2016; Snæbjörnsdóttir et al., 2020; White et al., 2020). To date, the maximum demonstrated rates of mineralization have been on the order of tens of thousands of tons of CO₂ per year. In scaling to injection rates of billions of tons of CO₂ per year, there is concern that slow reactions will limit the rates of mineralization (Tutolo et al., 2021). In a case where OAC and OAE may complement each other, research is ongoing as to whether this reduced CO₂ carbonation efficiency at the gigaton scale can be increased by pretreating, or co-treating (Awolayo et al., 2022), the injection formation with the acid generated by OAE. This approach aims to leverage the orders-of-magnitude increase in mineral dissolution rates and solubilities at low pH values to enhance the concentrations of carbonate forming cations. By enabling CO₂ mineralization and acid neutralization at gigaton-per-year scale, this may provide a pathway to enable the deployment of electrochemical OAE, and CDR approaches such as OAC, that require the durable storage of CO₂, at climate-relevant scales. That said, pre-existing zones of high permeability may localize injected acid, leading to increased mineral dissolution. This feedback is often termed “fingering” or “wormholing” (Szymczak and Ladd, 2014). Such effects must be minimized for acid pretreatment to realize its potential for CO₂ mineralization.

Additionally, through its ability to optimize feedstock properties, the acid generated in electrochemical OAE also potentially enables hybrid approaches with other CDR methods, including enhanced rock weathering (ERW), containerized enhanced weathering, and Biomass Carbon Removal and Storage (BiCRS). Similarly, the CO₂ generated in OAC can also be used in hybrid approaches that sequester CO₂ in non-geological locations such as concrete.

Despite the fundamental differences between OAE and OAC, some seeming similarities remain. First, with the addition of the equipment and energy needed for controllable contact between alkalinity and air, both OAE and OAC are equally capable of operating as closed or hybrid systems from the perspective of MRV. Second, as seen in Figure 1, the MRV required for OAE and OAC appear to be the same, but there is one important difference: OAE traces the CO₂ removal into a plume of TA-enhanced seawater with a typical level of DIC, while OAC traces the CO₂ removal into a plume of seawater with drastically reduced DIC and a typical level of TA. Care must be taken in the case of OAC to drive the process along the intended path on the TA-DIC-pH plot, given the almost complete loss of its buffer system after the CO₂ extraction step. For OAC, after Step 1 (acidification) and Step 2 (CO₂ extraction from seawater), the CO₂ has been removed from the seawater and this CO₂ must be durably sequestered. It is important to note, however, that this is not the CO₂ relevant to MRV. Rather, as shown in Figure 1B, the MRV required for OAC is performed on Step 4 (CO₂ extraction from air into the ocean) that occurs after Step 3 (alkalinity restoration). Just like the MRV for OAE, OAC must quantify the amount of CO₂ that flows from the air to the ocean to restore equilibrium. As an example, even if one ton of CO₂ is removed during Step 2 of OAC, if only half a ton of CO₂ is removed from the air in Step 4, perhaps because of alkalinity downwelling from the surface ocean prior to equilibration with the air, then only half a ton of CO₂ removal can be claimed. For both OAE and OAC, since the timescale for air-sea gas exchange is longer than the characteristic timescale for dilution, most of the CO₂ removal will occur far from the point of dispersion. As a result, MRV will rely on direct measurements of the seawater carbonate chemistry in the vicinity of

the outfall (Cyronak et al., 2023; Schulz et al., 2023) combined with ocean modeling to estimate CO₂ removal beyond the range of direct detection (Fennel et al., 2023).

In addition to developing MRV for the measurement and verification of CO₂ removal, MRV methodologies must also be designed to quantify the environmental and ecological impact of CDR interventions, a subset of MRV sometimes labeled “eMRV.” For example, both OAC and OAE release seawater with elevated pH back to the ocean, resulting in a mixing zone near the point of dispersal where the pH is at its maximum before decreasing as it diffuses away from the mixing zone and mixes with untreated ocean water. Experiments are underway to determine the effect of these changes in carbonate chemistry within the mixing zone on marine ecosystems, with some experiments starting to report results (Gately et al., 2023). The results of these experiments will inform eMRV measurement strategies and establish safe bounds of operation for OAC and OAE.

Scaling CDR to gigatons of CO₂ removal per year is a daunting challenge that will almost certainly require the deployment of multiple complimentary approaches. A major thrust of research and development in the next few years should focus on the potential for hybrid approaches, such as electrochemical OAE coupled to CO₂ mineralization, ERW, or BiCRS, or the coupling of OAC to CO₂ utilization in materials such as concrete. Determining the optimal times and places for deploying various CDR methods will require a clear understanding of their fundamental operating principles and relative merits. Such an understanding and direct comparison for electrochemical OAE and OAC has been lacking to date but is especially important given their superficial similarity in that they both electrochemically generate acid and base from salt. In this Perspective, I have aimed to provide a clear framework for comparing these two promising electrochemical mCDR solutions.

Data availability statement

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to the corresponding author.

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ME: Conceptualization, Funding acquisition, Writing – original draft, Writing – review & editing.

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Conflict of interest

ME is Co-Founder and Chief Scientific Advisor at Ebb Carbon, Inc.

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