Cover Sheet

Initial Exploration: Free Floating C02 Fixation Catalysts

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Abstract:

Climate change as an inextricable and dire threat to humanity is well established. The primary problem stems from an excess of carbon dioxide in earth's atmosphere and oceans causing a gradual increase in average temperature. With this there arises an immediate need for the alteration of this carbon dioxide into differing chemical states. Herein we perform an initial exploration of adapting the current field of "photocatalytic CO2 reduction" towards a mass geoengineering solution. We suggest that with the right strategy one could alter carbon dioxide into stable products at ambient temperatures, with no energy input other than sunlight, accomplishable by releasing CO2 fixation photocatalysts freely into the ocean. Such should be accomplishable, and it is possible that such could be done at low cost so as to affect a drastic reduction in free carbon dioxide within the Earth's oceans and atmosphere.

Keywords: Geoengineering; Climate Change; Photocatalytic Reduction; Carbon Dioxide; Ocean Science

Introduction:

Increasing global average temperatures due to increasing C02 emissions are well established. Beyond the acknowledgement of increase in C02 atmospheric concentration(1) and dissolved ocean concentration(2) discussion of this fact is beyond the purview of this paper.

Proposed solutions for reduction in free C02 are numerous. They include underground storage, or sequestration, of carbon dioxide(3). Mass plantation of new forests(4). And conversion of carbon dioxide into other products or “carbon fixation”, with many concentrating on commercially useful products(5).

C02 is a relatively stable molecule with high bond dissociation energy. As a starting basis this makes it relatively hard to break down, requiring larger application of thermodynamic entropy than more unstable molecules to alter. This challenge then translates into the high probability of requiring a large concerted effort, and thus a large amount of money, to reduce the free carbon dioxide content of the planet’s atmosphere and oceans.

The goal of removing C02 from the atmosphere, given its stability, should concentrate on means that require the least capital investment possible. Given this goal then, less inputs and human intervention in the process should be targets. The current proposal for most capital efficient C02 removal is the mass plantation of trees(4). While these trees might self sustainably grow at some point, high cost human intervention would be needed at the beginning, and large swaths of land as input for the forests to grow would be needed; and concerns have been raised regarding overestimation of reforestable land(6).

CO2 Reduction:

Our proposal primarily centers on the already well established field of photocatalytic “reduction” of C02 into other products; a subject that has received a lot of attention over the previous decades.

The principles of photocatalytic C02 reduction are straightforward and highly researched(7): Take a catalytic molecule that attracts and adsorbs carbon dioxide, and other possible reactants; attach a light absorbing dye to the catalytic molecule. The dye absorbs incident light, providing energy for a reaction of the attached molecules into the desired products. Please note reduction is often involved, but the charge of the ultimately desired products in this field is often arbitrary. Most often the environment chosen is an aqueous solution in a carefully controlled reactor environment, and the catalyst is used as a solid powder suspension therein. This in turn gives us our alteration; our targeted environment switches from controlled reactor to earth’s ocean. However to adapt this solution from a carefully controlled reactor environment to the environment of the world’s oceans presents multiple challenges.

The first challenge we address is whether the new target environment retains enough carbon for the target reaction, as well as if a reduction in carbon in the new target environment will lead to an overall reduction in free CO2 in the atmosphere. A study on carbon fixation utilizing increased phytoplankton nutrient seeding provides data showing a high concentration of CO2 saturating the ocean surface, equivalent to that already shown effective in many photocatalytic CO2 reduction papers. It also shows that carbon drawdown from the atmosphere increases dramatically as the ocean surface carbon content is reduced(8). Thus from a theoretical perspective we show removal of dissolved carbon dioxide can occur, and will lower both dissolved ocean content and atmospheric content.

The next challenge we address is the temperature needed for the reaction to take place. Fortunately for us low ambient temperature reduction of C02 is already an active area of research. Galinstant-cerium alloys have been found to catalytically react with dissolved C02 at “room” temperature(9) and very low onset potential; thus demonstrating high temperature environments are unneeded for the desired reaction type to take place. Further high conversion efficiency herein is important and will be discussed further down.

The next challenge we address is whether the desired reaction can take place in seawater at all. Fortunately successful photocatalytic reduction of dissolved C02 has already been demonstrated in polluted seawater using both carbon and copper variations of titanium-dioxide catalyst(10). Titanium dioxide variants are the most popular class of catalyst for C02 reduction, and this demonstration was also able to show catalytic activity under visible sunlight, the only illumination available for the reaction to take place in our proposal. While the catalytic activity in seawater was reduced from that in freshwater, we would note the productive activity of the catalyst is only of concern to us in regards to a more complex overall target that will be discussed further down below.

The previous also demonstrated reactivity with other C02 species; which is fortunate as a large portion of carbon trapped in the ocean is not in the form of carbon dioxide, but instead in the form of bicarbonate(11). Bicarbonate is a negative ion(HCO3-) and still fully energy favorable with photocatalytic reduction, including in open seawater. It is also a fully valid target for overall goal of lowering free carbon dioxide species in the environment, as it both recycles carbon dioxide back into the atmosphere and is reduced to carbonic acid, another problematic product that reduction of free CO2 aims to solve.

Thus a body of research establishes that the idea of a colloidal photocatalyst working to reduce C02 in ocean water is mechanistically feasible. While confirmation of any theoretical catalyst to be used in this scheme will have to be done, much larger challenges that the reaction taking place are of greater concern, and are discussed below. However we note that ambient environmental condition catalytic reduction of C02 is possible should not come as a surprise. Photosynthesis from plant forms, including phytoplankton already in the ocean, are a primary carbon sink source and already alter carbon in the desired environment without human intervention(12). The goal of a synthetic reaction in this case is to reduce inputs needed; nutrient limitations place a limit on phytoplankton and algae biomass(13) and a large body of research, ocean fertilization, has generally established this method as too costly and option, which is why they have already been discounted as a similarly scalable stratagem.

Environmental Considerations:

With the basic mechanical plausibility of the desired reaction taking place in the target environment established we now move onto complications encountered in environment, focusing on useful lifetime of the catalyst from a chemical and agglomeration standpoint. While the previously discussed research focuses on suspensions it should be noted the difference between a suspension and colloid is an arbitrary size cutoff of the suspended particles of less than a thousand nanometers for the latter, while any larger is the former, and we’ll be moving to the term colloid from hereon in.

A starting point for our proposal is general colloid stability in a marine environment. It is a vast and complex topic surveyed by Santschi(14). In this we find a large body of research from which to draw on, one of the most important conclusions being that stability in a marine environment is highly dependent on specific chemistry.

To narrow down our understanding of potential complications with catalytic stability we'll need to concentrate on research of ENPs, or engineered nanoparticles. While the following study is on one type of ENP, its findings support theories that predictive results can be generalized(15). Among the chief finding is that during deployment low concentrations should be heavily considered, as high concentrations of ENPs are the most sensitive attribute for sedimentation, which is the agglomeration of particles through adsorption until they lose buoyancy and sink, a primary cause of colloidal instability in marine environments.

We would caution against drawing too many inferences from these two papers and associated. They do not take into account specifically engineered photocatalytic particles, and so observed sedimentation rates are not one to one applicable over a long enough period of time. We see in previously cited sources much faster creation of products from specifically engineered catalysts than from bare TiO2 as one example, including removal of dissolved natural organic material dependent on chemistry (NOM)(16). This should drastically decrease sedimentation rate for our proposed catalyst in comparison, as adsorption of particles until sedimentation on bare TiO2 should occur much more quickly than for our hypothetical catalyst. Still, we felt it valuable to add this large body of research as what is and is not applicable to engineered catalysts from it will need experimentation to discern; especially with challenges such as a lack of photocatalytic activity being present at night most likely causing any experimental catalyst to react similarly to any other non catalytic colloid.

Beyond initial deployment conditions we can assume an extremely low concentration of ENP over the roughly three hundred and sixty square kilometers of available ocean surface. With previous research showing catalytic concentrations only start to degrade performance at >1 gram/liter we can consider the scalability of the proposed technique to only be limited to catalytic costs; as to reach this concentration even within the top cubed meter of the ocean surface would require three hundred and sixty billion tonnes of catalyst. While the entire ocean is not a viable target, as will be discussed later, we find below that even losing three orders of magnitude for viable spots we still have more than enough ocean to deploy the potential catalyst in without worry, assuming any reasonable effectiveness. Further we note that the assumption of targeting the top meter of the ocean is critical: it is the only area with the available sunlight and CO2 concentration for the target reaction to work.

If a catalyst is to be deployed then it in and of itself cannot be toxic, or at least very minimally toxic, to flora and fauna. For this we encounter an opposing effect compared to long term stability. Larger particle size sediments more quickly than smaller particle sizes, a result that should intuitively be hard to overcome as larger particle sizes would be more likely to encounter other solid particles over time. However larger particle sizes also have much less impact on the tested biological specimens (zebra fish embyros) (15). Specific research into this proposal may then have to rely on non toxicity of the particles themselves, rather than utilizing particle size as a means of reducing or eliminating harm to fauna. Non toxicity will be discussed further later.

For other long term stability concerns we may draw some basic inferences from previous research. Starting with the density of the water that our particle is to be suspended in, we see ocean surface density generally varies less by less than three percent(17) allowing a relatively stable target for particle buoyancy. We also find there's a relatively stable variation in ocean ph(18) and salinity(19). Combined with relatively stable catalytic activity from differing seawater samples(10) we can assume a relatively predictable and stable target environment.

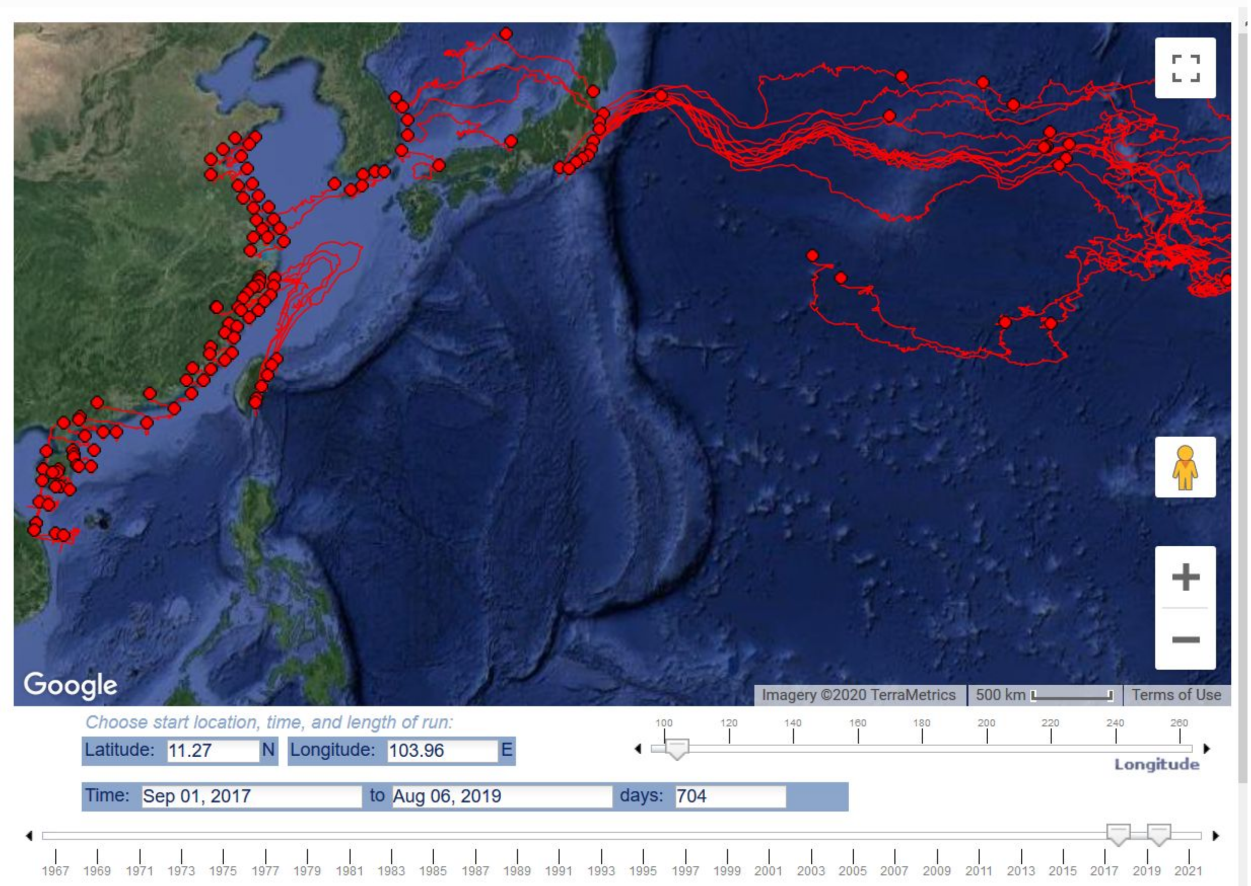
The challenge of long term particle stability and reducing or eliminating environmental harm of both the catalyst itself and any products it may create should be seen as a focus during exploratory research. While we address potential catalytic toxicity below most photocatalytic processes so far studied concentrate on products that would be harmful to the environment, assume a controlled reactor environment, and do not take environmental harm of the catalyst itself into account.

Cost Estimates and Deployment Concerns

With stability and mechanistic plausibility covered we now move onto the most important topic of concern, and the primary reason for this paper: cost. With no other human inputs required the only concerns we find for cost in this proposal is cost of engineered catalyst per weight, stability of catalyst in the environment, average sunlight the catalyst is exposed to over time, and reduction of CO2 over time per weight of catalyst. More generally we can sum these cost estimates to grams CO2 reduced per gram of catalyst deployed. We note deployment costs are hard to estimate as the deployment itself is releasing the catalyst into the ocean environment in a slow enough manner, and may ultimately be the same regardless of catalyst choice.

First we address lifetime concerns of deposition on shorelines. Here we rely on OSCURS(20) for simulation, and start each simulation just off the coast of a target area in the assumption that deploying near/from the coast is cheaper. An initial investigation of simulations shows dramatic difference in lifetime of current carried flotsam. Areas such as the coast of China tending to beach within less than a month, while areas such as the south western Baja California coast, and eastern Japan showing no beaching even after 365 days and 704 days of simulation respectively. See table 1. and fig 1. below for details.

Fig 1. Start dates, locations, and sim time all as shown.



Choose start location, time, and length of run:

Latitude: 11.27 N Longitude: 103.96 E

Time: Sep 01, 2017 to Aug 06, 2019 days: 704

Table 1.

Sims	**Location sim start**	**Average Time lasted**
40	Eastern China/Northern Vienam	>1 month
10	Eastern Japan	704 days+
5	Eastern Taiwan	>2 months
^Sep 2017 start date^		
10	Eastern China	>1 month
^March 2017 start date^		
10	Eastern China	>4 months
^June 2016 start date^		
10	Southern China	>1 month
10	Eastern Japan	365 days+
10	East Baja California, Mexico	365 days+
^December 2016 start date^		

Going into more detail, we also see change in year or month of simulation start having relatively little effect. While this is hardly a comprehensive list of viable deployment sites or initial deployment strategies, as all simulation start areas are near the coast, but not from rivers or other waste areas directly, it never the less reveals valuable information. Specifically that release areas, even beyond initial deployment, are not fungible, and will have to be taken into account in testing and any bidding processes. It also reveals that long term deployment of catalysts from the perspective of losing such by washing ashore is a challenge that can be fully overcome.

Next, cost of catalyst per weight we found to be difficult to estimate. Most recent studies of photocatalytic reduction already concentrate on low catalyst cost. We begin with an exemplar bulk material cost, and we find titanium dioxide at around US $3k per tonne averaged over the world(21), with many base material costs useable in proposed photocatalysts within the same order of magnitude, while others are far beyond that. What we found harder to estimate is manufacturing cost. While it is beyond the purview of this paper to estimate manufacturing quotes we find that both low complexity of manufacturing and low material cost is being taken into account in some proposed photocatalysts already(22).

We next consider the amount of CO2 reduced over time. This has been a focus of photocatalytic research, and so multiple strategies can be found. Herein we will be taking the micromole reduced per hour per gram of catalyst, or ***μmol h^{-1} g^{-1}*** as a general measure of efficiency.

For the highest photocatalytic efficiencies, we find 47k μmol h^{-1} g^{-1} with CO as the primary product as the highest efficiency per hour we could locate(23), which was done under visible light and ambient temperature. Converting to CO2 reduced per gram we find 2 grams of CO2 reduced per hour (of sunlight) per gram of catalyst. It should be noted that while carbon, a primary material for the catalyst, is incredibly cheap currently carbon nanotubes and graphene are both costly to manufacture. Ruthenium, used as a light sensitizer, is also highly expensive, though far cheaper dyes have been demonstrated in other studies. Other high efficiency photocatalysts have been reported such as 25k μmol h^{-1} g^{-1} for cobalt oxides on silica(24) though we would note this used sacrificial material and cobalt is as of this writing US $35 per kg, though this particular material is highly variable in cost.

That being stated we have hope low cost catalytic materials may soon perform in a similar range as these high cost ones, or the higher cost catalytic components such as carbon nanotubes might come down dramatically in price.

For an example of reachable price we use copper doped titanium oxide. While we mentioned this earlier as an average quote, if we take the lowest quote we find titanium oxide as low as $1,605 a ton, copper powder at $2,800 a tonne from(25), and a simple single step doping process with particle size control already demonstrated(26), thus while we'll not provide a specific quote for this exemplar we have hope that catalytic cost might prove exceedingly low.

But how well can previous photocatalyst research predict efficiency and cost? So far we have mostly discussed catalysts designed exclusively to do one thing: produce commercially valuable products. Most commonly carbon monoxide and further derived products. However most of these products are undesired for our case, as they would prove harmful to the environment and useless as a means of income. However other products, such as carbonaceous products, have been found to be selectively produced at lower onset potential(9). This suggests other products may prove to be an even more efficient catalytic target. Within the environment there is also an abundance of ions that are also potentially helpful reactants(27).

As for the catalytic degradation in the environment part, we find that a subject too complex to estimate. Photocatalytic degradation over time is incredibly variable, with some lasting indefinitely from the perspective of brief laboratory tests that found negligible degradation over the run. We again note the subject of catalytic sedimentation is also beyond the specific purview of this paper, as we could find no practical long term estimates of photocatalytic sedimentation rates in seawater, and as noted earlier sedimentation rates of ENPs are highly dependent on chemistry.

With the previous in mind we can make a cost estimate from a reasonable set of assumptions. The formula for which is incredibly simple and straightforward.

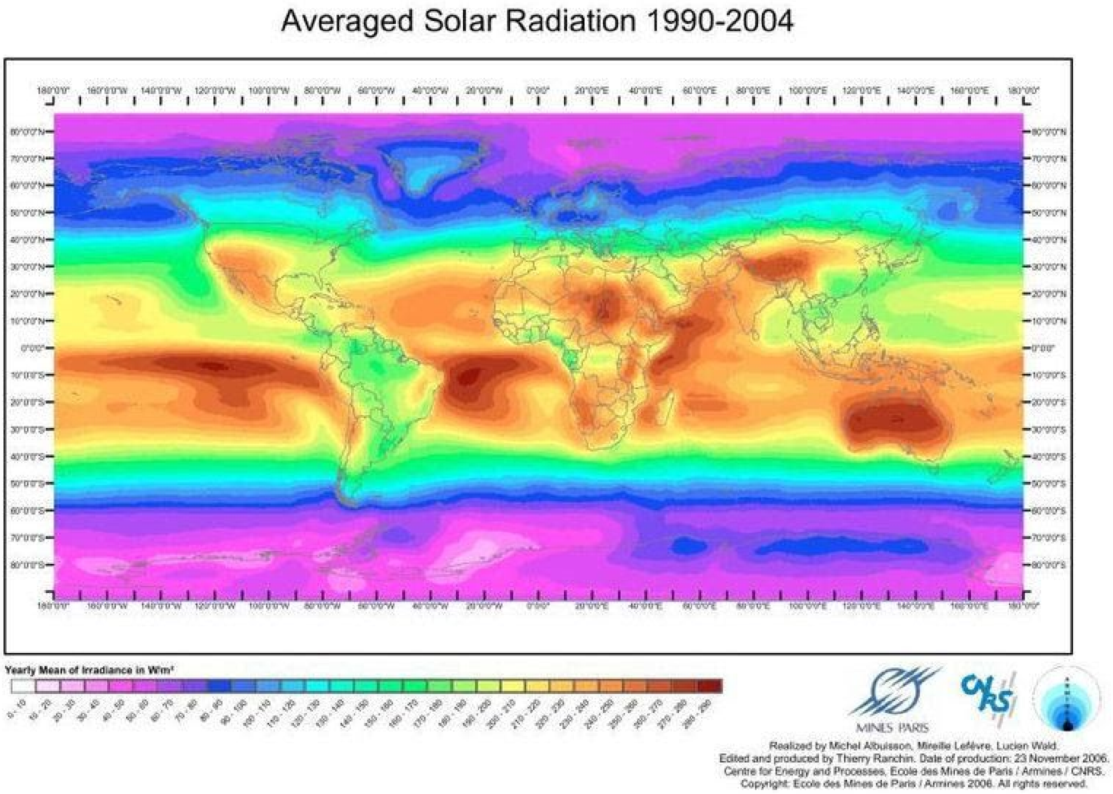
$$((T * Rw) * Sf) \div Wc = cRw$$

Eq. (1)

Where *T* represents the average effective lifetime of the catalyst in days, *Rw* is the carbon reduction (fixation) over the course of a day in units of *w*, *Sf* is the average fraction of catalytically effective sunlight during a day the catalyst receives, *Wc* is the unit *c* cost per deployed weight of the catalyst, and *cRw* is the cost per weight of carbon reduced.

For *Wc* a simple, we'll assume a high efficiency catalytic bulk material process might produce catalysts at USD $10,000 per ton. For *Sf* we found fig 2 below, which notes a high variability solar resources over the ocean; however we'll assume an *Sf* of five effective hours a day for the following examples. We note catalytic efficiency may easily prove better than this for sunlight, and ocean current targeting can improve sunlight availability significantly, but a bad case scenario will suffice for now. For *Rw* we'll assume in this example eliminating 2 grams CO2 an hour per gram catalyst for a total of 48 grams a day maximum

Fig. 2 acquired from (28)



Averaged Solar Radiation 1990-2004

Realized by Michel Albuisson, Mireille Lefèvre, Lucien Wald.
Edited and produced by Thierry Ranchin. Date of production: 23 November 2006.
Centre for Energy and Processes, Ecole des Mines de Paris / Armines / CNRS.
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Plugging these into our equation we get 10 grams of CO2 reduced a day on average, we'll assume an *T* of 200 days for this example. Doing so, we get a price $5 per tonne of carbon reduced. This matches the cost per carbon reduced of mass tree plantation(4) if they manage an incredibly efficient trillion dollar total deployment cost. This hypothesis however would accomplish the target reduction over a much lower time horizon, and with an unlimited scope for deployment as opposed to a severely limited one. As another hypothetical example, replacing the above with $5k per tonne catalyst an average lifespan of 400 days, we need not even increase efficiency or sunlight to hit a cost of $1.25 per ton reduced. With the 40 billion tonnes emitted annually(29) stopping, and reversing, global warming could cost $50 billion a year on average.

But are these cost targets reasonable? We first note that even if the first hypothetical target were matched, this is already lower than most countries that have such laws in place set the cost of carbon emitted per ton(30). While our examples is based on the catalytic efficiency that currently relies on a high cost material, improvements are almost certainly possible in regards to cost. One component of that catalyst relies on CNTs, however CNT(carbon nanotube) production has been hypothesized to be producible in vast quantities at as low cost as USD $660 per ton(31). With material such as phosphorous and carbon monoxide (already a demonstrated as an efficient product in multiple photocatalysis papers) being potentially low in cost all that would be needed to match our highest performing photocatalyst(21) at low cost is the replacement of ruthenium. Combined with optimizations of *T,* which at the moment may be near indefinite, optimizations of *Sf* which are easily accessible, and still relatively unknown potential for *Rw* we not only find the above targets not only reasonable if a concerted, multi team effort were made, but find it entirely reasonable that improvements over those targets may be found.

With the potential for a low cost efficient catalyst suggested we briefly cover potential catalytic toxicity. As an example, cytotoxic effects were found with absorption of μm scale CNTs(32), it also found agglomeration of CNTs towards that length, similar to other ENPs, was found in a marine environment, though we do note that CNTs in particular showed greater dispersion characteristics than other studies ENPs. However we note that doped, specifically engineered CNTs did not show any toxic effects and were found to be safe for in vivo medical procedures(33). Thus we find cytotoxicity of any potential catalyst, and in turn product, is heavily dependent on specific chemistry.

Our proposal then suggests it is possible for a photocatalyst targeting CO2 fixation to be deployed into the open ocean, in a similar manner to ocean fertilization, to affect a reduction in atmospheric CO2. Further it suggests that a concerted effort to shift photocatalytic carbon reduction research to such a geoengineering effort could produce a highly effective, relatively low cost solution for such.

However for this to succeed we find varying goals: Low cost, very little to no negative environmental impact from both the catalyst itself and products, extensive marine environment active lifetime, and efficient CO2 reduction capabilities. Within the very large possibility space already present and active in so much research we find the possibility that a catalyst that meets all these requirements to be high enough to warrant investigation.

Once a catalyst is found manufacture and deployment may prove easier and faster than any other proposed solution we could find. Manufacture of ENPs is well established as an industry, and deployment could, in principle, be the same as pollution or at worst ocean fertilization; so while deployment areas and specifics are of direct concern deployment costs may prove low.

For a few last notes, we have noted that while this solution would fall under the term “geoengineering” we have observed a large negative bias towards this term. We thus encourage developing a different term for this strategy, as well as other possible strategies. We propose “climate control” as an example of a new friendlier term, in order to affect more positive coverage. We also encourage any potential deployment of this solution to focus on clear, unambiguous communication and goals, which should lower any potential anxiety over its effects(34).

Finally, we do note that while this proposal has concentrated on releasing catalysts into the ocean surface as a long term means of CO2 reduction, a cycling of surface seawater into and out of a hypothetical reactor could, in principle, be used to create economically viable products with some alteration of the proposed strategy. Kelley, 1970(35) confirms dissolved ocean surface CO2 saturation, and also gives a useful starting point for differing dissolved CO2 seawater areas should a reactor setup as hypothesized be attempted.

Conclusion

We have shown that there is a significant probability that an artificial photocatalyst exists which could be released into the marine environment and affect a slowing, stopping, and even reversal of climate change due to greenhouse gases in a comparatively inexpensive and controlled manner within a short time horizon. We suggest that while this carbon fixation solution will, in all likelihood, require large initial resource costs in personnel and capital for investigation and optimizations, results may well be worth the cost. We also noted that generation of revenue products from CO2 catalysis could draw inspiration in method from this proposal.

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