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Earth System Dynamics Discussions

Interactive comment on "Ideas: Photoelectrochemical carbon removal as negative emission technology" *by* Matthias M. May and Kira Rehfeld

Anonymous Referee #2

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I have read the paper Photoelectrochemical carbon removal as negative emission technology by May and Rehfeld. The paper presents an interesting idea that is well suited to ESD. Overall the paper seems reasonable enough, although there are a number of places were the authors should be more specific. There also needs to be additional discussion of several specific points in order to provide a fuller context. The introduction also has a few statements that are opinion and not based in scientific analysis and need to be modified since this is a scientific paper. The idea presented is interesting enough without putting up artificial strawmen to knock down! With these modifications the paper will be suitable for publication.

Specific Comments:

Lines 7-10

The last sentence of the abstract (and some associated text in the introduction) contains some assertions that are not supported by the literature. These detract from the paper overall. Specifically:

First "should (i) rely on an unlimited source of energy (solar),", contains a number of implicit assumptions. "unlimited" implies that negative emissions should be maintained indefinitely. If taken literally this would eventually decrease CO2 concentrations below preindustrial levels! Not a goal I suspect the authors intend nor has this been proposed. If we posit an eventual goal of a zero carbon energy system, then the role of negative emissions is a finite one whose role is to "draw down" atmospheric CO2 levels until a "safe" leve is reached (e.g. UNFCCC). Therefore, an unlimited source of energy is not required, just a source that is sufficient for the amount of negative emissions that might be needed.

While of course it is important that any such process "result in a safely storable product", there is no evidence for the second portion of this statement, that this needs to be liquid or solid, not gaseous. There is a related misstatement later about geologic CO2 storage since, in fact, at the depths where CO2 is injected for storage pressures are high enough that CO2 is liquid. A substantial body of literature has verified that CO2 injection at a number of sites around the world, the most well known of which is likely the Norwegian Schlipner natural gas field, has shown that CO2 injection results in stable, trapped CO2. Again, this does not detract from the idea proposed by the authors, but statements in a scientific paper do need to be scientifically accurate! There is also a rather long history of CO2 injection for enhanced oil recovery, which has also provided substantial information on sub-surface behavior of CO2 (albeit in a condition not designed for 100% long-term storage).

Line 15. "therefore, the currently most feasible option appears to be the use of natural photosynthesis to generate biomass through afforestation or ocean fertilization. "

The authors do not appear to be aware of the literature here. First, it is not clear if ocean fertilization is feasible (experiments have not always provided support for this idea), nor are the side effects of ocean fertilization well enough understood.

The authors are deliberately ignoring geologic Carbon Capture and Storage (CCS), which is the leading option, along with reforestation, and related strategies, examined in scenarios to date. This is already being practiced today at a number of sites and there are no known technical barriers to large-scale application. While there may be legitimate concerns with this option, some of those concerns are also likely to apply to the option proposed by the authors. A more balanced discussion is needed (particularly since the authors later suggest geologic injection of the carbon-based liquids produced by their methods, which is inconsistent with the neglect of geologic CCS). A discussion of CCS and, presumably, contrast with the author's proposal needs to be added.

Page 1 Line 22. I am aware of course of the Heck et al. (2018) estimate, however much other literature does not come to the same conclusion (for example see the SSP scenarios, which are produced by models that integrate energy land and economics into a consistent projection framework). This not to discount the issue, but note that this is not a settled issue scientifically, and there are many nuances that prohibit such a blanket statement. For example, there is not one hard and fast estimate for the amount of negative emissions that are needed, this depends on the specific target and the rate that the world energy system is decarbonized.

However even taking the author's statement that 10 Gt/year of CO2 is needed, residue biomass sources (e.g. rice straw, corn stover, etc.), which do not require any additional land, could supply this amount by mid-century as crop production expands globally to support glorwing population and affluence levels. There is abundant literature on this point.

Page 2 line 10. "These could be stored in underground reservoirs such as depleted

oil fields, but also used as precursors for organic construction materials." The problem with using these liquids as precursors for organic construction materials is that these materials will eventually oxide and return carbon to the atmosphere. It is possible that this could still result in a long-term sink, but at only a fraction of the initial flow. (There are a number of papers that examine the substitution of wood for concrete as a building material that can be used for references and guidance here and should be cited.)

Further, if the authors are proposing use as construction materials, than the energetic cost of converting liquids to solids should be discussed.

Page 2 Line 17. As noted above this statement is not correct. First CO2 is not a gas at the relevant pressures, and there is abundant literature and data that indicate that CO2 can be safely injected into formations where it appears to be trapped on a long-term basis. (Some of these statements may indicate a misunderstanding of how deep CO2 injection works. Injected CO2 does not form some sort of large "bubble", but instead moves into rock pore spaces where chemical and physical reactions then occur.) .

Page 3 Line 6. Reference needed for what "previously largest project for solar electricity production in the Sahara desert" since this is a moving target!

Page 4, line 10. This "Unlike solar energy, however, large-scale mechanical wind extraction from the atmosphere might be limited. (Miller et al., 2011)" is another strawman that is irrelevant. We do not need unlimited energy for this process, just sufficient, affordable energy. In some locations that might best be supplied by wind, some solar, and in others biomass.

Page 4 line 16 - "While we estimate the costs for photoelectrochemical CO2 removal to roughly 65 Eur per ton" (add reference to the appendix where the details are given)

page 4. What is missing here are a discussion of transportation and storage costs. For any system that is sufficiently large scale, pipelines would likely be the most efficient mechanism for transport. Costs of pipeline transport of CO2 are well known (this is already a common practice) as also, of course, petroleum products, so a direct comparison of costs per km per tonne of C should be feasible and would be very useful. There are unlikely to be that many suitable storage locations in the places with the highest solar irradiance. So long-distance transport would likely be necessary. This is not necessarily a huge barrier (current scenarios with CO2 with CCS envision long-distance transport of CO2, and the world currently transports large quantities of petroleum across the globe - this would actually be a useful comparison. Are we talking about a volumetric transport rate per year that is on the order, much less than, or greater than current international shipments of oil by tanker? See the data in the "Review of Maritime Transport" series by the UNCTAD).

The volume needed for storage is a significant potential issue for this proposed negative emissions mechanism and needs to have more discussion and some order of magnitude estimates.The total storage volume over, say a 50 to 100 year period should be estimated by the authors and compared with potentially available storage locations. The authors mentioned depleted oil fields - the problem here is the same one mentioned by the authors as an issue for CCS with CO2, liquids injected into the oil fields may come to the surface (given that oil already comes to the surface in production wells).

There will also be some cost for storage. Injection into geologic formations (such as old oil wells) requires compression and pumping. The cost of this is well known and should be added to the cost estimates in Appendix B.

One advantage of CO2 sequestration is that CO2 is harmless, although it does pose a danger at concentrations large enough to exclude oxygen (e.g. this is the sort of low probability high consequence event considered by regulators in developing CO2 pipeline, and ultimately CCS, regulations). The potential health and environmental issues associated with the proposal should also be mentioned. One would presume that there could be environmental damage due to spills of these hydrocarbons from pipelines and/or leakage into groundwater or to the surface from storage sites. Are

there likely to be tradeoffs between carbon density, transportation issues (e.g. low enough viscosity to be transported by pipeline), and low toxicity or other environmental impacts?

Page 5 the statement “these idealised assumptions result in a maximum electronic photocurrent density of je âĽČ26mAcm−2” needs support. How was this value obtained. What assumptions were used to obtain this specific value.

Similarly here: “It follows that ideally ca. 19% of the incoming solar photons transform a CO2 molecule to the liquid,” as the percent value does not seem obvious or related to the above values.

The values seem reasonable, but it is important to state one’s assumptions so that readers know exactly what is being referred to here.

It is also important to note that these values are ideal cases. This needs to be emphasized in the main paper. This is fine, and is a reasonable starting point for bounding the possibilities, but it needs to be made clear that this is just the theoretical limit, which is unlikely to be approached commercially. (If we could approach the efficiency of nature, it would be a great starting point.)

This “ηe = 0.5” seem backwards, as the variable is described as “how many electrons are consumed for the formation of one product molecule from CO2 and water”, which would suggest the value should be 2, not 0.5. But perhaps the definition was not written correctly?

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