Addendum to: Understanding environmental trade-offs and resource demand of direct air capture technologies through comparative life-cycle assessment

Addendum to: *Nature Energy* https://doi.org/10.1038/s41560-021-00922-6. Published online 28 October 2021.

https://doi.org/10.1038/s41560-023-01312-w

Kavya Madhu, Stefan Pauliuk, Sumukha Dhathri & Felix Creutzig

In this Addendum, we clarify the assumptions made when constructing the product system for the high-temperature aqueous solution (HT-Aq) direct air capture (DAC) unit.

The HT-Aq DAC product system uses information published by Carbon Engineering, but we made several distinct changes to that system—in particular, in the calcination step—resulting in a simplified hypothetical new product system. Carbon Engineering's DAC prototype includes an oxy-fuel-fired circulating fluid bed calciner that uses an in-bed combustion process to convert calcium carbonate ($CaCO_3$) pellets into CO_2 and CaO. Such a calciner design allows for the co-capture of CO_2 streams emitted during the processing of $CaCO_3$ and the heat supply via combustion of natural gas inside the calciner. This co-capture of fuel-related carbon reduces the carbon emissions from the combustion of natural gas in the calciner and comes at the expense of additional demand for CO_2 compression, liquefaction, transport and storage.

Table 1 | Calculation of the carbon capture efficiency (CCE) for HT-Aq DAC with co-capture of combustion-related CO_2. The case without co-capture is reported in the paper.

System type:	HT-Aq DAC without co-capture	HT-Aq DAC with co-capture	Formula/source
Variable (unit, *symbol*)			
Atmospheric CO_2 captured (t, *a*)	1.00	1.00	By definition
Fuel CO_2 released during combustion (t, *b*)	0.28	0.33	HT-Aq DAC without co-capture: main paper and Supplementary Data 1, Co-capture case: Keith et al. (2018)[1], Figure 2: 112 t CO_2 captured per hour, 13.4 t CH_4 burned per hour, stoichiometry gives 0.329 t CO_2 (combustion) per t CO_2 captured
Amount of methane required (t, c)	0.10	0.12	HT-Aq DAC without co-capture: main paper and Supplementary Data 1, Co-capture case: Keith et al. (2018)[1], Figure 2: 112 t CO_2 captured per hour, 13.4 t CH_4 burned per hour
Methane supply chain emissions (t, *d*)	0.10	0.12	HT-Aq DAC without co-capture: main paper and Supplementary Data 1, Co-capture case: Scaled up from the case without co-capture using the methane flow *c*.
CO_2 vented (t, *e*)	0.28	0.00	By definition
Fuel CO_2 captured for storage (t, *f*)	0.00	0.33	By definition
Total CO_2 captured for storage (t, *g*)	1.00	1.33	Total CO_2 captured for storage = Atmospheric CO_2 captured (*a*) + Fuel CO_2 captured for storage (*f*)
Lifecycle CO_2e emissions to capture 1t CO_2 (t CO_2e, *h*)	0.54	0.24	HT-Aq DAC without co-capture: main result (see SD.1, Table F1.a, cell J4) Co-capture: Lifecycle CO_2e emissions to capture 1 t CO_2 (*h*) = Lifecycle CO_2e emissions to capture 1 t CO_2 (*h*, modelled case) – methane supply chain emission (*d*, modelled case) – fuel CO_2 captured for storage (*f*, co-capture case) + methane supply chain emissions (*d*, co-capture case)
Lifecycle CO_2e emissions to transport and store total captured CO_2 (t CO_2e, *i*)	0.05	0.06	HT-Aq DAC without co-capture: main result (see Supplementary Data 1, Tab F, Table2.a, cell I40) Co-capture case: Scaled up from the case without co-capture using the capture flow *g*.
Life cycle CO_2e emission to capture, transport, and store 1 t atmospheric CO_2 (t CO_2 e, *j*)	0.59	0.30	Total life cycle CO_2 emission to capture 1 t atmospheric CO_2 (*j*)= Life cycle CO_2 emissions to capture 1 t CO_2 (*h*) + Life cycle CO_2e emissions to transport total captured CO_2 (*i*)
CCE (%)	**41**	**70**	***CCE% = 100*(a–j)/a***

In Carbon Engineering's setup, it is physically impossible to separate the two CO_2 streams at the outlet of the calciner[1].

For the comparative LCA, we decided to model the HT-Aq DAC product system with different energy supply options and then compare it to the product systems of other DAC technologies with the same energy supply. Therefore, the calciner in the modelled product system for HT-Aq DAC has two CO_2 streams at the outlet, calculated from the separate mass balances for limestone decarbonization and methane combustion. For the energy supply baseline, the combustion-related CO_2 flow in the product system is not co-captured and stored.

This modification allows us to model various sources of thermal heat, and it ensures comparability between the different DAC products systems for different energy supply options, including the current electricity grid mix and natural gas supply. One must note, however, that such devices are currently not commercially available at large scale, and commercially available calciners retain both CO_2 streams. With the co-capture of CO_2 in the current design of Carbon Engineering, their prototype operates at a better carbon capture efficiency (CCE, see paper) than our modelled baseline product system without co-capture. We have now calculated the CCE of HT-Aq DAC where both CO_2 streams are retained and subsequently sequestered to be 70% (compared to 41% for our HT-Aq result without co-capture). These numbers are calculated from Table 1 below.

The separation of the calciner heat supply process also enables us to consider different types of electricity and heat supply in a potential calcination process with indirect heating, as done in the sensitivity analysis (see Fig. 2 of the main paper). In such an indirectly or jacket-heated calciner, the heat source (e.g., natural gas burners, electrical heaters, etc.) is located outside the shell of the calciner. Such a calciner can utilize sources of energy other than gaseous fuel, including high-temperature solar-thermal heat[2]. Assessing the large-scale technical and economic feasibility for utilizing an indirectly heated calciner in the current Carbon Engineering plant design is outside the scope of this paper.

To clarify the modelling used in the study, in the main paper, text in the Abstract, main text "Goal and scope of the comparative assessment" and "DAC carbon capture efficiency" sections, Methods "Life-cycle CCE" and "HT-Aq DAC" subsections, and Supplementary Note 4 and the cover sheets of Supplementary Data 1 and 2 have been modified. The updates are available in the HTML and PDF versions of the article.

References

1. Keith, D. W., Holmes, G., St Angelo, D. & Heidel, K. A Process for capturing CO_2 from the atmosphere. *Joule* **2**, 1573–1594 (2018).
2. Greco-Coppi, M., Hofmann, C., Ströhle, J., Walter, D. & Epple, B. Efficient CO_2 capture from lime production by an indirectly heated carbonate looping process. *Int. J. Greenh. Gas Control* **112**, 103430 (2021).