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RESEARCH ARTICLE

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The carbon dioxide removal potential of Liquid Air Energy Storage: A high-level technical and economic appraisal

Abstract Liquid Air Energy Storage (LAES) is at pilot scale. Air cooling and liquefaction stores energy; reheating revaporises the air at pressure, powering a turbine or engine (Ameel et al., 2013). Liquefaction requires water & CO_2 removal, preventing ice fouling. This paper proposes subsequent geological storage of this CO_2 – offering a novel Carbon Dioxide Removal (CDR) by-product, for the energy storage industry. It additionally assesses the scale constraint and economic opportunity offered by implementing this CDR approach. Similarly, established Compressed Air Energy Storage (CAES) uses air compression and subsequent expansion. CAES could also add CO_2 scrubbing and subsequent storage, at extra cost. CAES stores fewer joules per kilogram of air than LAES – potentially scrubbing more CO_2 per joule stored. Operational LAES/CAES technologies cannot offer full-scale CDR (Stocker et al., 2014) this century, yet they could offer around 4% of projected CO_2 disposals for LAES and $< 25\%$ for current-technology CAES. LAES CDR could reach trillion-dollar scale this century (20 billion USD/y, to first order). A larger, less certain commercial CDR opportunity exists for modified conventional CAES, due to additional equipment requirements. CDR may be commercially critical for LAES/CAES usage growth, and the necessary infrastructure may influence plant scaling and placement. A suggested design for low-pressure CAES theoretically offers global-scale CDR potential within a century (ignoring siting constraints) – but this must be costed against competing CDR and energy storage technologies.

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1 Introduction

The need to address carbon emissions from fossil fuels, which are responsible for anthropogenic global warming, means a shift to variable renewable energy generation is anticipated. It is widely expected (Mathiesen et al., 2011) that this shift will entail a move to electricity as a major fuel for traditionally chemically-fuelled use cases, such as transport (typically via vehicle batteries) and heating (via heat pumps). As such, a major expansion of world electricity demand is predicted – in addition to growth anticipated from population increase and industrialisation (IEA, 2017).

In general, most fossil electricity is dispatchable – in that it can be deployed on-demand. As demand rises (falls) daily, coal and gas fired stations are fed with more (less) fuel, adding more (less) electrical power to the grid. With exceptions, including biofuels and dam hydropower, renewable electricity is typically not dispatchable. Solar and wind power can be fed into the grid only at the time of their production, and any excess may be wasted. In a low-fossils grid, this tends to create a situation where there is a risk of large-scale under-supply (over-supply) of electricity, principally depending on the time of day and season. In an effort to balance supply and demand, it is widely expected that very large-scale storage will be required – as part of a broader mix of technologies, economic incentives, and behavioral interventions.

A wide range of storage technologies has been proposed (Jülch, 2016). These vary in purpose, predominantly by use case (e.g., whether mobile or fixed), storage duration (sub-second to inter-seasonal), and capacity (from device batteries to grid-scale).

Liquid Air Energy Storage (LAES) (Ding et al., 2016) and Compressed Air Energy Storage (CAES) are storage technologies generally suited to mid-to-large scale plants, and medium duration storage (Morgan et al., 2015). The need for cryogenic or high-pressure storage means that the resulting energy storage medium is quite low density. By contrast, power-to-fuels is more energy-dense and stable; a denser and more stable fuel benefits seasonal storage. Very small CAES/LAES installations suffer inherent limitations to storage efficiency, due to area/volume scaling effects on capital costs and heat transfer. While use of these technologies for spinning reserve is possible (Luo et al., 2015), other technologies may be better suited to this use case (batteries, capacitors and flywheels).

LAES/CAES is well-placed to address perhaps the main issue facing renewable energy – bringing solar energy from day to night. Swanson's law (Carr, 2012) gives a steep learning curve for solar power costs, implying that it will become the cheapest of all current low-carbon sources. As such, it can be assumed that the primary challenge is therefore to carry this solar energy into the night. This is particularly the case in tropical latitudes, which experience near-constant top-of-atmosphere insolation during the year. Mid-latitudes have an additional challenge, which is to carry summer sun to wintertime. This, as discussed earlier, is perhaps best addressed by power-to-fuels.

Notwithstanding such peripheral complexities (geographic seasonal storage variations, heterogeneous generation mix, short-term grid balancing), the energy storage problem crudely reduces to bringing solar energy into the night.

As such, our case-in-point technologies of LAES/CAES appear suitable for scaling, as their low costs of maintaining storage (and consequential mid-term duration) is suited to handling both certain daily storage, and uncertain weekly variances. Furthermore, these approaches are based on decades-old underlying technologies, and seemingly lack the potential constraints that may plague other storage technologies – such as materials (e.g., batteries) or siting (e.g., pumped hydro – although particularly CAES designs are geology-dependent). Of course, doubts remain as to the relative economic merits of this approach (Jülch, 2016), and our analysis is not intended to "pick winners" among a wide range of promising storage technologies. Rather, we seek to appraise the technical and economic case for adding a carbon dioxide removal (CDR) by-product to the LAES/CAES processes, with consequential consideration of any resulting economic impact.

Specifically, we attempt to answer the following research questions:

1) What proportion of expected CDR requirements could be met using large scale deployment of LAES or CAES?

2) What would be the economic opportunity of adding a CDR by-product to the business model for these industries?

3) Could modification to LAES or CAES potentially provide a viable way to provide all required CDR services?

2 Background to CDR

The role of this section is simply to place our novel LAES/CAES proposals into a proper context, for readers less familiar with this area of research. It also introduces the engineering behind Direct Air Capture (DAC) systems (which have much in common with the CO_2 removal systems of LAES).

CDR is an umbrella term for a wide range of different technologies capable of removing CO_2 from the atmosphere-ocean-biosphere system (Kriegler et al., 2013). A comprehensive treatment is beyond the scope of this paper – but prominent techniques are briefly summarized below, for context and comparison.

Bio-energy with Carbon Capture and Storage (BECCS) (Muratori et al., 2016) relies on the capture of post-combustion CO_2 from conventional power stations, fuelled by biofuels. It is currently operating in large-scale facilities, on a pilot basis (by re-firing extant plant with biomass). Though widely-discussed, sourcing biofuels without releasing carbon is a major challenge. These emissions may come from agricultural and transport machinery, fertilisers, or land-use change. Furthermore, concerns remain as to whether BECCS can really be made into an economically viable energy source – or whether it is best regarded as simply a disposal technique (Fajardy and Mac Dowell, 2018). As such, BECCS may be regarded as a competitor to biochar (Sun et al., 2014) – which is a pyrolysis process yielding a chemically-stable form of solid carbon char for shallow burial (Gurwick et al., 2013). While exothermic, biochar production is often envisaged absent energy recovery (You et al., 2017). BECCS, as with all other gas-concentrating techniques, relies on disposal of the concentrated gas stream. This is typically expected to be conducted by injection into geological formations. Deep saline aquifers are one such choice, wherein CO_2 remains stable. Alternatively, basaltic rocks may be used – wherein CO_2 reacts chemically, binding the elemental carbon in the lithosphere for geological timescales.

Direct Air Capture relies on various chemical techniques to capture CO_2 from ambient air, for potential geological disposal. As such, DAC is very similar to the modified LAES/CAES processes we later propose. DAC typically relies either on a high-temperature process (typically calcining) to produce high-purity CO_2, or a low-temperature pressure/temperature/humidity swing to adsorb and release an enriched stream of CO_2. The approaches vary in their suitability for different geographies, and for different CO_2 use or disposal approaches. For example, humidity swing typically relies on very dry, desert-type conditions; calcining produces feedstock-grade streams for industrial use. As an alternative, novel approach, DAC may potentially rely on cooling air to the point where CO_2 desublimates (von Hippel, 2018). The resulting solid CO_2 can be compressed and handled as a liquid with standard technology, or the CO_2 can be rewarmed to the gas phase. In either case, it must then be disposed of – just as in the chemically-based DAC approaches. Such thermal CDR approaches potentially lend themselves to applications within CAES/LAES, which rely on thermodynamics.

Enhanced Weathering (EW) (Köhler et al., 2010) relies on the fact that a wide range of basic rocks react slowly with CO_2 that is either present in the atmosphere or dissolved in the ocean. By grinding, distributing and spreading these rocks in suitable geographies (e.g., onto shallow ocean shelves, or farmland), associated reaction kinetics can be manipulated so as to produce meaningful weathering rates for the decadal or centurial removal of CO_2. This process is distinct from the other CDR techniques listed, in that it is wholly ambient. No kind of handling of ambient or purified CO_2 is required to complete the process. EW may also be combined with various DAC or flue gas treatment approaches, to give an accelerated mineralisation step.

CDR costs projections vary widely, within and between methods. Synthesis papers provide a degree of helpful constraint (Fuss et al., 2018) – BECCS and DACCS (Direct Air Capture and Carbon Sequestration) costs projections range upwards from around 100 USD/t CO_2; EW and biochar range from 50 and 30 USD/t CO_2, respectively (albeit with less certainty). Some techniques, e.g., soil carbon, are poorly cost-constrained cited above – due to the presence of cost-negative implementations (e.g., by profitably improving soil moisture retention).

For clarity, all concentration CDR relies on a CO_2 storage stage, at extra cost over concentration costs. Elements include transport, compression (if required) and injection/monitoring. These costs depend partially on injection well location. There is an interplay between the costs of electricity transport versus CO_2 transport, varying with volume and distance, though an analysis of locations and transport costs is beyond the scope of this paper. Additionally, our first-order analysis does not calculate variances in disposal costs – as these are highly dependent on the detail of implementation, and may be relatively minor.

It is against this background of alternative technologies and costs profiles that we attempt to appraise CDR by LAES/CAES, in costs and scale.

3 Analysis and calculations

LAES and CAES rely, respectively, on mechanical refrigeration and direct compression of ambient air. This is ordinarily powered electrically (Kantharaj et al., 2015), but direct-drive technologies (hydro, wind) could conceivably be used. In the case of LAES, the air is cooled (typically to –196°C at ambient pressure). While both water and CO_2 are relatively minor components of air, their mechanical properties necessitate removal from the stream, to prevent equipment fouling. This removal process is ordinarily achieved by adsorbing target molecules onto molecular sieves – but alternative techniques and processes are available for this purpose. As such technologies are inherent to LAES, we do not divert ourselves with a detailed discussion of their differences. Suffice to say, we assume that (with reasonable modifications) the CO_2 streams internal to the LAES process can be captured by the existing plant and turned into a stream of acceptable purity for carbon capture and storage (CCS) – it being an approach not requiring very high purity CO_2. Similarly, such scrubbing technology can be applied to CAES – albeit at extra cost, as this is not a necessary part of the standard CAES process.

A more concentrated CO_2 stream is preferred for CCS – avoiding the costs of transporting and injecting extraneous materials. However, double-digit percentages of impurities pose no particular problem for subsequent steps.

LAES plants favor medium- to large-scale, as thermal losses are a function of surface area – which is minimised with fewer, larger containers. CAES has historically been combined with large geological storage reservoirs, although a containerised implementation was proposed by (the now defunct) Lightsail (Spector, 2017). CAES also benefits from thermal storage enhancements; these pre-cool (re-heat) compressed air, before (after) storage. Accordingly, similar impediments to very small CAES plant exist.

Maximum viable scale limitations may apply by dint of the benefits of co-location adjacent to waste heat sources (paper mills, steelworks, cement factories, etc.) – as both LAES and CAES can be thermally boosted, providing effectively over 100% round-trip efficiency. However, such opportunities will be minimal, in a high-renewables world. Further limitations of grid capacity apply; storage is often preferentially located near usage or production nodes. Location near usage offers resilience and allows storage of any on-site microgeneration; location near production nodes allows generators to restrict power flows through limited grid access points, which is important with variable and geographically-sensitive renewables. The largest storage facilities would, by contrast, have to be located on the main grid power lines, unless ultra-scale solar and wind farms are used. While acknowledging the need for such considerations in electrical system design we postulate a system optimised for CCS. This would consist of large LAES facilities, sited close to favorable geology. The availability of such sites close to high-power grid connections is a cost-influencing limiting factor – albeit one that it beyond the scope of this paper, as it relies on site-level appraisal. For clarity: The scale of deployment we envisage are far beyond those at which useable waste heat is available.

Such detailed utilities engineering discussions aside, we present below a “back of envelope” model for calculation of the potential for CDR integration into LAES & CAES –

considering the following simplifying assumptions.

First, we assume that solar is the overwhelmingly dominant energy source – not wholly unreasonable, considering the predictable and sharp falls in costs embodied in Swanson's law (Carr, 2012). We further assume that all energy used is electrical, and that all stored electrical energy passes through LAES or CAES. Note: This is not the equivalent of arguing that LAES/CAES is the only storage approach in use; applications such as electric cars are expected to be important and may require multi-step storage. For example, an electric car may charge its battery from solar energy at night, via LAES storage. As such, multiple round-trip storage losses may be significant – but with potentially low input energy costs, the storage penalty may be economically less significant than the cost of installing alternative generation technologies.

We assume that storage is simple – i.e., that there are no waste heat sources for boosting its temperature above ambient (boosting its pressure, and therefore its capacity to do mechanical work). This assumption is realistic at large scales, where waste heat opportunities may be saturated.

In grossly simplified form, therefore, we assume that all energy is electrical, all storage is LAES/CAES, and all generation is solar. Furthermore, because storage of liquid or compressed air in large manufactured containers or rock formations is inexpensive, the costs of intraday LAES are assumed to be dominated by the power in/out calculations, not the container costs.

By making these simplifying assumptions we aim only to provide a first-order technical and economic feasibility study of intraday storage.

4 LAES and CAES calculation introduction

In this section we calculate: The total mass of CO_2 that could be removed by near-global adoption of LAES/CAES with CO_2 capture; the degree to which this address anthropogenic CO_2 removal requirements; and the related market size of this effort. As the basis for our calculations, we assume:

(1) Large-scale LAES/CAES with CO_2 capture begins when the atmosphere has reached ~450×10^{-6} CO_2 (by volume) – which is the expected value by approximately 2040, depending on various emission scenarios;

(2) Although the goal could be to return the atmosphere to pre-industrial CO_2 levels of 280×10^{-6}, a less demanding (higher) level will still be acceptably safe, which we take to be 350×10^{-6} (Hansen et al., 2008; Strahan, 2013);

(3) Industrial-scale LAES/CAES fully adopts CO_2 capture and sequestration, which recovers the vast majority of the CO_2 processed through the LAES/CAES system;

(4) LAES/CAES with CO_2 capture becomes the dominant means to temporarily store energy, as discussed above;

(5) Atmospheric mixing occurs quickly enough that no LAES/CAES system is ingesting the CO_2-depleted air of another LAES system's output – and instead is ingesting air with the worldwide average CO_2 fraction for that point in time (see further discussion below);

(6) CO_2 drawdown is linear with time (a simplifying approximation);

(7) We ignore changes in carbon cycle responses, as these require complex modeling. This introduces some uncertainty into our calculations and we address this via a sensitivity analysis after presenting those calculations. Nevertheless, these uncertainties do not impact our primary purpose of approximating the LAES/CAES scale required for meaningful CDR and comparing alternative systems. Essentially assumptions provide a meaningful way to estimate the upper limit for the contribution of LAES/CAES CDR.

The equation governing this carbon capture is then:

$$CC = N_{yr} GEU(t)\; f_{LAES}(t) f_{CO_2}(t) f_{rmv} / \varepsilon_{LAES} \rho_{LAES}, \quad (1)$$

where CC is the total amount of CO_2 in kg captured over a period of N_{yr} years, $GEU(t)$ is the time-dependent global energy use, $f_{LAES}(t)$ is the time-dependent fraction of global energy temporarily stored by LAES with carbon capture, $f_{CO_2}(t)$ is the time-dependent atmospheric fraction of CO_2, f_{rmv} is the fraction of CO_2 that passes through the LAES system that is removed, ε_{LAES} is the round-trip efficiency of LAES, and ρ_{LAES} is the energy density of LAES in kWh of energy per kilogram of air. In practice, ε_{LAES} and ρ_{LAES} are likely to be time-dependent as well, though only weakly so. For clarity, we do not write the LAES/CAES abbreviation out in full above – although the equation applies to both technologies with different values. For inputs into the above equation, we assume numbers appropriate for an upper limit calculation. Readers may easily scale our results based on their preferred input values.

4.1 LAES calculation

We chose N_{yr} = 100 years; $GEU(t)$ = 240 trillion kWh, which is the expected energy consumption during the year 2040 (EIA, 2013); $f_{LAES}(t)$ = 50%, constant with time, meaning half of all electricity used is first stored by LAES (accompanying round-losses must be accounted for); $f_{CO_2}(t)$ = 450×10^{-6} drawn down linearly to 350×10^{-6} (by volume) (ignoring carbon cycle response, as discussed above), equivalent to an average of 400×10^{-6} (equals to 610×10^{-6} by mass); f_{rmv} = 100%; ρ_{LAES} = 0.13 kWh/kg (Strahan, 2013); and ε_{LAES} = 70% (She et al., 2017). For these input numbers, $CC = 8.0\times10^{13}$ kg = 80 Gt of CO_2 captured in 100 years. Equation (1) is linear in all terms, with $f_{LAES}(t)$ by far the most uncertain. In fact, this term is essentially zero now and needs to be increased to the highest reasonable value for meaningful CDR. Thus a sensitivity analysis of Eq. (1) and this process essentially reduces to estimating a reasonable maximum value for $f_{LAES}(t)$ and linearly scaling that from the above estimate. For example, for $f_{LAES}(t)$ = 33%, CC = 53 Gt of CO_2 captured in 100 years. Table 1 explores the expected carbon capture per year (column 2) and per century (column 3) for $f_{LAES}(t)$ = 50%, 10%, and 1%, respectively. Table 1 also presents yearly revenue (column 4) and fractional decrease in LAES operating costs (f_{save}) under these three scenarios.

The first example (CC = 80 Gt) is 4% and the second (CC = 53 Gt) is 2.7% of the potentially 2000 Gt of CO_2 drawdown required, so maximizing LAES for CDR will not alone meet society's entire drawdown needs. Nonetheless, even at 30 USD per metric ton of captured CO_2, this represents a market value of 2.4 trillion USD (N.B.: disposal cost is additional). Operating such LAES systems over 100 years will process a total mass of 1.3×10^{17} kg of air, which is 2.6% of the atmosphere. We note that this atmospheric mass fraction further supports the assumption (number 5 above) that most air parcels will not be processed twice before they have mixed to the average atmospheric CO_2 fraction.

Under the assumption that the cost of LAES is dominated by power in/out calculations (as opposed to capital costs), then f_{save} becomes:

$$f_{save} = value_{CO_2} f_{CO_2}(t)/cost_{elec}\rho_{LAES}(1-\varepsilon_{LAES}), \quad (2)$$

where $value_{CO_2}$ is the value of captured CO_2 in USD/t, $cost_{elec}$ is the time-dependent cost of electricity. If we assume for this calculation that $cost_{elec}$ equals to the current wholesale cost of solar electric power of 0.077 USD/kWh, and the other variables and their values are as given above, then f_{save} = 0.6%. This is unrealistically conservative, however, because solar electricity generation prices continue to fall, and we can expect $cost_{elec} \cong$ 0.013 USD/kWh by the end of the century (Gerlach et al., 2015), which would yield f_{save} = 3.6%. A higher price for captured CO_2 would proportionally increase savings. For example, using the above numbers and $cost_{elec}$ = 0.013 USD/kWh and $value_{CO_2}$ = 100 USD/t, then f_{save} = 11.9%. Although not dramatic, these values of f_{save} are still significant – potentially displacing non-CDR plant. In addition, these savings are not dependent on the worldwide scale of LAES/CAES adoption, and are instead available to any LAES/CAES storage with incorporated CO_2 capture once there is financial support for carbon capture. With $value_{CO_2}$ likely increasing and $cost_{elec}$ likely decreasing over the next few decades, a large change is anticipated in the relative savings for CDR-equipped LAES operators.

4.2 CAES calculation

Because LAES cools air to below the sublimation point of CO_2, it is natural to consider coupling LAES with CDR. Analogously, CAES dramatically increases the pressure of stored air, which in turn affects the liquefaction temperature of CO_2. This facilitates thermal removal – although adsorption removal could also be used. In this section, we investigate to what degree the operating conditions of CAES would facilitate capturing CO_2. For the purposes of this investigation, we assume CAES with air stored at 70 atm of pressure at a temperature of 45°C. These conditions are similar to those at two currently operating CAES facilities (Kaiser, 2015) and coincidentally are near the critical point for CO_2, which is at 72.8 atm and 31.1°C.

While the critical point defines the upper limit of the liquid-vapor boundary for a substance, because CO_2 is not the dominant component of the atmosphere, we cannot simply cool this compressed air from 45°C to 31.1°C and obtain liquid CO_2. The liquid-vapor boundary is instead defined by the partial pressure of a substance, and for CO_2 at 400×10^{-6} by volume within air at 70 atm of pressure, the partial pressure of CO_2 is 0.0280 atm. At this partial pressure, the temperature at which liquefaction starts is approximately –114°C. As air is further cooled, more CO_2 liquefies out of the air, decreasing its partial pressure and requiring that the temperature drop further for more CO_2 to liquefy. At –129°C, approximately 90% of the CO_2 would liquefy, and at –140°C, approximately 99% of the CO_2 would liquefy. Because cooling requires energy and equipment complexity, there is a trade-off between greater cooling (for greater CO_2 recovery) and the increased energy cost of doing so. We find the trade-off is best balanced around –129°C, with 90% CO_2 capture (von Hippel, 2018).

The energy cost of cooling air in this manner was presented by von Hippel (2018). We modify Eq. (1) of that paper slightly (for simplicity), by dropping the term for passive thermal heat radiating – under the assumption that a CAES facility does not want to waste the heat of compression, which will be used later when the gas is expanded to recover the stored energy. The energy cost is then:

Table 1 LAES carbon capture and revenue

LAES	CC/y	CC/century	Revenue[a] (billion USD/y)	f_{save}[b]
$f_{LAES}(t)$ = 50%	0.8 Gt	80 Gt	24 to 80	0.6% to 11.9%
$f_{LAES}(t)$ = 10%	0.16 Gt	16 Gt	4.8 to 16	0.6% to 11.9%
$f_{LAES}(t)$ = 1%	0.016 Gt	1.6 Gt	0.5 to 1.6	0.6% to 11.9%

Table notes: [a] depends primarily on $value_{CO_2}$; [b] depends primarily on $value_{CO_2}$ and $cost_{elec}$.

$$EC = \frac{\left(|Q_{air}|(1-e_{air}) + |Q_{CO_2}|(1-e_{CO_2})\right)}{COP} + \frac{G_{sep}}{\eta} + E_{HE}, \quad (3)$$

where EC is the energy cost in joules of separating CO_2 from 1 m^3 of air; $|Q_{air}|$ and $|Q_{CO_2}|$ are the heat removed from the volume of air and CO_2, respectively; e_{air} and e_{CO_2} are the fractional recovery of Q_{air} and Q_{CO_2} by the heat exchanger, respectively; COP is the coefficient of performance for the refrigeration system; G_{sep} is the Gibbs free energy associated with the entropy change of separating CO_2 from air; η is the efficiency of the refrigeration system during CO_2 liquification; and E_{HE} is the energy needed to move 1 m^3 of air through the heat exchanger (von Hippel, 2018).

Figure 1 presents EC, the energy cost of removing CO_2 from the compressed air in CAES in GJ/t, as a function of dT – i.e., the temperature difference between the warm and cold air streams in the heat exchanger. EC values are presented for three levels of refrigeration performance (low, medium, and high values of COP) and two levels for the energy required to move air through the system (standard and advanced E_{HE}). The values of η match those that are currently achievable (η = 0.15) and potential available after development (η = 0.30 and 0.50). The values of E_{HE} match those that are currently achievable (E_{HE} = 940 J/m^3 at standard temperature and pressure) and expected from prototype technology (Koplow, 2010) (E_{HE} = 188 J/m^3 at standard temperature and pressure). The gray band running across the bottom of Fig. 1 indicates the estimated range of energy required to operate chemical-based direct air capture at 1.20 to 1.73 GJ/t (Stolaroff et al., 2008).

Figure 1 indicates that coupling the high-pressure stream of a CAES facility to additional equipment that cooled the compressed air to –129°C is tractable, though not competitive with the energy cost of the chemical-based DAC approach. Instead, even with advanced refrigeration with substantially more efficiency than currently available (the blue and green lines) and even with highly efficient heat exchangers with a temperature difference, dT, of only 1°C to 2°C, CAES coupled with refrigeration to remove CO_2 will require ~10 times as much energy to remove that CO_2 from the atmosphere as the chemical-based DAC approach. As such, it would not generally be competitive with LAES for CO_2 removal, both because of the energy cost and the substantial additional equipment that would have to be added to a CAES system for this additional cryogenic cooling. This does not mean that CAES with cryogenic cooling for DAC should be dismissed, however. If there are developments in CAES operations that cause these systems to be operated at higher pressure, or more importantly, if their stored air is held at lower temperature, the additional cooling for carbon capture would require less energy and be more economically competitive.

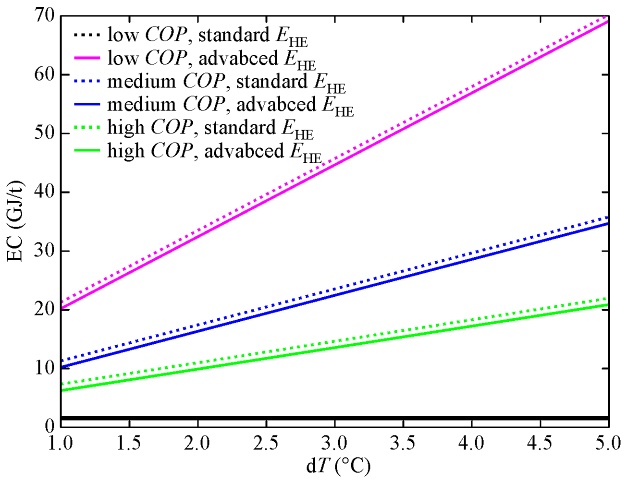

Fig. 1 Energy cost of removing CO_2 with CAES.

Finally, we return to the total mass of CO_2 that could be removed by near-global adoption of CAES (rather than LAES) with CO_2 capture. The primary functional differences for CO_2 capture between these two approaches are the amount of air they process, as a function of round-trip energy efficiency and the energy density of each. The ratio of air processed by CAES relative to LAES can be approximated as $\Re = \varepsilon_{LAES}\rho_{LAES}/\varepsilon_{CAES}\rho_{CAES}$. At present, studies of advanced CAES (Energy Storage Association, 2018) and LAES (She et al., 2017) both claim ~70% round-trip efficiency for these technologies. The energy densities are ρ_{LAES} = 0.77 MJ/kg and ρ_{CAES} = 0.12 MJ/kg (Strahan, 2013). CAES would thus process (0.7×0.77)/(0.7×0.12) ≅ 6.4 times more air and thus potentially capture 6.4 times more CO_2 than LAES. This would be a potential CO_2 capture of ~500 Gt over 100 years.

Table 2 presents the CO_2 capture potential as well as associated revenue for this approach to CAES, setting $f_{CAES}(t)$ = 50%, 10%, and 1%, respectively. Note that these impressive numbers would require CAES technology beyond what currently is in use, where carbon-based fuels are burned to warm the compressed air (stored solar thermal is a potential candidate, as is geothermal). Furthermore, CAES would require more extensive and expensive system modifications to capture CO_2 than would LAES, as outlined above. Further, geological implementations of this technology are more site-dependent, because the volumes of storage required for CAES are larger than those of LAES by the energy density ratio (here 6.4). LAES containers store air at near ambient pressure, which can be accomplished with large above-ground containers, whereas CAES stores at high pressures, thus currently utilizing large local geological storage (noting Lightsail's containerised alternative). For CAES to be the energy storage of choice, it would have to store ~6 times the volume as LAES. For these reasons, it is premature to estimate what fraction of the world's energy storage needs could be handled by CAES.

Table 2 CAES carbon capture and revenue

CAES	CC/y[a]	CC/century	Revenue[b] (billion USD/y)
$f_{CAES}(t) = 50\%$	5.1 Gt	510 Gt	150 to 510
$f_{CAES}(t) = 10\%$	1.0 Gt	100 Gt	30 to 100
$f_{CAES}(t) = 1\%$	0.1 Gt	10 Gt	3 to 10

Table notes: [a] assumes current value of $\rho_{CAES} = 0.12$ MJ/kg; [b] depends primarily on $value_{CO_2}$.

Despite these issues with bringing current CAES to a scale where it could meaningfully reduce atmospheric CO_2, it is worth pushing this calculation a bit further – in order to ask whether an alternative strategy for CAES would be to implement it at lower pressures, optimising the process for air volume maximisation. This could be done using sub-sea storage, using diving bells or polymer-based bags (Dorminey, 2014). This would provide planners the opportunity to swap cavern geographical limitations for restrictions on continental shelf availability (or in bag mooring technologies), and propose an energy storage solution highly optimised to recover CO_2. By reducing the pressure, the volume of air stored needs to rise. The following calculation shows how a global CAES system could be configured to maximise atmospheric recovery.

Starting with Eq. (1), we can calculate the density of compressed air commensurate with capturing 2000 Gt CO_2 over 100 years. This calculation yields $\rho_{CAES} = 19.1$ kg/m^3 at a pressure of 17.25 atm. For an average CO_2 fraction of 400×10^{-6} by volume during the drawdown, consistent with the above calculations, this requires CAES systems with a volume that, when multiplied by the number of breathing cycles, equals 1.7×10^{17} m^3. If we assume maximum use of CAES, with each system breathing fully once per day (consistent with the idea of bringing solar energy from day to night) over 100 years, this requires a global CAES volume of 4.7×10^{12} m^3. The required pressure, of approximately 17 atm, can be achieved by sub-ocean storage at just over a depth of 170 m – or by using an equivalent water column to regulate pressure, in a sub-surface cavern or mine. This immense volume is a challenge. If sub-ocean storage is used for example, the vertical height of an individual CAES unit may be of order 10 m. This corresponds to a container top-to-bottom pressure difference of 1 atm – although various segmented designs are possible. This height would then require that a surface of 4.7×10^5 km^2 be allocated to this storage – almost 0.1% of the earth's total surface area. So, while theoretically possible, it would require an immense engineering enterprise and a single-technology energy storage approach. This spatial demand is unlikely to make this approach competitive for full-scale CO_2 removal – although it may play a more modest part.

5 Conclusions

Our calculations allow us to draw three principle conclusions. First, the scale of LAES or current-technology CAES at a realistic maximum deployment is entirely inadequate to address the global CDR requirement – even ignoring carbon cycle response. Deployed at full scale for a century, it would be substantially below required levels (our crude calculations indicate a figure of 4% of projected disposals for LAES and $<25\%$ for current-technology CAES). This figure overlooks the geological restrictions on current CAES – as may be possible, were manufactured vessels used.

Secondly, and more encouragingly, the model indicates a total centurial-scale economic opportunity for CO_2 fractionation for CDR to be of the order of 2T USD (at 30 USD per metric ton of captured CO_2) for LAES and potentially of a substantially greater scale for CAES – assuming that CDR flows are demanded by the market at a price roughly equating to that of the other sources of CO_2 that are currently available or proposed. We do not present detailed economic benefit figures for CAES, due to uncertainty on costs of adding the CDR equipment. This economic opportunity equates to on the order of 20 billion USD/y revenue for the LAES industry and potentially much more for the CAES industry. This represents a notable contribution to the costs of operation of LAES and CAES plants – potentially serving to make them cost-competitive in circumstances where they may otherwise lose out to competing technologies. Notably, the relative costs savings (as a percentage of plant operating costs) become proportionally far larger as the price of input energy falls – moving from around 1% to around 10%, over the course of a few decades. While relatively small on the scale of the global energy system, the absolute revenue streams available may have a very significant influence on the design and siting of LAES and possibly CAES plants. Specifically, this approach (which assumes wide deployment of LAES and/or CAES) will favor very large plants, which would be located close to CCS infrastructure. However, we note also the costs of moving electricity – both capital costs and transmission losses. Consequently, we suggest detailed calculations are required, to identify the trade-off between gas and electricity transport – if few areas exist with both good grid connections and good gas disposal geology. Accuracy in calculations depends, to a significant extent, on maturation of the CCS industry.

Finally, we note the potential for a radical redesign of CAES, to effect CDR. Unlike LAES and current-technology CAES, this is based on technology that is at an early stage of development. We show that this could theoretically scale to act as the principal CDR technology. This would come at additional financial cost and requires immense subsurface storage capacity – although it is potentially competitive with other CDR approaches, in favorable environments.

In summary: Neither LAES nor current CAES technologies could provide all necessary CDR – but CDR revenue streams may influence strongly the prevalence and design of LAES and potentially CAES systems. A highly-modified CAES scheme, with unknown economics, is the only way to effect global scale CDR using these technologies.

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