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Supplementary information

The technological and economic prospects for CO_2 utilization and removal

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Supplementary Information

These supplementary materials contain further detail on methodology (S1) of deriving breakeven costs (S1.2) and scale estimates (S1.3) and a more detailed presentation of results (S2) underlying the scale and cost estimates for each of the 10 pathways with further information on TRLs and permanence (S3), supplementary figures (S4),further challenges to scaling (S5) and other potential pathways (S6).

S1 Methodology

S1.1 Overview

S1.1.1 Conventional Utilisation Pathways

Estimates for the cost and scale potential for the conventional utilisation pathways were informed by a scoping review for pathways 1 through 4 (chemicals, fuels, microalgae and concrete building materials), and by an expert opinion survey procedure (for pathways 1 through 5), both enacted using standard accepted frameworks [1-3]. See Ref [4] for detailed methodology supporting cost and scale potentials for pathway 5 (CO_2-EOR).

Scoping Review

For pathways 1 through 4, scoping reviews were undertaken by employing standardised search terms in the literature databases Scopus (n=2,846) and Web of Science (n=5,585) for peer-reviewed academic papers, published within the last 10 years. Forwards and backwards snowballing of references and citations were also employed. We excluded grey literature as well as non-reviewed conference proceedings.

Table S1 | Search terms

[urea or methanol] and [carbon dioxide or CO_2] near [utilisation or conversion or sequest* or carboxylat* or reduc* or capture] not [biomass or biochar or reservoir or fuel]
[polymer] and [carbon dioxide or CO_2] near [utilisation or sequest* or storage or carboxylat* or reduc*] not [adsorption or geopolymer or reservoir or *aerobic]
[methan* or DME or dimethyl ether or syngas or gasoline or diesel] and [carbon dioxide or CO_2] near [utilisation or conversion or capture or sequest* or carboxylat* or reduct*] and *econo*
[alga* or microalga* or algal biofuels] and [carbon dioxide or CO_2] near [utilisation or sequest* or storage or capture or fix]
[concrete or cement or industrial waste or aggregate* or building material or construction material] and [carbon dioxide or CO_2] near [utilisation or capture or sequest* or carbonat* or cur*] not [soil or biochar or reservoir or forest*]

Following the database searches, the abstracts of the relevant papers were screened for relevant content. Promising papers were read and data recorded pertaining to study type, CO_2 potential and cost estimates as they were presented in the primary literature. Papers were excluded if the quality of analysis was judged to be insufficient. The number of papers surviving each screen and included in this review is set out in Table S2.

Table S2 | Total number of papers identified by scoping review

Search	Results (Scopus)	Results (Web of Science)	Content screen*
Chemicals including polymers	1,160	2,565	42
Fuels	406	860	51
Algae	771	780	32
Building materials	509	1,380	62

*individual papers containing useful information on CO_2 potential, global scale, or cost estimates

Aggregated estimates from the literature on scale and costs of the carbon utilisation pathways considered are presented in Table S3.

Table S3 | Number of papers providing estimates of scale or cost

Pathway	Scale estimates	Technical process information	Breakeven cost estimates	Total papers*
CHEMICALS	7			
Urea		4	2	4
Methanol		22	22	24
Polymers		6	1	6
FUELS	3			
Methanol		24	17	26
Methane		6	5	12
Fischer-Tropsch Fuels		6	6	8
Dimethyl Ether		3	2	3
MICROALGAE	2	20	64	30
BUILDING MATERIALS	5			
Industrial waste aggregates		40	8	41
Cement curing		7	3	16

*including papers found via snowballing. Some papers supplied multiple estimates; others supplied partial information.

Expert Opinion Survey

A structured expert opinion survey was carried out for pathways 1-5, following the 'IDEA' (Investigate; Discuss; Estimate; Aggregate) protocol[3]: using telephone or face-to-face interviews, standardised questions were investigated and clarified. Experts provided private, individual best-guess point estimates together with associated credible intervals and their confidence in their estimates, for scale and, where willing, cost. The estimates were discussed in terms of reasoning, evidence, and background variables. Respondents were asked to review and confirm their estimates on follow up. Finally the expert estimates and a co-author estimate were averaged with credible intervals interpolated to an 80% confidence level.

S1.1.2 *Non-conventional Utilisation Pathways*

For the non-conventional pathways, estimates of the potential scale and cost of CO_2 *removal* pathways provide a starting point for estimating CO_2 *utilisation*. Estimates for the global CO_2 *removal* scale and cost for pathways 6 through 10 (BECCS, enhanced weathering, forestry techniques, land management, and biochar) were derived from a previously published process of expert judgement supported by a scoping review (covering 6,326 documents from Web of Science and Scopus). See Ref [5], and the associated website www.co2removal.org for detailed methodology. Estimates for global utilisation potential and breakeven cost of utilisation for these five pathways were then made by a process of structured estimation and expert judgement for individual pathways as detailed below.

S1.2 Breakeven Cost Estimates

As described in the main paper, breakeven CO_2 costs ("β"), in units of 2015\$/t CO_2u, represent the incentive for CO_2 utilisation that would be necessary to make the pathway economic. For a process producing Q tonnes of product at price p per tonne of product, with costs (excluding any subsidies for CO_2 and any payments for the acquisition of CO_2 such as transport, capture or purchase) per unit described by c per tonne of product, and utilising υ tonnes of CO_2, breakeven costs β are defined by the requirement that revenue less costs plus the subsidy is zero:

$$pQ - cQ + \beta\upsilon = 0 \tag*{1}$$

$$\text{Hence } \beta = (c - p).(Q/\upsilon) \tag*{2}$$

The price of the product, p, is normally set by market forces. In many instances, such as methanol production, market prices are a function of supply and demand (in a given time and place) based on production processes without CO_2 utilisation, because these are currently cheaper than processes using CO_2 (i.e. $\beta > 0$; a subsidy is required for utilisation processes to breakeven). In other instances, such as urea production, market prices (in a given time and place) are already based on CO_2 utilisation, and hence a positive subsidy is not required to support production at market levels – firms can and do pay for the CO_2 input and still yield economic returns (i.e. $\beta < 0$). In the case of EOR, the "product" being produced by CO_2 utilisation is extracted oil – if CO_2 is utilised this is because it reduces the cost per unit of extracted oil and is thus profitable without subsidy (again $\beta < 0$).

Payments for the acquisition of CO_2 are netted off in order to normalise assumptions across different studies about the cost of CO_2 as a feedstock. Some studies assume free CO_2; others factor in costs of purchase, transport, or capture such that these inputs vary from 0 to \$200/t CO_2.

S1.2.1 Conventional Utilisation Pathways

Breakeven costs for conventional utilisation pathways are the total 2015 USD production costs (capital costs and operating costs) per tonne of CO_2 utilised, adjusted for any revenues, by-product revenues, and any attributed CO_2 credit or tax.

Values of c, p and Q/υ (Eq 2) were recorded or calculated (depending on the available information) from individual papers in the scoping review. c is the recorded cost (other than any CO_2 input costs) of the product made using CO_2. p is the sum of revenues, any by-product revenues, adjusted for any attributed CO_2 subsidy or credit. Q/υ is the tonnes of product produced per tonne of CO_2 utilised (the inverse of the carbon intensity of the product). If p or Q/υ were not supplied in individual papers, then, providing the products were of identical chemical structure to that made using the conventional process, we used the relevant commodity price (Table S4) and standard conversion factors (Tables S9 and S11). In the calculation of β we normalised for currency (to USD, using an exchange rate of EUR1.2, £1.5 and A\$1), inflation (using the US CPI index), and units (using standard conversions).

Box S1: Worked example of breakeven cost extraction from scoping review

Koenig et al 2015 calculate a prospective cost of production of \$12.41-21.35/GGE (gasoline-gallon-equivalent) for Fischer-Tropsch fuels. They assume a ratio of 4.19 tonnes CO_2 to 1 tonne of hydrocarbon output. The paper uses three potential product examples – crude oil, hexane and diesel. The selling price of diesel is cited in the paper at \$3.8/gallon diesel (Table 4) . Standard conversations are 1 gallon diesel = 0.88 GGE = 0.0032 tonnes diesel. CO_2 is purchased in the model at \$50/t (Table S3) and using standard conversions.

Taking the lower end of the range, the prospective cost of the carbon utilisation product is \$12.41/GGE * 0.88 = \$11.49 / gallon diesel, compared with the market price of \$3.8 / gallon diesel. At these costs, a large subsidy for CO_2 utilisation would be required to make the process breakeven. Using Equations 1 and 2, we calculate the breakeven cost is $\frac{\$(11.49 - 3.8)}{0.0032\text{ t diesel}} \cdot \frac{1\text{ t diesel}}{4.19\text{ t }CO_2} - \$50\text{ /t }CO_2 = \$524\text{/t }CO_2$.

Despite our best efforts, the netting out of some costs and the inclusions of relevant co-benefits may be incomplete or unavoidably inconsistent. We also note that some cost estimates in the literature may suffer from promoter bias[6], leading to lower cost estimates, and that cost estimates derived from laboratory results are almost always lower than those derived after the completion of pilot plant operations[7]. On the other hand, updated commercial cost estimates are frequently not in the public domain; excluding these implies higher cost estimates.

Breakeven cost ranges reported represent the 25th and 75th percentile of the sample of estimates recorded in the scoping review.

Table S4 | Product price proxies for conventional utilisation pathways

Sub-pathway	Product or basket of products	Reference
Urea	\$200/t urea	[8]
Polyol	€1.2/kg PPC	[9]
Methanol	\$400/t methanol, or as given in individual papers	[10]
Aggregates	Specific to individual case studies in scoping review	
Cement curing	Specific to individual case studies in scoping review	
Methane	\$360/t SNG	[8]
FT fuels	\$600/t aviation fuel or \$1250/t diesel or \$1670/t gasoline	[11]
DME	\$660/t DME	[12]
Microalgae	\$1000/t biodiesel, or as given in individual papers \$20/kg shellfish-derived glucosamine, or as given in individual papers	[13] [14]

S1.2.2. Non-conventional Utilisation Pathways

For non-conventional utilisation pathways, we considered the relationship between CO_2 utilised and CO_2 stored or removed in order to derive estimates of the costs per tonne of CO_2 utilised ($\kappa = c.(Q/\upsilon)$), and using Equation 1 to find breakeven costs β via:

$$\beta = \kappa - p.(Q/\upsilon) \qquad 3$$

For some non-conventional pathways (*e.g.* soil and biochar), the CO_2 utilised is additional to the CO_2 stored. In other words, CO_2 removed from atmosphere is either stored in soil or used to enhance crop growth, but not both. Previous estimates have attributed all costs of CO_2 removed to the cost of CO_2 stored, without attributing any cost to the CO_2 utilised. Here we derive κ costs per tonne of CO_2 utilised by rebasing estimates of costs per tonne of CO_2 stored, σ, from a previously published scoping review [5] using S tonnes of CO_2 stored and υ tonnes of CO_2 utilised (Table S8):

$$\kappa = \sigma.S/(S + \upsilon) \qquad 4$$

This apportions the cost of the activity (e.g. soil management techniques) between sequestration and utilisation on a per tonne basis.

For other non-conventional pathways (*e.g.* forestry and BECCS), the CO_2 removed is identical to the CO_2 utilised and the CO_2 stored, i.e. $S = \upsilon$ and $\kappa = \sigma$.

Removal cost estimates, σ, for pathways 5 through 9, are sourced from Ref [5]. For details, see www.co2removal.org. These removal cost estimates were extracted only on a global basis. Estimates of cost were identified as having different forms, i.e. 1/ deployment or capital costs (for instance, the cost of converting land use to forestry,); 2/ opportunity costs (for instance, the lost revenue from competing land uses, principally agriculture); 3/ CO_2 prices at which a pathway is deployed at a given scale; and 4/ normalised average CO_2 prices required to sequester a unit of CO_2 for a given pathway. While the aim was to identify only the last of these cost categorisations, it was rarely reported and often derived from widely varying assumptions. In order to avoid inappropriate comparisons, conversions and exclusions were carried out.

To approximate values of p and Q/υ (Eq 3), we used a representative commodity price or price basket from the FAOSTAT database or publicly available sources (Table S5) and the amount of CO_2 required to make an equivalent amount of product (where appropriate, using a conversion factor from C to CO_2 of 3.664 = 44.0095/12.0107). Where necessary values of β were normalised for currency, units and inflation as per S1.2.1.

In the calculation of p for non-conventional pathways we have made a number of very broad assumptions, which are not limited to the following:-

- BECCS services are limited to electricity generation.
- Grazing is limited to pure grassfed systems rather than mixed crop-livestock systems.
- We do not account for local pricing inequalities or market distortions.
- There are no significant additional costs of utilisation above those subsumed within the costs of removal, with the exception of forestry, where we estimate additional costs of harvest and processing, but not additional establishment costs such as road construction, which could be considerable.

Table S5 | Revenue proxies for non-conventional utilisation pathways

Sub-pathway	Product or basket of products	Reference
BECCS	Wholesale electricity price/kWh in top candidate BECCS countries (China, US, India, Germany, Brazil, France, UK, Mexico, Italy & Spain)	[15]
Forestry techniques	Industrial roundwood in the rough (Latin America, Asia and Africa)	[16]
Cropland SCS	Top 10 global crops by volume (cassava, maize, oil palm fruit, potatoes, rice, soybeans, sugar beet, sugar cane, vegetables and wheat)	[16]
Grazing SCS	Grassfed cattle, goat and sheep meat & milk	[16]
Biochar	Top 10 tropical crops by volume	[16]

S1.3 Scale Estimates

S1.3.1. *Conventional Utilisation Pathways*

Estimates of potential scale presented in the paper for conventional utilisation pathways are derived from the aggregated expert opinion survey, estimates from the literature review, and structured estimates made by the author group, by an unweighted mean.

For the structured estimation process within the author group, for chemicals, fuels, and building materials, we calculated a 2050 global CO_2 utilisation potential υ (in tonnes CO_2 yr^{-1}) as the volume of product Q, multiplied by the amount of CO_2 required to make an equivalent amount of the product (υ/Q). To estimate Q we constructed simple scenarios based on market sizes, market growth rates, and the potential for CO_2 utilisation products to penetrate the market. υ/Q was either calculated according to conversion factors $(v_{CO2}\, M_{CO2}) / (v_p\, M_p)$ where v is the stoichiometric coefficient and M is the molar mass[17] with adjustments made to account for conversion inefficiencies, or drawn from experimental achievements (for instance, see Tables S9, S11 and S13). Structured estimates for CO_2-EOR and microalgae are described in S.2.3 and S.2.5.

S.1.3.2 Non-conventional Utilisation Pathways

Global utilisation potentials for pathways 6-10 were calculated as the tonnes CO_2 yr^{-1} utilised in 2050 according to our broad definition of utilisation under each pathway, described in detail under Section S2. For each pathway these were cross checked to be congruent with removal potential estimates.

S2 Results

Table S6 | Summary calculation of conventional utilisation pathway breakeven costs, scoping review

Pathway	*c* **Cost of new product** **\$/t product** **25th-75th percentile**	*Q*/*v* **Ratio of CO_2 to product** **median**	*p* **Revenue** **\$/t product** **median**	*β* **Breakeven costs** **\$/t CO_2** **median, 25th-75th percentile**
Chemicals	280 to 870	1.4	400	0, -80 to 320
Fuels	440 to 2800	1.7	400	390, 0 to 670
Microalgae	2000 to 5240	1.9*	1000	380, 230 to 920
Building Materials	8 to 80	17%	25	17, -30 to 70

*tonne CO_2 per tonne biomass

Table S7 | Summary of conventional utilisation sub-pathway breakeven costs, 25th and 75th percentiles of scoping review

	Sub-pathway **25th to 75th percentile** **\$/t CO_2**		**Pathway** **25th to 75% percentile** **\$/t CO_2**
Urea	-190 to -15	Chemicals	-80 to 320
Polyol	-2590		
Methanol (chemical)	-50 to 370		
Methane	480 to 720	Fuels	0 to 670
FT fuels	520 to 1500		
DME	580 to 1800		
Methanol (fuel)	-50 to 370		
Microalgae	230 to 920	Microalgae	230 to 920
Aggregates	-30 to 70	Building materials	-30 to 70
Cement curing	20 to 1000		

Table S8 | Summary calculation of non-conventional utilisation pathway breakeven costs

Pathway	**Tonnes sequestered** **Mt CO_2 yr^{-1}**	**Tonnes utilized** **Mt CO_2 yr^{-1}**	**Cost of tonnes sequestered** **\$/t CO_2**	**Cost of tonnes utilised** **\$/tCO_2**	**Revenue approximation** **\$/t CO_2u**	**Breakeven costs** **\$/t CO_2**
BECCS	500 to 5000	500 to 5000	100 to 200	100 to 200	40	60 to 160
Forestry techniques	500 to 3600	70 to 1100	5 to 50	4 to 44	41	-40 to 10
Land management	2300 to 5300	900 to 1900	0 to 100	0 to 72	94 to 100	-90 to -20
Biochar	300 to 2000	170 to 1000	90 to 120	60 to 77	134	-70 to -60

S2.1 Chemicals

Chemical transformation of CO_2 can be direct, via electrochemical, photoelectrochemical, biochemical or thermochemical pathways), or indirectly through the use of hydrogen (which might be generated by water electrolysis or potentially novel hydrocarbon decomposition over metal salts [18]) to convert CO_2 to CO by the reverse-water-gas-shift reaction, followed by Fischer-Tropsch (FT) synthesis of fuel from hydrogen and CO. CO itself is an important platform chemical. Here, we assess the potential for CO_2 utilisation in methanol, urea and polymers.

Methanol can be used either directly as a fuel (see below) or as an intermediary in the synthesis of gasoline via the Mobil zeolite catalysis process and chemicals such as dimethyl ether (DME), methyl-butyl ether, and acetic acid [19].

The majority of global urea production is produced using ammonia and CO_2 via the Basaroff reactions $2NH_3 + CO_2 \rightleftharpoons NH_2COO^-NH_4^+$ and $NH_2COO^-NH_4^+ \rightleftharpoons NH_2CONH_2 + H_2O$ [20]. Most of the CO_2 used to produce urea comes from CO_2 generated from the steam reforming of natural gas feedstock to create hydrogen during the production of ammonia via the Haber-Bosch process $N_2 + 3H_2 \rightarrow 2NH_3$, at high temperature and pressure. Ammonia and urea plants are usually co-located, and ammonia plants often vent CO_2 to the atmosphere (at 0.5 kg CO_2 / kg ammonia). CO_2 capture and utilisation to boost urea yields reduces such CO_2 venting. While this does reduce CO_2 emissions, it does not constitute CO_2 removal (because the CO_2 originates from natural gas) or storage: once applied to agricultural land, urea reacts with water to release CO_2 back to the atmosphere (within a week of application) and ammonia, which decomposes slowly to supply nitrogen to crops.

CO_2 can be used directly in polymers by carboxylation reactions[21], substituting for other expensive feedstocks such as epoxides (for polycarbonate polyols) or highly toxic phosgene (for carbonates). The ring-opening copolymerization reaction facilitates the process of reacting CO_2 with epoxides to make polymers such as aliphatic polycarbonates[22]. Fossil fuels are substituted via the replacement of polyether polyols with CO_2-derived polyols and via the partial substitution of petrochemically-derived epoxide by CO_2 molecules. The majority of CO_2 polymers are currently deployed in the construction or automotive sectors. CO_2 derived polyols show better UV resistance and resistance to chemicals compared to polyether/ester polyols.

Theoretically, CO_2 could also be used directly or indirectly in the production of olefins such as ethene and propylene that are mostly used as a feedstock for subsequent synthesis of polymers[23,24]. Catalysts for CO_2-to-olefins reactions currently have low specificity and electrocatalysts for this purpose have very low yields and low selectivity.

Scale Estimates

The mean values of plausible low and high scenarios for methanol as a chemical feedstock were 20 to 250 mt CO_2 yr^{-1}. In relation to our future market scenarios, these ranges might reflect (amongst many other possibilities) growth of methanol chemical markets of 6% yr^{-1} to 2030 and 2.5% to 2050, with the low scenario reflecting a ~10% market share and the high scenario reflecting a ~85% market share.

The mean values of plausible low and high scenarios for urea were 260 to 350 mt CO_2 yr^{-1}. The low scenario might reflect a saturated market continuing from present (i.e. 100% market share) with annual growth rates of around 2.1% to 2050. The upper end reflects a higher growth rate scenario (around 3.5%) reflecting greater demand for fertiliser products.

The mean values of plausible low and high scenarios for polymers were 10 to 50 mt CO_2 yr^{-1}. Cross-referencing these values with our future market scenarios, the low scenario might reflect (amongst many possibilities) a 50% market share of the existing polyurethane polyol (18 Mt yr^{-1}) and BPA polycarbonate markets (3.6 Mt yr^{-1}), with market growth rates of 6% to 2030 and 2.5% to 2050. These growth rates are unlikely to be heavily impacted by a consumer backlash against single-use packaging products which are a different part of the market to CO_2-polymers. Our market growth rates result in a 1 bn Mt yr^{-1} 2050 polymer market, consistent with a global use at the European per capita level of 100 kg, and a global population of 8-9bn. The high scenario would assume significant penetration of other markets beyond PU polyol and BPA polycarbonates, or a very minor (~2.5%) penetration of a CO_2-olefin product into (for instance) ethene and propylene feedstock usages.

Table S9 | Estimates of scale, chemicals

Substrate	t CO_2 / t substrate (experimental) based on 5% conversion loss [35] and stoichiometric equations [17]	Current global production (mt yr^{-1})	Implied annual market growth rates to 2050 (low-high)	Implied market share for the CO_2 product in 2050 (low-high)
Urea	0.70	200[8]	2.1% to 3.5%	100%
Methanol (non-fuel)	1.30	40[8]	3.8%	10% to 85%
Low MW polymers, e.g. polyurethane polyols	0.22[25]	20	3.8%	50% to 100%
High MW polymers, e.g. BPA polycarbonate	0.40**	4[26]	3.8%	50% to 100%
Olefins (ethene / propylene)	3**	200[24]	2.3%	0% to 2.5%

**based on commercial or pre-commercial achievements for CO_2 by mass, e.g. Econic aliphatic polycarbonates 50%, Novomer carbonates for mattresses, car seats etc. 40-50%, Opus 12 3.13 t CO_2 per t ethene.

Our estimates of potential utilisation in chemicals are between 0.3 and 0.6 Gt CO_2 yr^{-1}, with the majority of that potential derived from urea, compared to previous estimates of the global potential for CO_2-based chemicals of 0.2 to 0.7 Gt CO_2 yr^{-1} [26-28].

Cost Estimates

The scoping review for the chemicals pathway identified a number of techno-economic analyses (n=26), but few (polymers n=1, urea n=2, methanol n=17) rigorous cost estimates of CO_2-based chemicals other than methanol, which is discussed under Section S2.9.

The single polyol estimate is based on a techno-economic and LCA study using polyether carbonate polyols, derived from CO_2, for flexible foam polyurethane applications[9]. For the overall polyol market, both CO_2 uptake and polyol application values are variable.

Costs are sensitive to assumptions of CO_2 per mass of product, given conversion efficiencies, process design and the particular technological pathway (for instance, electrolysis-based sub-pathways can be highly sensitive to the cost of H_2, and thereby electricity costs, e.g.[29]). Urea production is sensitive to raw material and utility costs[8]. Polymer breakeven costs (for the case study of polyols) were in the region of -\$2590/t CO_2[30] and urea from -\$185/t CO_2[31] to -\$13/t CO_2[32].

Table S10 – Summary of costs, chemicals - median values from scoping review

Pathway	c Cost of new product made with CO_2 utilisation (\$/t product)	p Selling price of product (\$/t product)	% differential for costs of product	Q/υ Ratio of CO_2 to product	β Breakeven costs (\$/t CO_2)
Urea	110	200	-45%	1.4	-99
Polymers	1440	2040	-30%	0.2	-2590
Methanol	510	400	28%	1.5	59

S2.2 Fuels

CO_2 to fuel pathways involve products such as methanol, methane (synthetic natural gas, SNG), dimethyl ether (DME) and various synthetic hydrocarbons, all candidates for the storage of renewable energy as well as standalone, or blendable fuels, or fuel precursors. They are formed via inherently energy-intensive

CO_2 reduction reactions which break one or both C=O bonds. Both CO_2 to methanol and its derivatives ('Power-to-Fuel') [19,33] and CO_2 to methane via the Sabatier process, $CO_2 + 4H_2 \rightleftharpoons CH_4 + 2H_2O$ ('Power-to-Methane') [34,35] are long-discussed concepts. Synthetic fuels formed via, *e.g.*, the reverse-water-gas-shift reaction using H_2 followed by Fischer-Tropsch, are candidates for the displacement of fossil-based fuels, particularly for sectors such as aviation and shipping [36,37].

To produce carbon-neutral fuel, a combination of atmospheric CO_2 and carbon-free energy is required[38,39]. Low-cost and reliable renewable energy appears to be a necessary condition for economic viability of such fuels. This might be provided by continuing falls in the cost of solar PV and electrolysis, or by the growth of semi-arid adapted plants in regions with plentiful reliable sunshine[40]. It is also sometimes proposed that otherwise-curtailed renewable energy on conventional grids be used to power CO_2-based fuel production; this is a long-term proposition if abundant surplus renewable energy is available, which may be unlikely at less than 50% renewable energy penetration[41]. In such instances, CO_2 conversion plants capable of reacting to periods of curtailment would be beneficial, but lower capacity factors imply lower returns on capital for such plants[41,42]. Alternative uses for otherwise-curtailed renewable energy may be more economically valuable. Provision of improved grid management and system flexibility might avoid curtailment such that renewable energy instead displaces high-carbon generation[43].

Scale Estimates

The mean values of plausible low and high scenarios for methanol as a fuel were 10 to 165 mt CO_2 yr^{-1}. Cross-referencing these values with our future market scenarios, these ranges might reflect (amongst other possibilities) growth of methanol fuel markets of 15% yr^{-1} to 2030 (reflecting Chinese production capacity targets[44]) and 2.5% to 2050, with the low scenario reflecting a ~3% market share and the high scenario reflecting a ~50% market share.

The mean values of plausible low and high scenarios for CO_2 methane products were 700 to 2770 mt CO_2 yr^{-1}. Cross-referencing these values with our future market scenarios, these ranges might reflect growth of natural gas markets of 2.5% yr^{-1} to 2030 and 1% to 2050 (roughly congruent with the IEA New Policies scenario [45]), with the low scenario reflecting a 3% market share and the high scenario reflecting a ~10% market share.

The mean values of plausible low and high scenarios for DME were 10 to 20 mt CO_2 yr^{-1}. Cross-referencing these values with our future market scenarios, these ranges might reflect (amongst many other such scenarios) growth of DME markets of 15% yr^{-1} to 2030 (reflecting Chinese production capacity targets[44]) and 2.5% to 2050, with the low scenario reflecting a ~3% market share and the high scenario reflecting a ~10% market share.

The mean values of plausible low and high scenarios for Fischer-Tropsch fuels were 220 to 1770 mt CO_2 yr^{-1}. Cross-referencing these values with our future market scenarios, these ranges might reflect growth of aviation fuel markets of 15% yr^{-1} to 2030 and 2.5% to 2050, reflecting IATA projections, with the low scenario reflecting a ~40% market share and the high scenario reflecting a 100% market share and further penetration into the global diesel market.

In these scale scenario explorations, we assume 5% conversion losses and the molar mass of octane is taken to be representative for FT fuels.

Table S11 | Estimates of scale, fuels

Substrate	t CO_2 / t substrate (experimental) based on 5% conversion loss [35] and stoichiometric equations in Refs [17,27]	Current global production (mt yr^{-1})	Implied annual market growth rates to 2050 (low-high	Implied market share for the CO_2 product in 2050 (low-high)
Methanol	1.3	30	7%	3% to 50%
Methane	2.6	5000[46]	1.6%	3% to 10%
DME	1.67	5	7%	3% to 10%
Fischer-Tropsch Fuels	2.93	20	7%	40% to 100%

Cost Estimates

The scoping review for fuels identified a literature characterised by theoretical models and techno-economic analyses of systems models (n=49), covering a broad range of prospective technological pathways and integrated systems analyses. There was little direct consideration of the impact on CO_2 utilisation *per se*, but as with the chemicals pathways these varied according to choice of technologies and assumptions on conversion efficiencies.

Cost estimates (n=30) are highly dependent on plant scale and capacity utilisation[47]. Particularly for electro-catalytic processes, the cost of electricity is also highly significant. All these factors relate to the integration and use of renewable energy. The bulk (>85%) of the cost of CO_2-to-methanol production via hydrogenation is the capital and operating cost of H_2 production [48]. Methanol breakeven costs (n=17) ranged from -\$110/t CO_2 [49] to \$640/t CO_2[50]. DME breakeven CO_2 costs (n=2) ranged from \$580/t CO_2[12] to \$1,800/t CO_2[30], FT fuel breakeven costs (n=6) ranged from \$43/t CO_2 [42] to \$1,560/t CO_2 [51] and methane breakeven costs (n=5) ranged from \$360/t CO_2 [52] to \$4,510/t CO_2 [53].

Table S12 – Summary of costs, fuels – median values from scoping review

Pathway	*c* Cost of new product \$/t product	*p* Revenue \$/t product	% differential for costs of product	*Q*/*υ* Ratio of CO_2 to product	*β* Breakeven costs \$/t CO_2
Methanol	510	400	28%	1.5	59
Methane	1739	360	383%	2.8	496
FT fuels	4159	1200	593%	4.2	767
DME	2741	660	315%	1.7	1184

The few global estimates available for utilisation of CO_2 in fuels range from 15 Mt yr^{-1} for DME and methanol [26] to 2 Gt yr^{-1} by 2050 [46,54]. Our 2050 estimates based on the assumptions above range from 0.9-4.2 CO_2 Gt yr^{-1}.

S2.3 Microalgae

The use of microalgae to make fuels (via reacting the microalgae-derived fatty acids with methanol to make fatty acid methyl esters that comprise biodiesel) or high-value carbohydrates and proteins, employs highly-adapted photosynthetic carbon conversion processes to displace fossil-based fuel carbon.

Carbon content in microalgal biomass ranges between 45-50%. CO_2 uptake efficiencies across the process can be highly variable [56] and outgassing to the atmosphere particularly from open water ponds has to be managed. Productivity yields are also highly variable according to geographical location and cultivation techniques [57].

Technological research has evolved from focusing on open raceway ponds to photobioreactors to hybrid systems or vertical flat panel reactors. Processing steps under consideration have also evolved away from solvent extraction techniques (algal lipid extraction and upgrading) towards hydrothermal liquefaction or whole biomass fractionation. These reflect attempts to capitalise on synergies such as high-value carbohydrate or protein co-products or non-fuel uses of whole algae (including bio-based plastics) , and the use of integrated biorefinery concepts [58]. Potential use of microalgae in wastewater treatment or biogas clean-up provides further routes to optimise economics and reduce resource use [56].

Technology Readiness Levels (TRLs) range from commercially viable (though not for bioenergy products) open-pond facilities to much less developed newer technologies as noted above.

Constraints to scale for microalgae include high resource requirements, particularly water and nutrients. Water use requirements will vary according to growth conditions. Assuming a 20% oil content, 3000-5000 litres of water, ~0.38 kg N and ~0.07 kg P would be required per 1 kg biofuel produced[59]. Typically, point sources of CO_2 are required, with research ongoing into new technologies to use atmospheric carbon capture to support higher microalgae growth.

Scale Estimates

For our structured estimate, assuming CO_2 removal efficiencies of 75%[56], extraction efficiencies of 90%, conversion efficiencies of 90% and a land packing factor for associated required infrastructure of 0.8[57], and assuming that 2% of non-arable global land (~46m ha) is available, the potential CO_2 utilisation capacity would be ~1.8 Gt CO_2 yr^{-1}. This would imply a 2050 global production of 500 bn litres of algal biofuel. By comparison, the IEA forecasts that current global biofuel production of 100 bn litres will need to quadruple by 2060 in a 2°C warming scenario. In other words, if all calculated microalgal potential were exploited, it would comprise over 100% of the expected market size of biofuels in 2050. Unsurprisingly, there are significant restrictions to achieving this level of implementation, particularly with regards to cost optimisation, the potential continued requirement for concentrated CO_2 to achieve required yields, and the complex economics of microalgae production. The US Department of Energy has a 5 bn gallon/yr production target of algal biofuels by 2030 (~20 bn litres); if the US alone pursued this pathway the scale of CO_2 utilised would equate, approximately, to around 100 Mt CO_2 yr^{-1}.

Our mean scale estimate as presented in the paper is a range of 160 to 870 Mt CO_2 yr^{-1}. The low end reflects a scenario whereby the US and other countries were supportive of the deployment of algal-based biofuels. The high end reflects a global scenario as above whereby microalgae fulfils around 80% of the 2050 global demand for biofuels.

Cost Estimates

A relatively large number (n=30) of technical economic analyses were identified in the scoping review for microalgae, from which 46 estimates of cost were extracted. These showed a wide variability in complexity and assumptions. The key variables are yield productivity of the algal strain, with an often-large gap between experimentally achieved yields and assumed yields in models [59].

Although algae productivity yields of 25-30 $g/m^2/day$ are routinely used in modelling assumptions, realised yields can be much lower. Average current global algal biomass productivity yields are estimated to be around 9.4 $g/m^2/day$[57] (~ 28 t/ha/year). The equivalent average lipid productivity (based on a lipid simulation model rather than on explicit oil content percentages) is 17 g $ha^{-1}yr^{-1}$.

The scoping review, which is necessarily retrospective, reflects the high costs of systems under consideration. Cost estimates range from -\$70/t CO_2[60] to \$5,300/t CO_2[61] (median: \$380/t CO_2) reflecting wide assumptions on achievable yields and capital costs, which comprise >90% of total production costs, making plant lifetime & depreciation assumptions critical. The study with negative breakeven costs[60] assumed by-product revenues from lipids, energy (both electricity and steam), credits from wastewater treatment, and from nitrogen-rich residue sales. With further technological refinements, costs are still expected to reduce considerably. The US Department of Energy's projected cost target for 2022 is \$490/t algal biomass (~\$270/t CO_2)[59].

S2.4 Building Materials

Of the 60-70 Gt yr^{-1} of materials extracted, harvested and consumed by humans, ~30 Gt are for construction[62,63]. Human use of building concrete and virgin aggregate is ~17 and ~15 Gt yr^{-1} respectively[63] and ~4 Gt yr^{-1} by mass of cement is used[63,64] in buildings and infrastructure.

Cement is produced by calcining carbonates such as limestone ($CaCO_3$), together with clays to provide silica, at high temperatures to produce four calcium silicate phases. The reverse of this process, 'carbonation', involves the reaction of metal-oxides and calcium silicates with CO_2 to form insoluble carbonates.

Building materials can utilise CO_2 in several different fashions, here disaggregated into two approaches. First, carbonation of existing materials generated from industrial waste,[a] including cement and demolition waste, can produce aggregates for building materials. Second, concentrated CO_2 can be used as an alternative for curing cement[65]. As carbonates are very stable (Figure S2), the CO_2 is potentially stored for hundreds or thousands of years, including post-demolition.

The 'process emissions' of cement production (as opposed to the emissions from the energy required for the calcining reaction) amounted to 1.5 Gt CO_2 in 2016 [66]. However, the net climate impact of CO_2 in concrete products needs to account for the fact that 11-43%[64,67] of the initial process emissions may be offset by subsequent gradual reabsorption of atmospheric CO_2 by the exothermic carbonation of hydrated cement.[166]

The key constraint to long-term scaling for aggregates may be that appropriate industrial waste feedstocks, currently 6.5 Gt yr^{-1}, are expected to decline as industry decarbonises,[68] although it is possible that fly ash, bottom ash and air pollution control residues from biomass combustion may serve as suitable substitutes. Restrictions on alkaline feedstock availability, poor cost-to-performance ratios for alternative products [69], saturated markets and the dynamic nature of CO_2 sequestration by mass are all hindrances to the potential for building material CO_2 utilisation. Furthermore, long term scaling will depend on the proportion of waste cement that has been already carbonated via natural weathering. For instance, in an experimental sample of waste cement, 11% was already carbonated[70].

Scale Estimates

Our mean values as presented in the paper for CO_2 cement curing are 100 to 1280 Mt CO_2 yr^{-1} in 2050. Cross-referencing these values with our future market scenarios, these ranges could reflect growth of concrete building products of 6% yr^{-1} to 2030 and 2.5% to 2050, with the low scenario reflecting (for instance) a ~12% market share of the precast concrete product market with a sequestration capacity of 240 kg per tonne of cement and the high scenario reflecting a 100% market share of precast concrete and a 70% share of pourable cement markets, with improved sequestration capacities of 340 kg per tonne of cement. These market numbers assume that cement is 12% of concrete; that 30% of the global concrete market is precast and another 30% is product made from pourable cement addressable for products made with CO_2-curing (at least 25% of the global market are reinforced products, which are not, at least at present, suitable for CO_2-cured cement).

Our mean values as presented in the paper for aggregates are 20 to 130 Mt CO_2 yr^{-1} in 2050. Cross-referencing these values with our future market scenarios, these ranges might reflect growth of industrial feedstocks of 2.5% yr^{-1} to 2030 and 2% to 2050, with 4% of those feedstocks being carbonated in the low scenario and 20% in the high scenario, together with 5% of global cement and demolition waste. Volumes of cement and demolition waste are derived from Ref [63], assuming 65% is concrete. Cement and demolition waste gets separated into fines (waste cement and waste concrete; we assume 17% is suitable for carbonation) and recycled concrete aggregates, which has around a 1.1% sequestration capacity[71]. Volumes of cement and demolition waste forecast for 2050 are congruent with the observation that a global convergence of per capita material consumption at industrial levels by 2050 equates to a fourfold increase in global material stocks and a 1.3x increase in annual global net additions to stocks[63]. Sequestration capacities for industrial materials are based on experimental work carried out where possible from the same laboratory (Table S13).

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[a] *In situ* carbonation provides a permanent form of CO_2 storage in silicate rocks, according to $Mg_2SiO_4 + 2CO_2 \rightarrow 2MgCO_3 + SiO_2$, but does not provide any products or revenue generation, and is therefore not included in this review. *Ex situ* carbonation can also be performed on ultrabasic rocks such as olivine, but these materials are not used as products in buildings.

Table S13 | Structured estimates of scale, building materials

Substrate	t CO_2 / t substrate (experimental)	Current global production mt yr^{-1}	Implied annual market growth rates to 2050 (low-high)	Implied market share for the CO_2 product in 2050 (low-high)
Coal pulverized fuel ash	0.097[72]	600[72]	2.1%	4% to 20%
Cement & demolition waste	0.055[71]	4200[63]	2.1%	0% to 5%
Steel slag	0.165[72]	400[73]	2.1%	4% to 20%
Cement kiln dust	0.223[72]	1000[72]	2.1%	4% to 20%
MSWI bottom ash	0.062[72]	80[72]	2.1%	4% to 20%
MSWI fly ash	0.089[72]	20[72]	2.1%	4% to 20%
Precast concrete products	0.029*	~6400[63]	3.8%	12% to 100%
Pourable cement products	0.04*	~6400	3.8%	0% to 70%

*2.9% corresponds to 240kg CO_2 per tonne of cement as achieved commercially by Solidia; 4% corresponds to the stoichiometric upper bound of 340kg CO_2 per tonne of cement.

Our mean estimate that the building materials pathway may be able to sequester 0.1 to 1.4 Gt CO_2 yr^{-1} by 2050 compares with previous approximations of sequestration capacity of 60-170 Mt for carbonated steel slag[74], 70-100 Mt for inorganic carbonates[26,75], and 1.24 Gt CO_2yr^{-1} for carbonates and silicas[76].

Cost Estimates

Much of the scoped literature for building materials (n=51) comprised of small-scale experiments or pilot studies investigating the impact of CO_2 technologies on material properties and the maximum amount of CO_2 sequestration achievable per mass. CO_2 sequestration by mass is a highly dynamic variable dictated by substrate composition and stoichiometry, depth, surface area, and curing duration, with reported results ranging from <0.01 t CO_2 per t recycled concrete aggregate [77] to 0.25-0.30 t CO_2 per t steel slag [78,79].

Few rigorous analyses of costs were identified (n=9). Breakeven costs of these pathways (using product prices as reported in individual studies, to reflect the wide geographical variation) ranged between -\$8/t CO_2[79] to \$1050/t CO_2[80]. Costs are highly product- and location-specific, and sensitive to transportation requirements as well as pre-treatments such as curing or additional grinding[81,82]. Revenues in these papers were derived from various sources including concrete blocks as well as co-products or other revenue streams such as high-value precipitated calcium carbonate (PCC) co-products or avoided gate fees for the treatment of hazardous waste material.

The costs per tonne of CO_2 of carbon-curing of cement are high when there are low levels of achieved sequestration capacity in the concrete products. Experimental results report a wide range of sequestration capacity[83].

Table S14 – Summary of costs, building materials - median values from scoping review

Pathway	c Cost of new product \$/t product	p Revenue \$/t product	% differential for costs of product	Q/υ Ratio of CO_2 to product	β Breakeven costs \$/t CO_2
Aggregates	21	18	20%	24%	-1
Cement curing	56	71	-21%	7%	48

Table S15 | Variations in CO_2 sequestration capacity, building materials

Substrate	t CO_2 / t substrate (Steinour equation*)[68,72]	t CO_2 / t substrate (median, scoping review)
Coal pulverized fuel ash	0.264	N/C
Cement & demolition waste	N/C	0.123**
Steel slag	0.3	0.162
Cement kiln dust	0.115	0.115
MSWI bottom ash	0.475	0.070
MSWI fly ash	0.120	0.026
Precast concrete products	N/C	0.056

*the theoretical formula typically used to estimate maximum CO_2 uptake capacity[84]

**reported levels likely reflect pre-sorted material rather than raw demolition waste

S2.5 CO_2-Enhanced Oil Recovery

Oil fields are typically developed in up to three distinct phases. Primary recovery typically uses the reservoir pressure to facilitate production – normally only 20-40% of the oil is recovered in this phase. Secondary recovery involves the injection of water to maintain the reservoir pressure and to displace (or 'sweep') oil from the reservoir. Tertiary recovery or Enhanced Oil Recovery (EOR) is a broad term for a variety of techniques which can be used to increase the amount of crude oil that can be extracted from an oil field, and CO_2-EOR uses CO_2, which dissolves in oil reducing its viscosity, to sweep additional oil, to a production site[85]. Naturally occurring CO_2 provides approximately 80% of the required CO_2 with anthropogenic sources providing the remaining 20%.

Reservoir management, understandably, is typically optimised to maximise profit, rather than CO_2 sequestration. Given sufficient economic and policy stimulus, the next generation of EOR projects may place higher value on CO_2 sequestration [86].

Based on a survey of 54 of the world's largest oil basins (representing 95% of global estimated ultimately recoverable oil), more than 90% of the world's oil reservoirs are amenable to the application of miscible CO_2-EOR. This corresponds to a theoretical potential of 470 billion bbl of additional oil, implying a theoretical *cumulative* CO_2 injection capacity in the range of 70-140 Gt CO_2[87]. Developing this potential would depend on the availability of commercial CO_2 in large volumes, which could be made possible by widespread use of carbon capture and storage.

Our expert judgement is that the lowest plausible scale of CO_2 EOR for 2050 is around 100 Mt CO_2, in a business-as-usual scenario and compared to present-day utilisation volumes of 60-80 Mt CO_2. Our highest plausible scale estimate for 2050 is around 1.8 Gt CO_2, representing a 12.5% rate of growth compared to the rate of growth of CO_2 EOR in Texas over the period 2007-2010 of 10-11% yr^{-1} [4].

It has been estimated that, CO_2 capture accounts for 47% of the costs associated with a CO_2-EOR project, while CO_2 transport accounts for 29% and the operation of the EOR project accounts for the remaining 24% [88]. CO_2 supply pricing contracts are commercially sensitive but at oil prices of \$100/bbl, and under the assumption of no other revenue stream or incentive such as the 45Q tax credit, CO_2 needs to be available at <US\$45/t for the project to breakeven [85]. Other studies have shown that increasing the utilisation rate of CO_2 above that required to increase oil recovery requires supply prices of around US\$60/t [86].

Oil prices are currently \$60/bbl and would decline if demand for oil falls as non-fossil substitute technologies are adopted by consumers. EOR projects sit towards the higher end of the oil supply cost curve.

S2.6 BECCS

Bioenergy with carbon capture and storage (BECCS) sequesters CO_2 via the growth of biomass, which is then transformed into an energy product, with the by-product CO_2 then captured, concentrated and stored. Combining low-carbon bioenergy production with carbon capture and storage (CCS) results in a net removal of CO_2 from the atmosphere[89] and at the same time energy is produced (energy production ranges of 3–40 GJ t^{-1} C_{eq} for energy crops), providing economic value added[90].

The carbon storage component depends on the type of product. Electricity production with CCS would normally result in negative emissions. In contrast, the production of biofuels for mobile or diffuse uses implies that almost all the CO_2 is released again upon combustion[91].

To approximate the revenue impact of BECCS, we took the average weighted wholesale electricity price /kWh of the top ten candidate nations for BECCS systems (countries with large and well-connected electricity markets together with suitable geological capacities) – China, US, India, Germany, Brazil, France, UK, Mexico, Italy and Spain [92]. A carbon intensity of an average coal-biomass facility of ~740g/kWh [93] was used to derive a $/t revenue figure of $42/t CO_2.

S2.7 Enhanced Weathering

In the presence of alkalinity and divalent cations of calcium (Ca^{2+}) or magnesium (Mg^{2+}) from the dissolution of silicate minerals, atmospheric carbon dioxide forms stable carbonate via $CO_2 + MO \leftrightarrow MCO_3 + \text{heat}$. This process occurs thermodynamically spontaneously over geologic timescales. The acceleration of the process by increasing surface area by crushing and spreading silicate material over land or sea, is known as enhanced weathering (EW). Some products of dissolution precipitate in soils but most will eventually be transported to coastal zones and therein to the open ocean, where bicarbonate ions dissolved in the ocean eventually become sea floor carbonate sediments. CO_2 outgassing by oceans will offset a proportion of the removal of atmospheric CO_2 as the sinks rebalance, on a very long timescale [94].

EW in agriculture theoretically not only creates net carbon sinks, but may also (through by-products of weathering) be able to reduce nitrogen loss, counteract soil acidification and supply nutrients such as silicic acid to the soil[95-97]. These effects, while not well established, may be particularly important on highly weathered soils such as in the tropics.

The most Ca and Mg rich silicate rocks have the capacity to sequester >1 tonne of CO_2 per tonne of rock. These ultrabasic rocks have the lowest energy requirement (for grinding and transportation) per unit of CO_2 sequestered, but generally exist in environmentally sensitive locations and often have additional significant health risks from potential inhalation of dust or application to agricultural soil. For instance, olivine is nickel-enriched and therefore hazardous, whilst harzburgite (also olivine-rich) contains asbestos-related materials.

Basalts (a basic rock which can sequester 200-800kg CO_2 per tonne of rock[98]) are more abundant and accessible. Basalt minerals account for $6.8\text{x}10^6$ km^2 of the Earth's surface (with significantly more beneath the surface and under the oceans) [98]. The use of rocks as fertilizers has precedent (for instance in the commercial use of dolomite and limestone), but is generally precluded by the costs of crushing and transporting. A series of field trials conducted in 1961 found that application of crushed basalt at rates of 20 tonnes per hectare, together with standard N, P, K fertilizer application, increased sugarcane yields by ~30%[99]. We do not, however, translate this yield increase into an estimate of CO_2 utilisation here.

S2.8 Forestry techniques

Afforestation (the restoration of forest on land that has had no previous tree cover, or that has been deforested for 50 years or more), and reforestation (the restoration of forest on recently deforested land) (AF/RF), captures and stores carbon from the atmosphere through photosynthesis and growth of woody biomass[100]. For most relevant species, the rate of net CO_2 uptake follows an S-shaped curve reaching a maximum in 30-40 years after which the net uptake rate declines to zero as the forest matures (with the gross CO_2 uptake rate balanced by respiration and the decay of dead organic matter) [101]. It is this dynamic that dictates the rate at which biomass can be sustainably harvested from a given landscape for other uses, *e.g.*, for wood products or as a feedstock for BECCS.

Wood products for CO_2 utilisation are seen here as sustainably harvested wood products for use in the construction industry. Sustainably managed forestry is described by the IPCC as the strategy that maximises climate mitigation outcomes over unmanaged forests[93]. In practice this will depend on local conditions. See the Sankey diagram in Ref [102] for further analysis of products from the forestry sector. The FAO classification of industrial roundwood in the rough does not include fuelwood. The longevity of carbon in harvested wood products (HWP) for construction will vary according to product type[103].

To calculate the utilisation impact of AF/RF, we assume that in sustainably managed forests, 50-70% of above-ground biomass is utilised for solid wood, paper and biofuel purposes (with 30-50% left to decay along with the roots)[104]. Of the global wood harvest, 32% is used for construction (with 53% used for fuelwood, and the remainder for pulp and paper[102]. Assuming 0.5 to 3.6 Gt $CO_2r\ yr^{-1}$ in 2050 from afforestation and reforestation[5], and assuming that 75% of that afforestation or reforestation is sustainably managed[102], this would equate to 70 to 520mt CO_2u in wood products from AF/RF activities, assuming that 60% of the forest biomass is above-ground[102].

The yearly gross inflow of carbon into wood products was calculated at 1.4 Gt CO_2 in 2008[105], with a per capita inflow of 201 kg CO_2 / cap / yr. Assuming a 2050 population of 9.15bn[106], and unchanged per capita carbon inflows, the gross inflow of carbon into wood products would be ~1.8 Gt CO_2 for 2050. 38% of current roundwood product volumes are certified under FSC or PEFC frameworks[107]. Assuming that proportion stays the same, and considering the 90% of industrial roundwood that flows into construction rather than paper & packaging[102], we estimate (very roughly) that an additional 0.6 Gt $CO_2\ yr^{-1}$ might arise from sustainably harvested wood products for construction, additional to the volumes from newly planted forests.

To approximate the revenue impact of AF/RF activities, we assumed an industrial roundwood in the rough product and found the average value of production in Latin America, Asia and Africa of ~\$167 per cubic metre [16]. We used a conversion metric for wood weights averaged over mixed hardwoods, mahogany, teak and pine[b] to find a gross revenue per tonne of product of \$115/t. We assumed that additional harvesting and processing costs would be incurred, and we adjusted the cost/t product by \$39/t based on average costs of sustainable forestry practices in Asia and Africa, updated for inflation[108]. Assuming a C content of 50% resulted in a \$ revenue/t CO_2 utilised of \$41.

S2.9 Land management

Carbon stored in soils is a significant carbon sink (containing around twice as much carbon in organic form as atmospheric carbon) and has been significantly depleted by human activity over the last century.

There are several practices that come under the definition of soil carbon sequestration (SCS), each of which either increase carbon inputs to, or reduce carbon losses from, soils. Methods include growing cover crops, leaving crop residues to decay *in situ*, applying manure or compost, using low- or no-till systems, and using other land management techniques optimising soil structure and organic matter inputs. A comprehensive list of studies on management practices is in Table 1 of ref [110]. Reducing tillage tends to shift carbon distribution to shallow depths but it is now agreed (by some[111] but not all [112]), that no-till strategies do not, on their own, increase total C storage and require augmenting with increased inputs to the soil and other soil management techniques. Whilst some studies exclude no-till as a sequestration technique[113], others do not[110].

Changing land management practices to enhance soil carbon can also lead to improved yields.[114] Interventions that increase agricultural output by increasing CO_2 uptake are considered CO_2 utilisation

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under the broad definition adopted in this paper. However, some SCS techniques may lead to no yield improvements or even negative impacts on yields thanks to increased weed growth[115].

Sequestration potentials arising from SCS will be subject to saturation and their permanence depends on climatic conditions as well as management[116]. For cropland management sequestration rates may peak at about 10 years and continue at lesser rates over another 40 years. Sink saturation depends on soil type and climate zone – sinks saturate more slowly in colder regions[117].

Yield increases associated with soil carbon sequestration rates per t CO_2 ha^{-1} yr^{-1} have been estimated at between 0.7% to 1.5%, depending on geographical region, with a global average of 0.9%[118]. A literature review (of 32 studies) exploring the potential effect on crop grain yields of the implementation of the 4 per mille initiative found an average 1.3% increase in yields associated with a 0.4% annual increase in soil organic carbon (SOC) stock[119]. The proportional increase in net primary productivity (NPP) is 2%, assuming a global SOC sequestration technical potential of 1.4 Gt C[120] and a global agricultural SOC stock over 0-30 cm soil depth of 233 Gt[121] (which equates to a 0.6% annual increase in SOC stock).

Applying the range of 0.9-2% to global crop production (as an approximation of NPP of 8.92 Gt C[16]) scaled to 2050 using FAO projections[106] results in a potential utilisation range of ~0.4 to 0.9 Gt CO_2 yr^{-1}. If grassland productivity increases from SCS techniques were found to be similar to those estimated for croplands, given an estimated global grazing NPP of 10.2 Gt C[122], the global SCS utilisation potential would increase to a total of ~0.9 to 1.9 Gt CO_2 yr^{-1} in 2050.

To approximate the revenue impact of land management activities on croplands, we derived a weighted basket price per tonne of the global top ten global crops (by production volume) using gross production values [16], of ~$178/t. We used a C content of 50% to find a $ revenue/t CO_2 utilised of ~$94. To approximate the revenue impact of land management activities on grazing lands, we used a weighted basket of products from grasslands, being cattle meat/milk and sheep/goat meat/milk [16], and used the gross production values for those products to derive a $ revenue per tonne of product of ~$1500/t. We used a feed conversion ratio of 8 and normalised for C content of 50% to derive a $ revenue/t CO_2 utilised of ~$100.

S2.10 Biochar

Biochar is produced when biomass undergoes pyrolysis. The thermochemical conversion results in a material that has high resistance to decomposition. This product can then be added to soil to store carbon in a relative stable form[123] and improve soil fertility[124]. The greatest and most consistent yield improvements have been found to be on highly degraded soils[125,126]. However, field studies testing the agronomic benefits of biochar are still in their infancy. Many field experiments have found that yield gains are only achieved by adding biochar in the presence of nitrogen-based fertilizers (albeit a reduced rate)[127]. The variation in those experiments ranged from no observed effect to a three-fold gain in yields. Such variation in yield effects is likely to be a hindrance in the economic decision on the part of farmers to use biochar applications[128].

The stability of char is dependent on the temperature and speed of the pyrolysis process, and *in situ* half-life is under debate[123]. Ancient soils contain chars that are thousands of years old, but some experiments have suggested that modern biochars release up to 20% of their carbon content in the first year of application, with the remaining carbon stable for around 10 years[127]. Laboratory experiments suggest a half-life to be around 60-80 years[129]. Biochar degradability will also vary according to farming practices and by crops[125].

A recent meta-analysis[124] concluded that following biochar application, crop productivity increases on average by 10% - albeit with a wide range (-28% to 39%), and limited to short term studies of 1-2 years' duration. Assuming an average 10% increase in yield applied to a land area of 40-260m Ha[117], supporting 0.3-2.1 Gt C NPP, would result in a 2050 biochar utilisation estimate of 0.17 to 1 Gt CO_2 yr^{-1}.

To approximate the revenue impact of biochar application on croplands, we presumed that this activity would only occur in the tropics. We used the top ten tropical crops (by production volume) and derived a weighted basket price per tonne of \$252 [16] and a \$ revenue per tonne CO_2 utilised of ~\$134.

S3 TRLs and further pathway information

Table S16 | Pathway breakdown, technology readiness levels (TRLs) and permanence of storage

Pathway	Detail (if relevant)	TRL	Permanence
CONVENTIONAL UTILISATION PATHWAYS			
Chemicals	Urea Methanol Polymers	9 4-7 7	Days to Weeks Months Months to Decades
Fuels	Methanol Methane Fischer-Tropsch Fuels Dimethyl Ether	4-7 5-8 5-8 5-8	Months Months Months Months
Microalgae		5-7	Months
Concrete Building Materials	Cement & demolition waste Other industrial waste aggregates Cement curing	3-5 3-9 4-8	Centuries Centuries Centuries
CO_2-EOR		7-9	Millennia
NON-CONVENTIONAL UTILISATION PATHWAYS			
BECCS		5-7	Millennia
Enhanced Weathering		2-3	Millennia
Forestry Techniques	Afforestation and Reforestation Wood products	9 8-9	Decades to Centuries
Land Management	Soil Carbon Sequestration	9	Years to Decades
Biochar		4	Years to Decades

Technology readiness levels (TRLs) delineate maturity according to whether the technology is at basic principles (TRL1), invention and research (TRL2), proof of concept (TRL3), bench-scale research (TRL4), pilot scale (TRL5), large-scale (TRL6), inactive commissioning (TRL7), active commissioning (TRL8), or operational (TRL9).

S4 Supplementary Figures

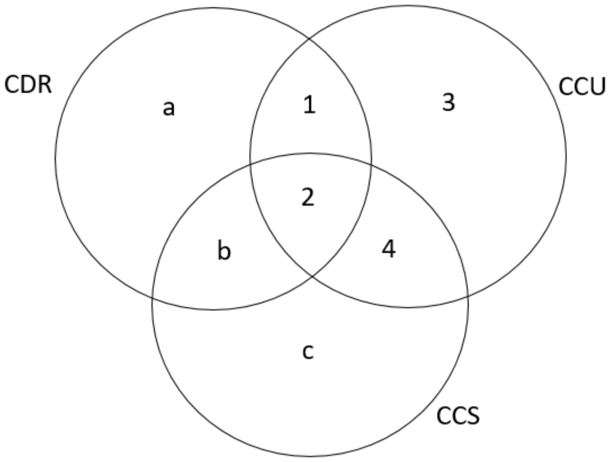

Figure S1 | CO_2 utilisation and related concepts. Some processes that utilise CO_2 (CCU) may also result in carbon dioxide removal (CDR) from the atmosphere (areas 1, 2). CO_2 utilisation with carbon capture and storage (CCUS, areas 2, 4) may lead to net CDR (area 2), but might also mitigate climate change by reducing net emissions (area 4, *e.g.* CCS on fossil exhaust gases with subsequent CO_2 use that offsets other emissions), without net removal. CCU can also function without storage or removal (area 3, such as production of short-lived polymers). All three concepts – CCU, CCS and CDR – might or might not result in a climate benefit, depending on execution and once all indirect effects are accounted for. Numbering 1-4 denotes techniques that utilise CO_2 with example techniques as follows: 1: biochar, 2: BECCS, 3: CO_2 polymers, 4: CO_2 EOR. Lettering a-c denotes techniques that do not utilise CO_2, with example techniques as follows: a: *ex situ* mineral carbonation, b: direct air capture with CCS (DACCS), c: CCS on fossil exhaust gases.

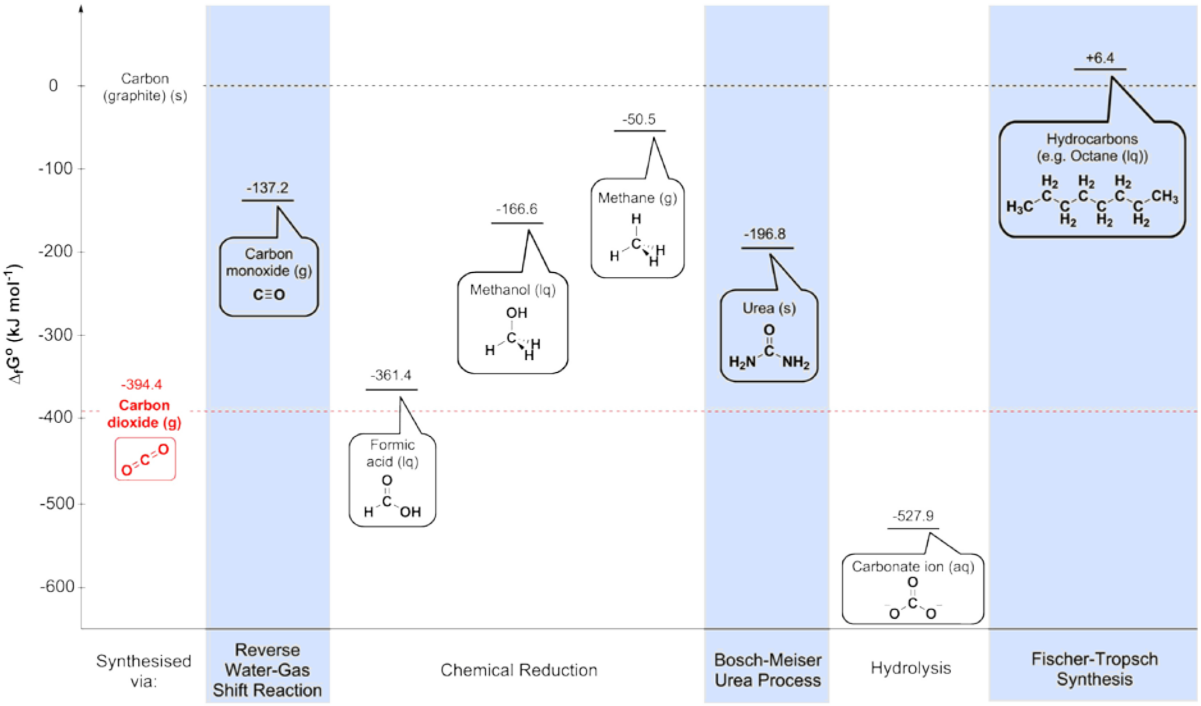

Figure S2 | The chemical energy (Gibbs free energy of formation) of different carbon-based compounds at 298 K[130]. CO_2 is a thermodynamically stable molecule and transformations to most other carbon-based molecules, including hydrocarbons or other oxidation states, involve substantial energy inputs. Hydrocarbons are useful fuels precisely because combustion to CO_2 is strongly exoergic. In contrast, mineral carbonates for building purposes (here shown in the hydrolysed aqueous form) are more thermodynamically stable than CO_2.

S5 Further barriers to scaling

Geographical considerations

The relative attractiveness of CO_2 utilisation pathways varies spatially. Industrial CO_2 utilisation may initially be constrained to locations with existing sources of high concentration flue gases[131] and cheap energy, such as “stranded” gas deposits, geothermal or hydropower or available renewables. The feasibility of enhanced weathering or concrete building materials will similarly be affected by the location of the necessary feedstocks[132,133]. Pathways such as BECCS and CO_2-EOR can only occur near appropriate sequestration sites[4,134]. For most land-use based pathways, potential scale is highly dependent on climate, land, nutrient and water availability, with the largest effects reported in warm, wet environments[57,96] or on degraded soils[135]. Rapidly industrialising and urbanising countries are a natural fit for pathways such as concrete building products or CO_2-derived chemicals or fuels[136]

Financing issues

It is highly simplistic to assume that economically viable utilisation pathways are automatically attractive to investors. Privately financing large-scale deployment is challenging where there is a long time-lag (*e.g.*, a decade) between the original investment and returns, or where the risks are substantial. Financing has already proved challenging for CCS at scale[137,138], and could also prove challenging for BECCS and utilisation pathways that require significant sunk capital investments, such as enhanced weathering. Policy can help; projects’ bankability can be improved by direct support, or more steady carbon prices that incentivize CO_2 utilisation in a way that reduces emissions or removes CO_2. Financing forestry and changes to land management may confront significant barriers such as land tenure problems, a lack of credible information, or general resistance to change.

Political and societal considerations

Socio-political factors might considerably reduce the feasible scale of deployment[139], and are poorly understood[139]. Public perception and acceptability – rather than economics or finance– can determine whether a technology succeeds or fails[140], and this in turn is affected by prevailing governance arrangements. Achieving the SDGs is growing in political salience and importance, and land-use based pathways could have particularly significant impacts — positive[141] or negative[142] on aspects such as food availability or environmental protection. Potential consequences are highly region- and technology-specific, and sometimes cross-border. Best practice analysis may take the form of collaborative and participatory governance approaches[143], with a strong focus on ethical implications[144]. Further, if new technologies threaten powerful incumbents, their political clout may manifest as a barrier. Conversely CO_2 utilisation pathways may, in some instances, help to circumvent political economic barriers by providing incumbent fossil producers with a stake in new industries.

S6 Other potential CO_2 utilisation pathways

Pathways that face considerable legal and governance hurdles, such as ocean-based utilisation and removal[145], are not considered here. There are several other potential CO_2 utilisation pathways which we did not include in our review of ten because they are understudied in the context of CO_2 utilisation, are too early stage, or are unlikely to scale. Pathways not examined here – which does not imply they are not worthy of further analysis – include plant biomass for the production of materials, chemicals, fuels and energy without CCS[146,147], [22], micro-organisms (such as cyanobacteria) for the production of fuels, chemicals and materials[148], wetland restoration[113], formic acid[149], fine chemicals[17], and the production of advanced materials such as graphene and carbon fibre[150]. Pathways not examined here – which does not imply they are not worthy of further analysis in the context of CO2 utilisation – include plant biomass for the production of materials, chemicals, fuels and energy without CCS[146,147], micro-organisms (such as cyanobacteria) for the production of fuels, chemicals and materials[148], wetland restoration[113], fine chemicals[17], and the production of advanced materials such as graphene and carbon fibre[150]. Bio-based feedstocks also exist, sometimes in commercial forms, in polymers, solvents, lubricants and fragrances[151-153]. One such synthetic plastic made from plants is polylactic acid (PLA). Biobased plastics may account for 1-3% of the total plastics market (300Mtpa). Broader materials production such as cotton, furniture and fibre for use in clothing, packaging, furniture and construction sectors are also potential biological CO2 utilisation pathways – for one example, treated biomass as cementitious building materials [154].

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