

# Curing time: a temporally explicit life cycle CO<sub>2</sub> accounting of mineralization, bioenergy, and CCS in the concrete sector†

Samantha Eleanor Tanzer, \* Kornelis Blok and Andrea Ramirez

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The decarbonization of concrete production requires a multi-pronged approach including the abatement of CO<sub>2</sub> emissions from cement production as well as storage of CO<sub>2</sub> within concrete itself. This study explores the decarbonization potential of combining bioenergy and carbon capture and storage (CCS) during cement production with the accelerated carbonation of fresh concrete and the natural carbonation of demolished concrete for the life cycle net CO<sub>2</sub> of 30 MPa ordinary Portland concrete. As both biomass and concrete reuptake CO<sub>2</sub> over time, the timing of CO<sub>2</sub> emissions and removals is explicitly accounted for. At current technology levels, the combination of bioenergy and CCS in cement production combined with the carbonation of demolished concrete was seen in our model to allow for net CO<sub>2</sub>-negative concrete. However, the concrete is CO<sub>2</sub>-positive until the CO<sub>2</sub> of production is reabsorbed by biomass regrowth and the carbonation of demolished concrete at end-of-life. In our model, accelerated carbonation was, by itself, an inefficient CO<sub>2</sub> storage mechanism, due to the penalty of energy use and injection losses. However, if it led to a gain in concrete strength, accelerated carbonation could result in lower CO<sub>2</sub> *via* reduced resource demand and cement production.

## Introduction

Concrete is the most abundant manmade material. Worldwide, 30 billion tonnes are produced annually.<sup>1</sup> The production of 4.1 billion tonnes of cement,<sup>2</sup> the binding agent in concrete, was itself responsible for 2.4 Gt of carbon dioxide emissions in 2019, representing 26% of all industrial CO<sub>2</sub> emissions.<sup>3</sup> To abate the worst impacts of the climate crisis, decarbonization of concrete is critical.

*Department of Engineering Systems and Services, Delft University of Technology, Jaffalaan 5, 2628 BX Delft, The Netherlands. E-mail: s.e.tanzer@tudelft.nl*

† Electronic supplementary information (ESI) available: The full modelling parameters and results in tabular format. Additionally, the production of concrete masonry units was also modelled, whose results follow the same trends seen for OPC. However, due to space limitations, that analysis was excluded here. Methods and results specific to CMUs are included in the ESI. See DOI: 10.1039/d0fd00139b



The cement and concrete industries anticipate that full decarbonization will require a multi-pronged approach, encompassing increases in energy efficiency, the use of wastes and biomass as fuel, and recycling; carbon capture and storage; as well as the decarbonization of transport and electricity.<sup>4,5</sup>

Cement production is the most carbon-intensive element of the concrete supply chain, requiring 3–4 GJ of thermal energy per tonne of cement, energy which today is provided mostly by fossil fuels.<sup>6</sup> Yet, about 60% of CO<sub>2</sub> emitted during cement production is from the calcination of limestone (CaCO<sub>3</sub>) into calcium oxide (CaO). Once in concrete, however, CaO reabsorbs CO<sub>2</sub> from the atmosphere, recarbonating into limestone. Estimates suggest that CO<sub>2</sub> reuptake by in-stock concrete offsets 20% of CO<sub>2</sub> emissions from current annual cement production.<sup>7</sup> Over a 50 to 100 year service life, concrete may reabsorb 10–30% of the CO<sub>2</sub> released during calcination of its constituent cement.<sup>8,9</sup> At the end of its service life, demolition greatly increases the exposed surface area of concrete, providing an opportunity for rapid recarbonization,<sup>10</sup> but less than 1% of demolished concrete is estimated to be recycled in an exposed environment.<sup>7</sup>

Another pathway to concrete carbonation is to inject CO<sub>2</sub> into fresh concrete. This “accelerated carbonation” was studied in the 1970s<sup>11,12</sup> as a method to increase the early strength and setting speed of concrete. Recently, several accelerated carbonation products have come to market claiming a reduced carbon footprint.<sup>13–15</sup> However, variation in product type and concrete recipe makes it difficult to quantify the decarbonization potential of accelerated carbonation by itself.

Furthermore, the concrete life cycle includes the sourcing of sand, aggregate, chemical additives, water and energy; demolition at end-of-life; and transport of bulk materials. All of these must be accounted for when assessing the CO<sub>2</sub> footprint of concrete.

This study explores the combination of decarbonization technologies to understand their impact on the lifecycle CO<sub>2</sub> balance of ordinary Portland concrete. We focus on accelerated carbonation, bioenergy use in cement kilns, and the capture and storage of CO<sub>2</sub> from cement kilns. As both concrete and biomass uptake CO<sub>2</sub> over time, we chart the balance of CO<sub>2</sub> emissions and removals over time. Additionally, the impact of strength gain from accelerated carbonation, sourcing of accelerated carbonation CO<sub>2</sub>, carbonation of demolished concrete, biomass rotation period, and the decarbonization of electricity and transport are considered.

## Methods

This study is a temporally explicit life cycle CO<sub>2</sub> accounting of concrete production, with and without the use of accelerated carbonation, bioenergy, and/or carbon capture and storage, based on the system in Fig. 1. The unit of analysis (functional unit) is the production of one cubic meter of 30 MPa ordinary Portland concrete, formed into an 20 cm-deep exterior wall segment with a 50 year service life. Production and use were assumed to occur in northwest Europe, which is reflected in the choices of technological efficiencies and supply chain data, but the system is otherwise geographically generic.

The cases considered in this study are summarized in Fig. 2. For all cases, the net life cycle CO<sub>2</sub> was estimated, as was the cumulative balance of CO<sub>2</sub> emissions and removals over time. CO<sub>2</sub> emissions from concrete production and upstream



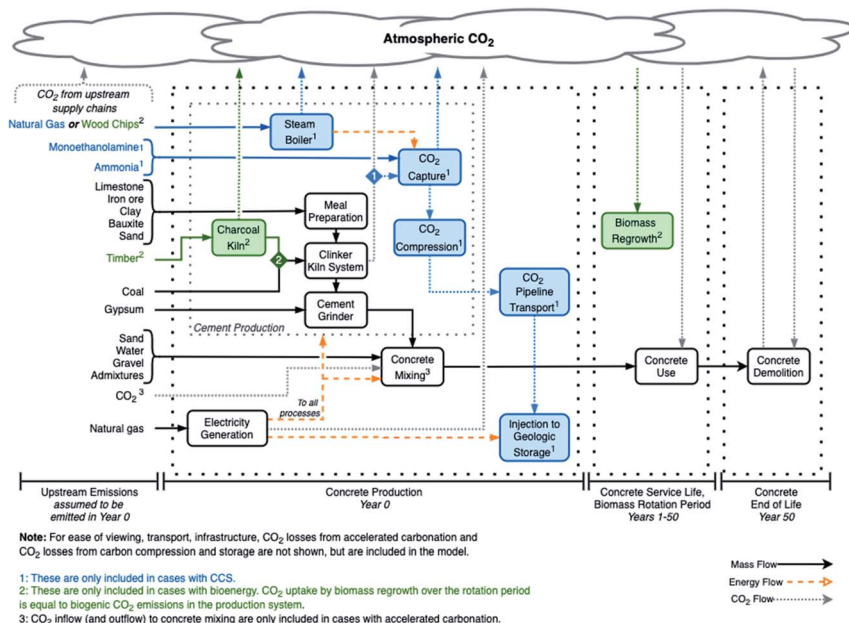


Fig. 1 System considered in this study.

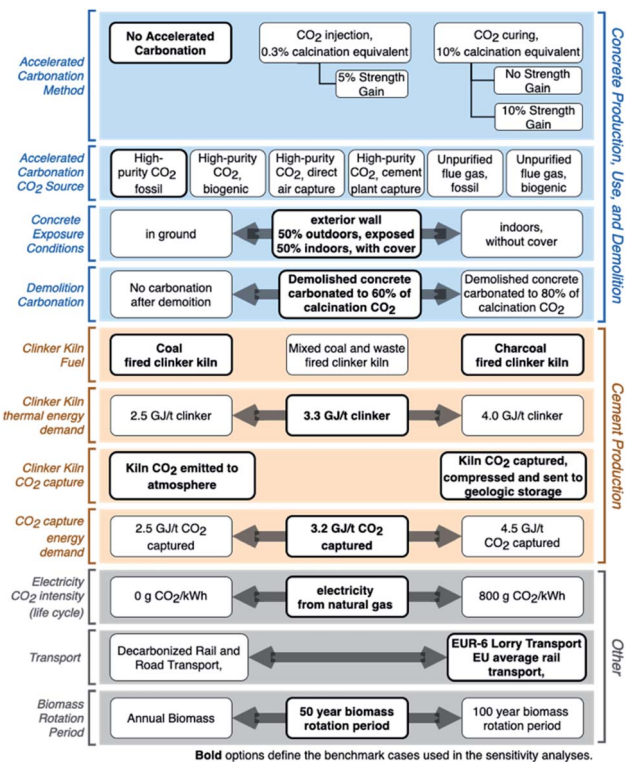


Fig. 2 Cases considered in this study.



supply chains of production inputs were assumed to be emitted in “year 0”. Afterwards, CO<sub>2</sub> is removed from the atmosphere by the natural carbonation of concrete and by replanted biomass. CO<sub>2</sub> emissions and removals associated with demolition occur in the year after the end of the concrete’s service life.

The process modelling and CO<sub>2</sub> balances were facilitated by a custom Python3 model. For CO<sub>2</sub> emissions in upstream supply chains, life cycle inventory data from ecoinvent 3.6 (ref. 16) were used. Tabular data for the model input parameters are available in the ESI.†

### Technology scenarios

Four scenarios of production technology and background systems were considered in this study, whose main parameters are summarized in Table 1:

Table 1 Main model parameters, by technology scenario

| Parameter  | Unit                             | Current, average | Current, benchmark | Future, conservative | Future, optimistic |
|--|----------------------------------|------------------|--------------------|----------------------|--------------------|
| Clinker kiln, thermal energy demand                            | MJ per kg clinker                | 3.7<br>(ref. 4)  | 3.3<br>(ref. 19)   | 3.0 <sup>a</sup>     | 2.8<br>(ref. 21)   |
| Waste fraction of clinker kiln fuel                            | % kiln fuel (LHV)                | 46%<br>(ref. 4)  | 0%                 | 60%<br>(ref. 23)     | 90%<br>(ref. 4)    |
| Biomass fraction of waste <sup>b</sup>                         | % waste                          | 16%<br>(ref. 4)  | n.a.               | 40%<br>(ref. 23)     | 50%<br>(ref. 4)    |
| Meal grinding electricity demand <sup>c</sup>                  | kW h per t meal                  | 23<br>(ref. 17)  | 12<br>(ref. 20)    | 12<br>(ref. 20)      | 12<br>(ref. 20)    |
| Clinker kiln electricity demand <sup>c</sup>                   | kW h per t clinker               | 26<br>(ref. 17)  | 23<br>(ref. 20)    | 23<br>(ref. 20)      | 23<br>(ref. 20)    |
| Cement mixing electricity demand <sup>c</sup>                  | kW h per t cement                | 40<br>(ref. 17)  | 16<br>(ref. 20)    | 16<br>(ref. 20)      | 16<br>(ref. 20)    |
| Total carbonation, after demolition                            | % of calcination CO <sub>2</sub> | n.a.             | 60% <sup>a</sup>   | 60% <sup>a</sup>     | 75% <sup>a</sup>   |
| CO <sub>2</sub> capture, thermal energy demand                 | MJ per kg CO <sub>2</sub>        | 3.2<br>(ref. 24) | 3.2<br>(ref. 24)   | 3.0 <sup>a</sup>     | 2.5 <sup>a</sup>   |
| CO <sub>2</sub> capture, electricity demand <sup>c</sup>       | MJ per kg CO <sub>2</sub>        | 38               | 15                 | 15                   | 15                 |
| Direct CO <sub>2</sub> intensity of electricity                | g CO <sub>2</sub> per kW per h   | 269<br>(ref. 18) | 360                | 54 <sup>a</sup>      | 0                  |
| Upstream CO <sub>2</sub> of electricity <sup>d</sup>           | g CO <sub>2</sub> per kW per h   | 90<br>(ref. 16)  | 48                 | 18                   | 0                  |
| CO <sub>2</sub> intensity of road transport (life cycle basis) | g CO <sub>2</sub> per t per km   | 84<br>(ref. 16)  | 84                 | 34                   | 0                  |
| CO <sub>2</sub> intensity of rail transport (life cycle basis) | g CO <sub>2</sub> per t per km   | 51<br>(ref. 16)  | 51                 | 20                   | 0                  |

<sup>a</sup> Assumption. <sup>b</sup> Assumed to be 50% short-rotation biomass (e.g. agricultural wastes), and 50% long-rotation biomass (e.g. sawdust). <sup>c</sup> Electricity efficiency beyond current state-of-the-art was neglected due to its negligible impact in scenarios with decarbonized electricity. <sup>d</sup> Upstream CO<sub>2</sub> was approximated by subtracting the direct CO<sub>2</sub> intensity of electricity generation<sup>18</sup> from the total life cycle CO<sub>2</sub> in the ecoinvent 3.6 (ref. 16) process for European average electricity generation. Decarbonization of upstream emissions was assumed to occur at the same rate as for direct CO<sub>2</sub> emissions of electricity generation.



**Current, average**, using cement production efficiencies and kiln fuel composition from the European Cement Association, CEMBUREAU<sup>4</sup> and average cement production electricity use,<sup>17</sup> and EU-average grid electricity.<sup>18</sup> At end-of-life, the concrete was assumed to be demolished and immediately re-used in a subsurface application, without further carbonation. This scenario is designed to represent current production conditions.

**Current, benchmark**, assuming “state-of-the-art” cement production.<sup>19,20</sup> To provide a clearer picture of parameter influence in the sensitivity analysis, simplified energy provision was assumed, with the clinker kiln using only one type of fuel (coal in fossil cases, charcoal in biomass cases), and with electricity provided by a natural gas combined cycle power plant with an efficiency of 56.6%. It is also assumed that at end-of-life, concrete rubble is exposed until 60% of the calcination CO<sub>2</sub> has been recarbonated, as this is an immediately implementable decarbonization option.

**Future, conservative**, with improvements in kiln and CO<sub>2</sub> capture efficiencies, increased use of waste, and partial decarbonization of transport and electricity. Additional electricity efficiency improvements, beyond current state-of-the-art were not considered, due to their negligible impact.

**Future, optimistic**, with a “practical minimum” clinker kiln efficiency,<sup>21</sup> increased use of wastes,<sup>4</sup> and fully decarbonized electricity and transport sectors, as envisioned to be available no later than 2050 in the EU.<sup>22</sup>

### Concrete production

This study considered the production of ordinary Portland concrete (OPC), with a 28 day compressive strength of 30 MPa. 25–35 MPa concrete represents 60% of the 255 million m<sup>3</sup> of ready-mixed concrete produced by members of European Ready Mixed Concrete Organization in 2018.<sup>25</sup> This study used the concrete recipe in Table 2, taken from the ecoinvent 3.6 process for 30–32 MPa ready-mix concrete.<sup>16</sup> The use of cement replacers, such as fly ash or slag, is outside the scope of this study.

In the benchmark case, inputs to concrete production were assumed to be transported 200 km by heavy lorry to the construction site, where concrete mixing occurs with water available on-site. To minimize variation between cases, accelerated carbonation was assumed to happen on the site of concrete production and use.

### Accelerated carbonation

Two cases of accelerated carbonation were considered:

Table 2 Recipe for 30 MPa concrete used in this study<sup>16</sup>

| Ingredient            | kg per m <sup>3</sup> concrete |
|-----------------------|--------------------------------|
| CEM I Portland cement | 344                            |
| Sand                  | 859                            |
| Gravel                | 960                            |
| Water                 | 207                            |
| Admixtures            | 1.2                            |



**0.3% calcination CO<sub>2</sub> injection into the concrete mixer**, based on commercially available technology.<sup>26</sup> A small quantity of CO<sub>2</sub> is injected, equalling approximately 0.3% of the calcination CO<sub>2</sub> emitted during the production of the concrete's constituent cement. This has been shown to increase the strength of the concrete, allowing for approximately a 5% reduction in cement.<sup>26</sup> Therefore, in these cases, each m<sup>3</sup> of concrete has 17 kg less cement, with an additional 14 kg of sand to maintain volume.

**CO<sub>2</sub> curing to 10% of embodied calcination CO<sub>2</sub>** where the concrete is exposed to a high-CO<sub>2</sub> atmosphere in a pressurized environment. The literature of CO<sub>2</sub> curing of cement and concrete varies widely in product recipe, curing environment, observed CO<sub>2</sub> uptake, and change in concrete properties. In particular, both strength gain and strength loss have been reported. A comparison of several CO<sub>2</sub> curing studies is included in the ESI.† In our model we assumed that two hours of CO<sub>2</sub> curing in a constant pressure environment of 150 kPa resulted in a CO<sub>2</sub> uptake of 10% of calcination CO<sub>2</sub>, with no change in strength.

In the benchmark cases, the CO<sub>2</sub> used for accelerated carbonation was assumed to have been captured from industrial flue gas, purified to 95%, and transported *via* lorry to the concrete production site. CO<sub>2</sub> uptake efficiency was initially assumed to be 60%,<sup>27</sup> with unabsorbed CO<sub>2</sub> emitted to the atmosphere. As accelerated carbonation was assumed to occur at a construction site, dedicated pipeline transport of the CO<sub>2</sub> was assumed to be unrealistic.

The study also considered cases where CO<sub>2</sub> curing led to a 10% strength gain, with a corresponding reduction of cement use. In combination, we also explored the impact of four other sources of the CO<sub>2</sub> used in accelerated carbonation:

- 95%-purity CO<sub>2</sub> from the system's own cement plant, when outfitted with CCS.
- 95%-purity biogenic CO<sub>2</sub> from the production of bioethanol, assumed to only require compression and transport.
- 95%-purity atmospheric CO<sub>2</sub> from an on-site direct air capture (DAC) unit.<sup>28</sup> This includes an electricity demand of 366 kW h per t CO<sub>2</sub> and a thermal energy demand of 5.9 GJ per t CO<sub>2</sub>, supplied by natural gas. In the DAC CO<sub>2</sub> capture process, approximately 95% of CO<sub>2</sub> from natural gas use is also captured.<sup>28</sup>
- raw flue gas (10–20% CO<sub>2</sub>), both fossil and biogenic, that has only been cleaned of SO<sub>2</sub> and NO<sub>x</sub>.

### Natural carbonation

Concrete absorbs CO<sub>2</sub> as the CaO in the cement recarbonates, first at the surface then gradually penetrating into the concrete mass with decreasing uptake over time as in Fig. 3. The carbonation rate depends on the composition of the concrete, exposed surface area, and exposure environment. It is typically modelled using Fick's diffusion law, and calculated with the equation<sup>9</sup>

$$\text{CO}_2 \text{ uptake} = \sum (k_i \times \text{DOC}_i \times A_i) \times (\sqrt{t} \div 1000) \times U_{\text{fcc}} \times C$$

where

CO<sub>2</sub> uptake, in kg, is the total CO<sub>2</sub> carbonated over period  $t$ .

$k_i$  in mm per year<sup>0.5</sup>, is the carbonation rate factor, based on the concrete strength and exposure conditions.<sup>9</sup>



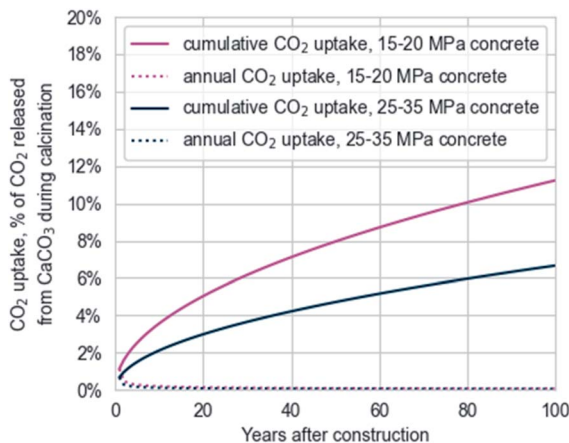


Fig. 3 Example of natural carbonation rate of concrete exposed to rain.

$DOC_i$ , in percent, is the maximum degree of carbonation specific to the exposure conditions.<sup>9</sup>

$A_i$ , in  $m^2$  is the surface area of the exposed concrete.

$t$ , in years, is the length of exposure.

$U_{tcc}$ , in  $kg\ CO_2$  per  $kg$  cement is the maximum theoretical uptake of  $CO_2$ , equal to the  $CO_2$  released during calcination. For the CEM I Portland cement, the value is 0.49.

$C$ , in  $kg$  cement per  $m^3$  concrete, is the cement content of the concrete.

Concrete use was assumed to be as an exterior wall, with a depth of 20 cm, with an external surface exposed to rain, and a painted interior surface. In cases with accelerated carbonation, natural carbonation was assumed to begin from the level of carbonation already present in the concrete. *e.g.* If 5% of calcination  $CO_2$  was carbonated by accelerated carbonation in 18 MPa concrete exposed to rain, carbonation during concrete service life was assumed to continue as if 20 years of natural carbonation had already occurred (as if starting at the year 20 point in Fig. 3).

### End of life

At its end of its 50 year service life, the concrete was assumed to be demolished with a life cycle  $CO_2$  footprint of 9  $kg$  per  $t$  demolished concrete.<sup>16</sup> As concrete rubble has a large surface area, natural carbonation up to 60–80% calcination  $CO_2$  can be achieved by leaving rubble exposed to air for several weeks.<sup>29</sup>

While the EU reports a 90% average recovery rate for construction waste,<sup>30</sup> the predominant fate of recovered concrete is road underlayers or backfilling,<sup>7</sup> where it is not exposed. Therefore, no additional carbonation of demolished concrete was assumed in the “Current, Average” scenario. Deliberate carbonation of demolished concrete was included in the benchmark scenario and both future scenarios.

### Cement production

Cement production was modelled for CEM I Portland cement.<sup>31–33</sup> Losses from kiln dust or conveyance between processes, were neglected. Production inputs



were assumed to travel 200 km by rail to the cement plant. The energy demand for cement production is provided in Table 1.

**Meal preparation.** The raw ingredients for cement production consist primarily of limestone, with smaller fractions of silicon, aluminium, and iron, typically provided by clay, sand, bauxite, and/or iron ore. This study assumed a meal composition based on a real-world mix of a cement plant in Norway<sup>32</sup> that is 77% limestone. The meal was assumed to be crushed and ground in a ball mill.

**Clinker kiln system.** The pulverized meal is fed into a kiln, which is heated in stages to 1300–1500 °C. Between 450–900 °C, CO<sub>2</sub> is released from the limestone (CaCO<sub>3</sub>) during calcination. In this model, a modern short dry kiln with a preheater and a precalciner was assumed. The non-waste fraction of kiln fuel was assumed to be coal, which is replaced with charcoal in the bioenergy cases. The benchmark scenario assumed that no waste was used as fuel.

**Cement mixing.** The cooled clinker is mixed with gypsum and/or other additives to form cement. CEM I Portland cement consists of 95% clinker and 5% gypsum and was assumed to be processed in a roller mill.

### Carbon capture and storage

In cases with carbon capture and storage (CCS), flue gas from the cement kiln was sent to a post-combustion CO<sub>2</sub> capture unit using a monoethanolamine (MEA)-based absorption process. Flue gas cleaning using ammonia and limestone to remove SO<sub>2</sub> and NO<sub>x</sub> was assumed,<sup>31</sup> as this is current practice. While there are more advanced solvent-based capture systems, and demonstrations of calcium-looping-capture and direct separation technologies for cement production are underway,<sup>34,35</sup> MEA-based capture is a mature and commercially available technology, so was chosen for the benchmark scenario, which focuses on currently available technologies.

In the benchmark scenario, the CO<sub>2</sub> capture process was assumed to require 3.2 GJ per t CO<sub>2</sub> of low-temperature steam,<sup>24</sup> provided by a dedicated boiler with a 90% efficiency. The boiler was assumed to use natural gas or, in cases with bioenergy use, wood chips. CO<sub>2</sub> from the boiler was emitted to the atmosphere. No heat integration with the cement plant was assumed in the benchmark case, as available heat can vary widely and may be in use for other purposes.

After capture, the CO<sub>2</sub> was assumed to be compressed to 110 bar, requiring 96 kW h per t CO<sub>2</sub>,<sup>32</sup> transported 200 km by pipeline, and injected into geologic storage, with an injection electricity demand of 8 kW h per t CO<sub>2</sub>.<sup>36</sup> It was assumed that 1% of CO<sub>2</sub> was lost during transport and injection.

### Bioenergy use and biomass regrowth

In cases with bioenergy use, charcoal replaces coal in the cement kiln and wood chips replace natural gas in the steam boiler for CO<sub>2</sub> capture. Charcoal was selected to ensure that the kiln would be able to reach sufficiently high temperatures, and for usability in existing kilns accustomed to coal-firing. Charcoal was assumed to be produced in industrial Missouri-style kilns, with 69% carbon recovery and ancillary CO<sub>2</sub> emissions of 543 kg per t charcoal.<sup>37</sup>

Timber for charcoal production was assumed to have a rotation period of 50 years, as was the long rotation fraction of biogenic wastes. CO<sub>2</sub> reuptake by





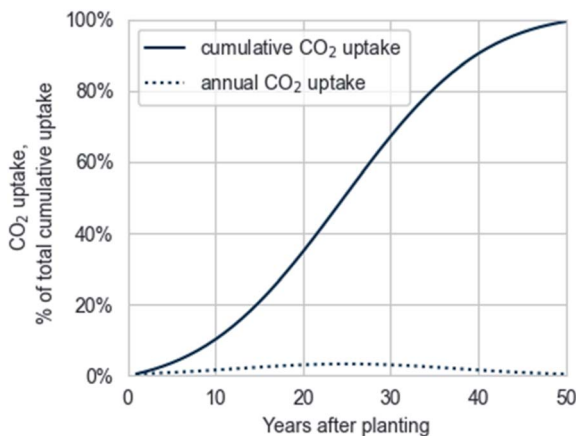


Fig. 4 Example of CO<sub>2</sub> uptake of biomass with a 50 year rotation period.

biomass over time was modelled using a Gaussian distribution, as visualized in Fig. 4, following equation<sup>38</sup>

$$\text{CO}_2 \text{ uptake in year } \tau = (2\pi\sigma^2)^{0.5} e^{-(\tau-\mu)^2/2\sigma^2}$$

where  $\mu$  is the rotation period halved, and  $\sigma$  is  $\mu/2$ . Biomass replanting was assumed to occur in year 0, and at the end of the biomass rotation period, 100% of biogenic CO<sub>2</sub> emitted during cement production is reabsorbed.

### Upstream CO<sub>2</sub>

Upstream CO<sub>2</sub> emissions were included for the inflows in Fig. 1, as well as for transport of materials to the cement plant and the concrete production site, transport of CO<sub>2</sub> to geologic storage, and infrastructure use for all processes. For this background system, life cycle inventory CO<sub>2</sub> data from ecoinvent 3.6 (ref. 16) was used, including the emissions of biogenic, fossil, and direct land use change CO<sub>2</sub>. Indirect land use change was not considered. The specific ecoinvent processes and CO<sub>2</sub> factors are provided in the ESI,<sup>†</sup> as are energy content and emissions factors of fuels used in this model.<sup>39,40</sup>

## Results and discussion

Full tabular results are available in the ESI.<sup>†</sup> Net CO<sub>2</sub> has been rounded to the nearest 10 kg CO<sub>2</sub> per t per m<sup>3</sup> concrete.

### Technology scenarios overview

Fig. 5 summarizes the net life cycle CO<sub>2</sub> for each concrete type and production scenario, before considering accelerated carbonation. The net CO<sub>2</sub> of the “current, average” scenario is 380 kg CO<sub>2</sub> per m<sup>3</sup> concrete. Excluding end-of-life carbonation, the benchmark scenario, without bioenergy or CCS, has a 15 kg m<sup>-3</sup> lower net CO<sub>2</sub> due to increased production efficiencies. Carbonation of demolished concrete accounts for a further reduction of 85 kg CO<sub>2</sub> per t per m<sup>3</sup>.



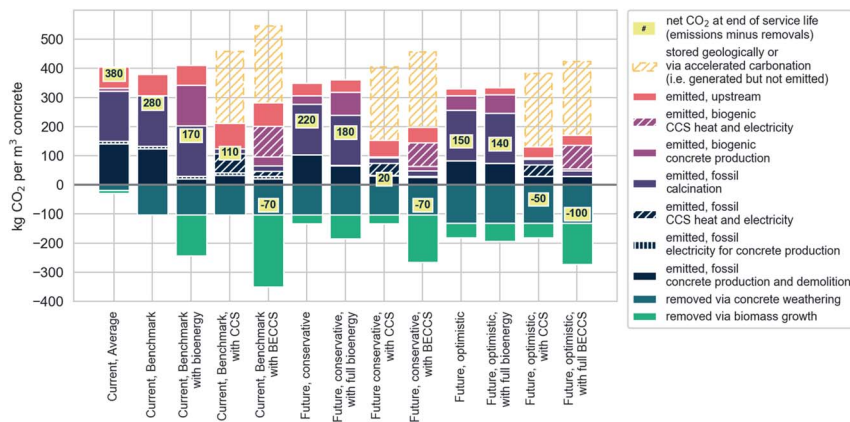


Fig. 5 Life cycle net CO<sub>2</sub> for 30 MPa ordinary Portland concrete in different technology scenarios. Note that the net CO<sub>2</sub> is the CO<sub>2</sub> balance at end-of-life. Concrete was assumed to be used as a 20 cm exterior wall. In all scenarios besides “Current, average”, concrete was assumed to be left to recarbonate following demolition.

For embodied cement production, the net CO<sub>2</sub> of the benchmark scenario is 880 kg CO<sub>2</sub> per t cement *versus* 920 kg per t for the “current, average” production scenario. All BECCS cases, as well as the “optimistic” future cases with CCS only (which have a 45% biogenic kiln fuel mix), resulted in CO<sub>2</sub>-negative concrete, with atmospheric CO<sub>2</sub> removals during biomass regrowth and concrete recarbonation exceeding CO<sub>2</sub> emitted during the concrete’s life cycle. However, as Fig. 6 illustrates, the net CO<sub>2</sub> refers to the CO<sub>2</sub> balance at concrete’s end-of-life; 50 years after most CO<sub>2</sub> emissions occur. In all cases, the concrete system is CO<sub>2</sub>-positive for at least 40 years. Additionally, all cases with bioenergy use have higher net CO<sub>2</sub> than their fossil counterparts for 15–25 years after concrete production until sufficient atmospheric CO<sub>2</sub> is reabsorbed by sustainably replanted biomass. This is seen in Fig. 6 where the lines of the bioenergy cases cross those of their fossil counterparts. The future scenarios’ bioenergy cases have flatter curves, attributable to high proportion of both annual biogenic wastes and fossil-based wastes as fuels for cement production. Finally, significant carbonation of demolished concrete is required to reach CO<sub>2</sub> negativity in all but the “future, optimistic” BECCS cases.

The benchmark cases are discussed below, followed by an assessment of accelerated carbonation options, then sensitivity analyses on natural carbonation, biomass CO<sub>2</sub> uptake, and production efficiencies.

### Benchmark cases

The benchmark case was estimated to emit 380 kg CO<sub>2</sub> per m<sup>3</sup> concrete during production and upstream processes, of which 280 kg are direct emissions from cement production (175 kg from calcination, 105 kg from fuel use). A further 70 kg m<sup>-3</sup> were emitted upstream, of which 60% were from transport of bulk materials. In our model, 20 kg CO<sub>2</sub> per m<sup>3</sup>, or 11% of calcination CO<sub>2</sub>, were removed by natural carbonation over the 50 year service life of the concrete. Finally, carbonation of the demolished concrete removed an additional 85 kg CO<sub>2</sub> per m<sup>3</sup> from



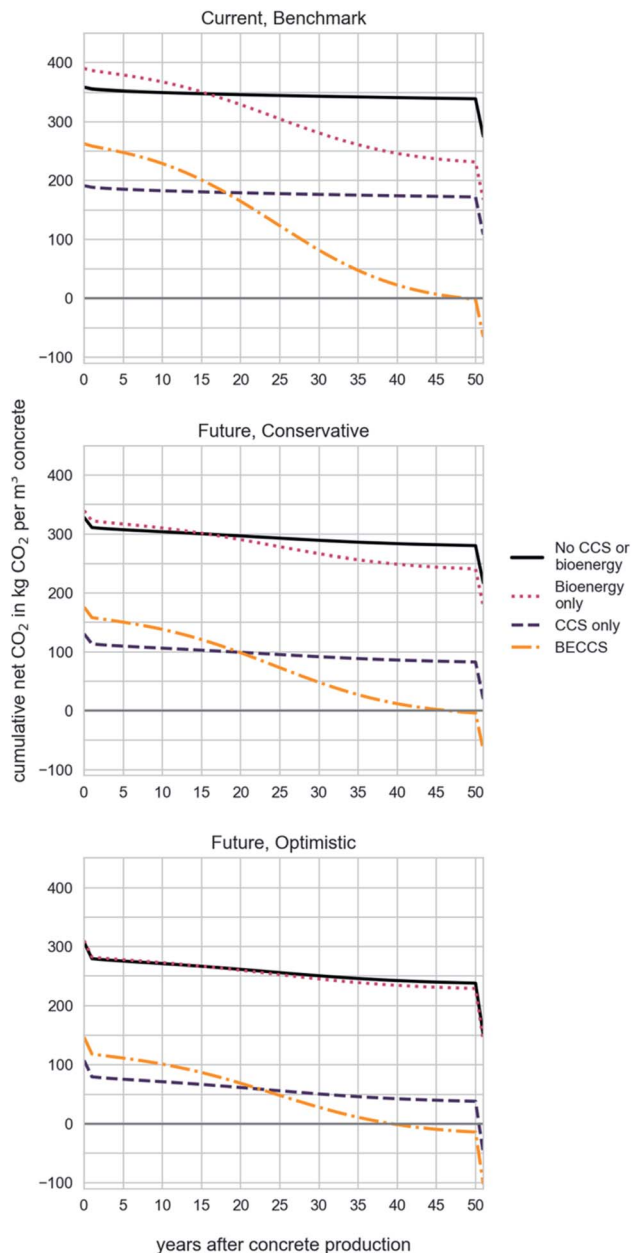


Fig. 6 Cumulative life-cycle net CO<sub>2</sub> of 30 MPa ordinary Portland concrete over time, in different technology scenarios, assuming a 50 year concrete service life, and a 50 year rotation period for long-rotation biomass. Concrete was assumed to be used as a 20 cm exterior wall and left to recarbonate following demolition.

the atmosphere, while the demolition process was responsible for 20 kg m<sup>-3</sup> of CO<sub>2</sub> emissions.

Without accelerated carbonation or CCS, the use of bioenergy in cement production decreased net CO<sub>2</sub> by 110 kg m<sup>-3</sup> concrete. CO<sub>2</sub> emissions of



production increased by 30 kg CO<sub>2</sub> per m<sup>3</sup>, but 140 kg m<sup>-3</sup> of CO<sub>2</sub> were reabsorbed by biomass regrowth. In contrast, the use of CCS in cement production alone reduced both CO<sub>2</sub> emitted and net CO<sub>2</sub> by 165 kg CO<sub>2</sub> per m<sup>3</sup>, corresponding to 250 kg CO<sub>2</sub> sent to geologic storage minus 85 kg CO<sub>2</sub> per m<sup>3</sup> emitted as a consequence of CCS, of which 65 kg were direct emissions from energy provision for the capture unit.

The combination of bioenergy and CCS in cement production generates 70 kg more CO<sub>2</sub> per m<sup>3</sup> concrete than CCS alone, stores 15 kg more CO<sub>2</sub>, and approximately 350 kg of CO<sub>2</sub> per m<sup>3</sup> are removed by biomass. The net effect is that concrete produced with BECCS cement was modelled to be net CO<sub>2</sub>-negative, at approximately -70 kg m<sup>-3</sup>, but only at end-of-life, after CO<sub>2</sub> reuptake by biomass and the carbonation of concrete both during service life and after demolition.

### Accelerated carbonation

Table 3 summarizes the modelling results for accelerated carbonation, in relation to the benchmark case without bioenergy or CCS use. The injection of 0.3% CO<sub>2</sub> during concrete mixing was the only case that resulted in a decrease of net CO<sub>2</sub>, (of 9 kg CO<sub>2</sub> per m<sup>3</sup>), though less than 1 kg CO<sub>2</sub> per m<sup>3</sup> concrete was stored in the concrete. The 5% reduction in cement demand due to the increase in concrete strength reduced CO<sub>2</sub> emissions by 15 kg CO<sub>2</sub> per m<sup>3</sup>, partially offset by decreased natural carbonation from the lower cement content of the concrete. Additionally, as the CO<sub>2</sub> intensity of cement production decreases, so does the apparent decarbonization benefit of reduced cement use. In the BECCS case, the net CO<sub>2</sub> of 0.3% CO<sub>2</sub> injection is actually 8 kg CO<sub>2</sub> per m<sup>3</sup> higher than without, as decreased use of CO<sub>2</sub>-negative cement increases the net CO<sub>2</sub> of the concrete. However, the benefits of decreased resource use are pertinent, even if they are outside the scope

Table 3 Impact of accelerated carbonation on net CO<sub>2</sub> of concrete, with main contributing factors

| Parameter  | No accelerated carbonation            | 0.3% CO <sub>2</sub> injection, 5% strength gain                      | 10% CO <sub>2</sub> curing, no strength gain | 10% CO <sub>2</sub> curing, 10% strength gain |
|--|---------------------------------------|---|--|---|
|  | kg CO <sub>2</sub> per m <sup>3</sup> | kg CO <sub>2</sub> per m <sup>3</sup> , as change from benchmark case |  |   |
| Net CO <sub>2</sub>  | 280                                   | -9  | +42  | +19   |
| CO <sub>2</sub> stored in concrete                           | 0                                     | +<1   | +17  | +15   |
| CO <sub>2</sub> emitted, CO <sub>2</sub> acquisition         | 0                                     | +<1   | +11  | +10   |
| CO <sub>2</sub> emitted, carbonation losses                  | 0                                     | <1  | +11  | +10   |
| CO <sub>2</sub> emitted, cement production and upstream      | 295                                   | -15   | 0  | -29   |
| CO <sub>2</sub> removed by natural carbonation, service life | 20                                    | -1  | -10  | -11   |
| CO <sub>2</sub> removed by natural carbonation, demolition   | 85                                    | -4  | -6   | -14   |
| Total electricity use (kW h per m <sup>3</sup> concrete)     | 24 kW h                               | -1 kW h   | +10 kW h                                     | +7 kW h                                       |



of this study. If scaled to the 2018 EU production of 250 million m<sup>3</sup> ready-mix concrete,<sup>25</sup> 0.3% CO<sub>2</sub> injection would store only 0.1 Mt of CO<sub>2</sub>, but a 5% reduction cement demand would reduce cement sector emissions by 3.5 Mt CO<sub>2</sub> per year.

As modelled, OPC cured with the equivalent of 10% of calcination CO<sub>2</sub>, without strength gain, increased net CO<sub>2</sub> by approximately 40 kg CO<sub>2</sub> per m<sup>3</sup> relative to the benchmark case without accelerated carbonation, bioenergy, or CCS. This increase was due to additional emissions from capture, transport, and injection of CO<sub>2</sub>; CO<sub>2</sub> lost during the injection process; and reduced natural carbonation. However, this process also stored 17 kg of fossil CO<sub>2</sub> into the concrete. If these avoided emissions are included in the net CO<sub>2</sub> of the concrete system, net CO<sub>2</sub> only increased by 23 kg CO<sub>2</sub> per m<sup>3</sup>. Even if CO<sub>2</sub> injection was assumed to be 100% efficient, with no losses of CO<sub>2</sub>, the net CO<sub>2</sub> would still be higher than without accelerated carbonation.

However, “avoided emissions” reflect a reduction in CO<sub>2</sub> emitted from preventing the release of CO<sub>2</sub> and are not a physical removal of CO<sub>2</sub> from the atmosphere, though both reduce the amount of CO<sub>2</sub> that would have been in the atmosphere than if the “avoided” CO<sub>2</sub> would have been emitted. Thus, for accelerated carbonation, claiming avoided emissions requires that the CO<sub>2</sub> stored in the concrete would have otherwise been emitted, and not sent to geologic storage or otherwise abated. If the CO<sub>2</sub> would have been otherwise abated, there are no avoided emissions. Finally, it is important that avoided emissions are not double counted. In other words, they should only be accounted once—either in the system of CO<sub>2</sub> generation or in the system of CO<sub>2</sub> storage (here, accelerated carbonation), but not both.

Fig. 7 shows the impact of CO<sub>2</sub> origin and concrete strength gain for 10% CO<sub>2</sub> curing. The 0.3% CO<sub>2</sub> injection case is not included, as the CO<sub>2</sub> quantity is too small. Assuming there is no gain in concrete strength, only CO<sub>2</sub> curing using a raw flue gas from a biogenic source had a net CO<sub>2</sub> on par with the benchmark case, if avoided emissions can be counted. A 10% gain in strength was sufficient to offset

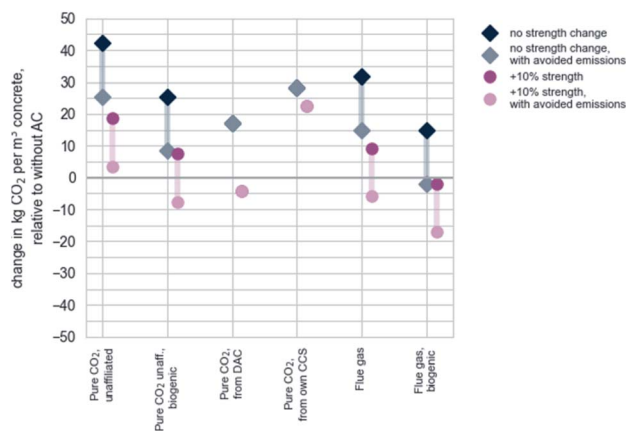


Fig. 7 Impact of CO<sub>2</sub> source on life cycle net CO<sub>2</sub> of concrete subject to CO<sub>2</sub> curing to equivalent of 10% of embodied calcination CO<sub>2</sub>, considered with and without strength gain and avoided emissions.



the emissions associated with accelerated carbonation, and achieve a small net CO<sub>2</sub> reduction (5–20 kg m<sup>-3</sup>) relative to the benchmark case when accelerated carbonation used raw flue gas, pure biogenic CO<sub>2</sub>, or CO<sub>2</sub> from direct air capture (if the CO<sub>2</sub> from DAC fuel use is also captured). However, in the cases where raw flue gas is used for curing, a 10% strength gain may be unrealistic, as the lower CO<sub>2</sub> concentration in the curing environment is likely to result in lower CO<sub>2</sub> uptake.<sup>41</sup> Lastly, using CO<sub>2</sub> from a CCS-equipped cement plant only increased CO<sub>2</sub> emissions, compared to the more efficient option of sending the CO<sub>2</sub> to geologic storage, as, in our model, the net impact of increased energy use and decreased natural carbonation from accelerated carbonation exceeds the CO<sub>2</sub> emissions of sending the CO<sub>2</sub> to geologic storage.

### Natural carbonation

**Exposure conditions.** Uptake of CO<sub>2</sub> by natural carbonation decreases over time, as the CO<sub>2</sub> must continually penetrate deeper into the concrete to achieve additional carbonation. In our model, for a 50 year service life, 50% of natural carbonation occurs by year 12, and 75% by year 28. At the benchmark exposure conditions, carbonation decreased to less than 0.3 kg CO<sub>2</sub> per m<sup>3</sup> concrete per year after year 20. Doubling the concrete service life increases CO<sub>2</sub> removal by only 8 kg CO<sub>2</sub> per m<sup>3</sup> over years 51–100.

As shown in Fig. 8, in the most favourable conditions for concrete carbonation—outdoors, uncovered, and sheltered from rain—OPC was estimated to absorb 23% of calcination CO<sub>2</sub> over 50 years, double that of the benchmark case. Indeed, an equivalent amount of carbonation occurs in 12 years in these conditions as in 50 years in the benchmark case. In contrast, OPC in ground, such as in road sub-layer applications, or otherwise unexposed, was estimated to absorb less than 5% of calcination CO<sub>2</sub> over 50 years.

**End-of-life carbonation.** Alone, natural carbonation after demolition has the potential to abate 20–30% of life cycle CO<sub>2</sub> emissions of OPC. The additional energy use could be negligible, if there is no additional transport distance, *e.g.* left to carbonate for several weeks at the site of demolition or reuse/disposal.<sup>29</sup> However, this abatement only occurs at the very end of the concrete's service life, 50 years after the emissions of its production.

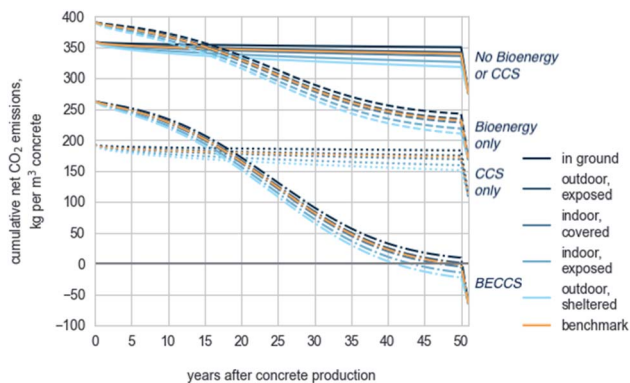


Fig. 8 Impact of concrete exposure conditions on net CO<sub>2</sub> over time.



Though end-of-life carbonation is a long-term decarbonization option for new concrete production, 368 Mt of mineral construction and demolition waste, mostly concrete, was generated in the EU in 2018.<sup>30</sup> Assuming that, as in our benchmark case, 85 kg CO<sub>2</sub> per m<sup>3</sup> concrete is absorbed by end-of-life carbonation, current demolition wastes could remove on the order of 30 Mt of CO<sub>2</sub> per year from the atmosphere, or over 25% of direct CO<sub>2</sub> emissions from EU cement production.<sup>6</sup>

### Biomass use

Fig. 9 shows the impact of biomass rotation period on the net CO<sub>2</sub> of concrete over time. In all cases, the net life cycle CO<sub>2</sub> of the bioenergy cases does not decrease below that of the corresponding fossil energy case until approximately halfway through the biomass rotation period. Until then, the amount of CO<sub>2</sub> in the atmosphere from the production of the concrete is higher than without bioenergy.

This model assumed the use of charcoal for clinker kiln fuel in the bioenergy cases to provide a clear picture of the use of long-rotation biomass. However, from a resource perspective, this is overly simplistic. If all 180 Mt per year of EU-28 cement production (2018)<sup>6</sup> was charcoal-fired, it would require over 40 Mt per year of timber, nearly a quarter of current annual European forestry production.<sup>42,43</sup> If all cement production also installed CCS, a further 20 Mt per year of wood chips would be needed to supply energy for the CO<sub>2</sub> capture reboiler. Instead, energy demand for CCS and part of the energy demand for the kiln could be provided by low-grade fuels, such as agricultural residues or dedicated annual energy crops, which would also decrease the average rotation period of the biomass.

### Efficiencies of production and background supply chains

Tabular results are available in the ESI.<sup>†</sup>

**Electricity.** The benchmark scenario assumes electricity with a life cycle CO<sub>2</sub> intensity of approximately 400 g CO<sub>2</sub> per kW per h. In cases without CCS, full decarbonization of the electricity supply chain decreased CO<sub>2</sub> emissions by less than 10 kg CO<sub>2</sub> per m<sup>3</sup> concrete in the benchmark case. This increases to 25 kg

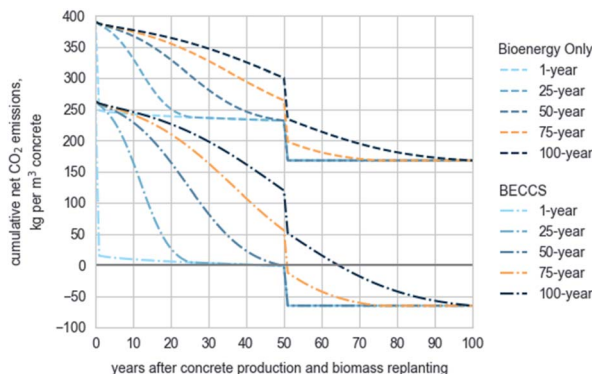


Fig. 9 Impact of biomass rotation period on net CO<sub>2</sub> over time.



CO<sub>2</sub> per m<sup>3</sup> in the CCS case, as the use of CCS in cement production doubles the electricity intensity of concrete production. If CCS is applied to the 180 Mt of 2018 EU cement production,<sup>6</sup> it require 22 TW per h of electricity, or over 2% of total current industrial electricity use.<sup>30</sup>

**Transport.** The production of concrete requires the transport of large quantities of bulk materials, and correspondingly the life cycle CO<sub>2</sub> of concrete is sensitive to transport assumptions. Our model assumed 200 km of rail transport of minerals to the cement plant and 200 km of heavy lorry transport of concrete production inputs, together responsible for over 10% of lifecycle CO<sub>2</sub> emissions in the benchmark case. A decarbonized transport sector could reduce CO<sub>2</sub> emissions by 40 kg CO<sub>2</sub> per m<sup>3</sup> of concrete. Conversely, longer transport distances, the use of less efficient or lower-capacity lorries, and/or a reliance on road transport for inputs to the cement plant will rapidly increase the CO<sub>2</sub> intensity of concrete production.

**Clinker kiln efficiency.** The efficiency of the clinker kiln was most significant to net CO<sub>2</sub> in cases without bioenergy or CCS. An exceptionally efficient clinker kiln, with a thermal energy demand of 2.5 GJ per t clinker would only decrease lifecycle CO<sub>2</sub> emissions by 25 kg CO<sub>2</sub> per m<sup>3</sup> concrete compared to the benchmark of 3.3 GJ per t clinker. In the BECCS cases, a high-efficiency kiln *increased* life cycle net CO<sub>2</sub>. This is the phenomenon of “inefficient BECCS”, where decreased energy efficiency increases the amount of biogenic CO<sub>2</sub> that can be captured and stored.<sup>44</sup>

**CO<sub>2</sub> capture efficiency.** In the cases with CCS, increasing energy efficiency of CO<sub>2</sub> capture to 2.5 GJ per t CO<sub>2</sub> decreases CO<sub>2</sub> emissions by approximately 10 kg CO<sub>2</sub> per m<sup>3</sup> concrete. While a reboiler duty below 3.0 GJ per t CO<sub>2</sub> is possible with MEA, it is more commonly seen with advanced solvents, and thus may also impact the life cycle CO<sub>2</sub> based on differing upstream impacts of solvent production, though solvent production currently represents less than 0.1% of CO<sub>2</sub> emissions in all cases.

If CO<sub>2</sub> capture is applied to the steam boiler providing heat for the solvent reboiler, a net reduction of 85% of boiler CO<sub>2</sub> could be achieved, 40 kg CO<sub>2</sub> per m<sup>3</sup> concrete in the CCS-only case and 90 kg CO<sub>2</sub> per m<sup>3</sup> in the BECCS case. However, the BECCS case would still take over half the biomass rotation period to reach carbon neutrality.

The use of waste heat, if available, could instead reduce CO<sub>2</sub> emissions by 50 kg or 105 kg CO<sub>2</sub> per m<sup>3</sup> concrete for the CCS-only and BECCS cases, respectively. However, in the BECCS case, this reduction in CO<sub>2</sub> emissions is offset by the reduced removal of atmospheric CO<sub>2</sub> by biomass, and therefore the reduced emissions and resource use is not reflected in net CO<sub>2</sub>.

### Considerations beyond the scope of this study

This paper considered concrete produced with ordinary Portland cement, and the decarbonization potential of low-CaO cements were outside the scope of this study. However, as of 2017, the average clinker ratio of European cement was 77%, with the remainder replaced with other cementitious materials such as fly ash and granulated blast furnace slag,<sup>25</sup> and decreasing the proportion of clinker is one of the major avenues of decarbonation proposed by CEMBUREAU, the European cement industry group.<sup>4</sup> The use of ash and slag as clinker replacers lowers CO<sub>2</sub> emissions within the system boundaries of concrete production and





may increase the rate of natural carbonation.<sup>9</sup> However, while fly ash and blast furnace slag can reduce the demand for fresh clinker, they are products of the combustion of fossil fuels and calcination of limestone, and therefore, the fate of the CO<sub>2</sub> from their system of origin should be taken into account when assessing their decarbonization potential. Furthermore, the availability of fly ash is expected to decrease as coal is phased out of the power mix.

Secondly, this study did not consider other contributors to global warming potential, such as methane and dinitrogen oxide emissions or indirect land use, which could be significant for bioenergy-based systems. We also did not consider other global warming impacts from concrete use, though, like avoided emissions, this is specific to the reference system considered, *i.e.*, whether concrete replaces surfaces with lower albedo, (*e.g.*, asphalt) or replaces surfaces that provide evaporative cooling (*e.g.*, grass).

Finally, this was a study on the *marginal* production of 1 m<sup>3</sup> of concrete, and therefore cannot embody the decarbonization potential of reducing the total production of concrete by improved construction design, increased reuse, or extended concrete service life. In particular, this study assumed a concrete use life of 50 years, based on the expected lifespan of modern reinforced concrete structures. This short lifespan for concrete is a modern phenomenon, resulting from the use of iron-based reinforcing bars (rebar). These allow for the construction of very large and strong structures, but corrode and expand as oxygen invades the concrete, causing irreversible structural damage.<sup>45</sup> However, the use life of concrete could be re-extended to multiple hundreds of years if it is unreinforced, or reinforced with non-corroding rebar, such as aluminium bronze.<sup>45</sup> This would greatly decrease the future impacts of the concrete industry, as the concrete stock becomes more durable, reducing overall resource use.

## Conclusions

This paper explored the production of ordinary Portland concrete considering different combinations of natural and accelerated carbonation, bioenergy use, and carbon capture and storage. The sensitivity analysis explored the impact of strength gain from accelerated carbonation, the origin of CO<sub>2</sub> used for carbonation; conditions of concrete use and demolition on natural carbonation; biomass rotation period; and efficiencies of electricity, transport, and cement production.

In our model, the aggressive use of BECCS in cement production and the deliberate natural recarbonation of demolished concrete together resulted in net-CO<sub>2</sub>-negative concrete at current technology levels, when considered on a life cycle basis.

However, net CO<sub>2</sub> is the balance of CO<sub>2</sub> emissions and removals for the entire concrete life cycle, measured at the end of the concrete's service life and after all biomass used for bioenergy has been regrown. Depending on the biomass rotation period and the rate of concrete carbonation, CO<sub>2</sub>-negative concrete may still have a net-positive CO<sub>2</sub> balance for the entirety of its service life and only reach CO<sub>2</sub> negativity when the demolished concrete is allowed to recarbonate.

Modelled with currently available technology, post-combustion amine-based CCS for cement production reduced life cycle CO<sub>2</sub> of concrete by 40%, and was the single most effective decarbonization intervention, but alone is insufficient to



result in negative emissions. Combined with the use of fully biogenic fuel in the cement kiln, biogenic fuel or waste heat for CO<sub>2</sub> capture, and allowing for carbonation of demolished concrete, BECCS was estimated to result in a life cycle net CO<sub>2</sub> of -70 kg CO<sub>2</sub> per m<sup>3</sup> concrete. However, 280 kg CO<sub>2</sub> per m<sup>3</sup> were still emitted during production and in upstream supply chains, more than with CCS alone. It is not until almost halfway through the biomass rotation period that the net CO<sub>2</sub> of BECCS is lower than in the CCS-only case, assuming that the biomass is indeed sustainably regrown. Using short rotation biomass for cement kiln fuel and encouraging carbonation of current concrete waste can be used to more rapidly decarbonate the concrete sector.

In this study, accelerated carbonation of ordinary Portland concrete did not appear to be an efficient method for CO<sub>2</sub> storage on its own. The CO<sub>2</sub> penalty from increased energy use and decreased natural carbonation exceeded the CO<sub>2</sub> stored, though this was highly sensitive to both concrete strength gain and the origin of the CO<sub>2</sub> used for accelerated carbonation. The potential benefit of accelerated carbonation seems to lie not in its ability to directly store CO<sub>2</sub> in the concrete, but rather if it can increase concrete strength and reduce the overall use of cement.

The natural carbonation of concrete is a slow process, and though estimates of total carbonation by global concrete stocks are impressive, the annual CO<sub>2</sub> uptake of in-use concrete is minor relative to the embodied CO<sub>2</sub> of its production. However, increasing carbonation during demolition and recovery by leaving the concrete waste exposed to air for a period of weeks is a promising decarbonization option that could be implemented in the near term.

Decarbonization of concrete production is a complex matter, and CO<sub>2</sub> emissions, while important, do not embody the full impacts of the system of concrete production and use. The net CO<sub>2</sub> must be taken in the context of the full specific systems for concrete production. Trade-offs between near-term *versus* long-term decarbonization and between decreased CO<sub>2</sub> *versus* increased energy use must be considered. Even if CO<sub>2</sub>-neutral, or CO<sub>2</sub>-negative, concrete is achievable, it is very likely to be at the expense of increased resource use. Therefore, the primary decarbonization priority should always be the reduced use of all concretes *via* all production methods.

## Author contributions

S. E. Tanzer: conceptualization, methodology, software, data curation, writing – original draft, writing – review & editing, visualization. K. Blok: writing – review & editing, supervision. A. Ramirez: conceptualization, writing – review & editing, supervision.

## Conflicts of interest

There are no conflicts to declare.

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