**Is crushed concrete carbonation significant enough to be considered as a carbon mitigation strategy?**

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Abstract

When addressing concrete carbonation as a carbon mitigation option, studies leave out the effect that a temporal difference between the CO\(_2\) emissions and uptake happening throughout concrete’s life cycle have on climate change. In this study, the role played by carbonation on concrete’s carbon mitigation potential is investigated through a dynamic life cycle assessment, to properly position CO\(_2\) uptake and release. The carbon balance in concrete structures built and demolished from 2018 to 2050 is modelled as a case study. The potential uptake due to crushed concrete carbonation is over 9% of the cumulative global warming effect of concrete manufacturing. It is comparable to the reduction potential of the most promising strategy, namely replacing clinker, totaling 12%. If stimulated in a wide scale, crushed concrete carbonation can push the industry towards meeting carbon mitigation targets faster. Future environmental impact assessments should rely on dynamic models to increasingly consider this phenomenon.

1. Introduction

Concrete has a remarkable performance in terms of versatility, strength and durability. For that reason, mankind has relied on it to build and operate urban environments for centuries [1]. However, awareness on the CO\(_2\) intensity that accompanies cement and concrete production has risen in the past decade [2–5]. Still, the technical features of cement and concrete seem to outweigh their environmental shortcomings, for their use continues to grow [6, 7].

The updated Cement Technology Roadmap [8] explores a possible low CO\(_2\) emissions pathway expected to reduce cement making’s direct CO\(_2\) emissions by 24% below current levels until 2050 [8]. The main carbon mitigation strategies envisioned by the roadmap are (a) improving energy efficiency, (b) switching to alternative fuels, (c) reducing the clinker to cement ratio and (d) integrating carbon capture into cement production.

While highest potential CO\(_2\) savings in cement and concrete manufacturing are usually associated with those main strategies, carbonation could imply a reduced contribution to climate change. Carbonation happens when the calcium oxide in cement-based products chemically reacts with atmospheric CO\(_2\), generating calcium carbonate (CaCO\(_3\)). Seldom considered in environmental assessments, carbonation is commonly investigated in durability studies of reinforced concrete structures, since the CO\(_2\) absorption can be a catalyst for steel corrosion. The few studies positioning carbonation as a potential carbon uptake solution typically compared direct CO\(_2\) emissions due to calcination during clinker production with CO\(_2\) uptake due to carbonation [9–11]. Greenhouse gas (GHG) emissions associated to fuel burning or other processes involved in concrete manufacturing are left out.

A common consensus in the few studies that position carbonation as a potential strategy to reduce concrete’s carbon footprint is that carbonation of crushed concrete is a promising strategy to increase natural CO\(_2\) absorption, due to the increased surface area exposed to atmospheric CO\(_2\) [7, 10, 12].
However, an important aspect systematically left out of concrete carbonation studies is the temporal difference between the CO₂ emissions during manufacturing, the slow gradual uptake during service life, and the comparably faster uptake happening with crushed concrete. As carefully discussed in life cycle assessments (LCAs) of biobased materials that position biogenic CO₂ uptake versus release at the end of life \[13, 14\], the effects of this carbon balance in the atmospheric radiative forcing, and consequently on climate change, is different depending on the timing of the emission or uptake. In order to properly position the uptake against the emission, one should rely on dynamic global warming characterization factors \[13\].

 Humanity has a short feasible time frame to limit global warming below 1.5 °C \[15\] and, therefore, a wide portfolio of carbon mitigation measures must be implemented. This study aims at investigating the role played by carbonation on concrete’s carbon-mitigation potential. A dynamic LCA was performed to estimate the carbon balance during years 2018–2050 (figure 1) of all concrete structures in Québec (Canada). The Global Warming Potential (GWP) of the most promising strategies envisioned by the Cement Technology Roadmap, namely increase of alternative fuel and of supplementary cementitious materials (SCMs) use, and deployment and increase in carbon capture and storage (CCS), is contrasted with that of carbonation. Concrete structures built 50 years before the initial year of assessment (1968) and onwards are expected to be crushed in 2018 and onward (assuming structures lasted for 50 years \[16, 17\])—to consider carbonation potential during concrete’s end of life.

2. Method

The following sections describe the research’s four main stages: (a) collection and/or extrapolation of data on cement and concrete production volumes and composition; (b) scenario definition for comparison between carbonation and typical carbon reduction strategies, (c) dynamic GWP calculation, and (d) calculation of impacts in other areas of protection.

2.1. Cement and concrete production volumes and composition

The amount of concrete produced in the set time frame was determined building upon the cement consumption per capita data \[18\] published for Ontario, assuming the same value applied for Québec. To address the variability in average per capita consumption values, all scenarios assessed were built for two figures: 260 kg and 300 kg of cement per capita. Past and future cement production figures are assumed to be correlated with population growth \[19\], which was gathered from historic data and increase estimates published in the Québec Census \[20\].

Total concrete production was determined based on the average amount of cement used for concrete manufacturing \[9\]. To further divide the annual production volumes between ready-mix concrete and precast concrete, the ready-mix production amount was extracted from Wernet et al \[21\] and Martineau \[22\]. It was assumed that the remaining amount
calculated via cement per capita consumption data referred to precast concrete manufacturing. This proportion between both concrete types was corroborated by production data on ready-mix and precast concrete from Nordic countries [12]. The production volume of different strength classes of ready-mix concrete and their composition were extracted from Wernet et al [21] and Martineau [22], whereas the same information for precast concrete came from Athena [23].

2.2. Scenario definition

2.2.1. Carbonation

Carbonation was estimated in three separate exposure conditions: (a) during concrete’s service life, (b) at their end-of-life, in landfills, with particles of 100 mm diameter, and (c) also at their end of life, but assuming that 80% of used concrete is crushed for recycling as aggregate, with particles of 20 mm diameter, and the remaining 20% is landfilled, with 100 mm diameter. To determine the exposure conditions during concrete’s service life, the thicknesses and the distribution of each type of concrete in different structural elements and products were extrapolated from Nordic data [12]. Quebec has the same Köppen–Geiger climate classification as the Nordic countries, which supports the assumption that the thicknesses of construction elements would be similar.

Having all exposure conditions, concrete strength classes and compositions, the theoretical estimation of CO₂ uptake from concrete’s carbonation [24] was made possible, following equation (1):

\[
\text{CO}_2_{\text{uptake}} = k \sqrt{t} \times c \times \text{CaO} \times r \times A \times M
\]

where \( k \) is the carbonation rate coefficient, \( t \) is service life (in years), \( c \) is the quantity of Portland cement per cubic meter of concrete, \( \text{CaO} \) is the amount of CaO content in Portland cement, \( r \) is the proportion of calcium oxide that can be carbonated, \( A \) is exposed surface area, and \( M \) is chemical molar fraction \((\text{CO}_2/\text{CaO})\).

The carbonation rate coefficients were extrapolated from Lagerblad [24]. Correction factors were applied based on type of SCM used and on the presence of surface treatments or covers. In this study, it was assumed that all concrete structures underwent surface treatment and/or were covered.

To estimate the uptake at concrete’s end of life, one can follow the same equation, coupled with information on (a) particle sizes and geometry—to determine exposed surface area, and on (b) the amount of concrete that already carbonated during the service life—to assess the remaining volume still to be carbonated. Particles were considered to be shaped as spheres. To keep the estimations conservative, the lowest carbonation rate for buried structures was used. The impact of crushing the old concrete into smaller particles for recycling as aggregates was also taken into consideration. Data on energy and water demand to crush the material came from Pradhan et al [25].

2.2.2. Cement technology roadmap strategies

The strategies listed in the cement technology roadmap that represent the largest CO₂ emission reduction potentials were considered. The following sections indicate assumptions and data sources for the development of each strategy’s impact assessment.

- SCM use
Two scenarios were set to model the clinker replacement possibilities with SCM:

* A conservative option, assuming that the reduced clinker content would be met by increased use of limestone filler, meeting the Canadian standard for the so-called ‘general use limestone cement’ [26].

* A more promising option, assuming that the reduced clinker content would be covered by increasing fly ash and ground granulated blast furnace slag use in cement making.

The conservative option implicates in not meeting the full clinker replacement ratio predicted by the cement technology roadmap for 2050, since limestone filler incorporation has a limit of 15% [26]. This lower threshold was set considering that most used SCMs are co-products from other industries and might not meet the production demand for cement. The latter option, while promising for CO₂ reduction, relies on the availability of co-products from steel and thermal energy production, which is expected to decrease considerably in next decades [27]. Data sources for production of different types of SCM, cement and concrete are summarized in the supplementary file.

- Alternative fuel use.
The Cement technology roadmap assumes that the global average of alternative fuel use in 2014 was 5.6% [8]. In Quebec, however, alternative fuel use represented 30% of the heating demand for cement making [28]. To model the evolution of alternative fuel use until 2050, the trendline for the increased global proportion from 2014 until 2050 listed in the international report was identified and applied to current data for the Canadian province.

Analogously to the previous strategy, two scenarios were set:

* A conservative option, assuming the increase could be met by using the same average waste fuel mix adopted today in Quebec [29], and

* A more promising option, assuming that the energy demand would be met by a waste mix based exclusively on biobased waste fuels.

For the latter, it was assumed that biogenic CO₂ emissions due to burning of biobased waste fuels did
not contribute to the climate change effect. The burning of biobased waste fuels modelling relied on the emissions listed in Treyer [30]. Any other upstream data related to wood chip production was disregarded based on an assumption that in the best-case scenario the biobased energy would be supplied by waste. In some cases the required transportation of waste biomass could be prohibitive, increasing CO₂ emissions. This was, however, not considered in this study. The total energy demand, the average fuel mix, and the average waste fuel mix used in cement kilns in Québec were taken from Athena [29].

- Carbon capture and storage (CCS).

The deployment of CCS was assumed to rely solely on the post-combustion capture technology with mono-ethanolamine, as this is the simplest to implement in existing cement plants [31]. Data on the heat and electricity demand and on absorbant requirement to capture and compress the emitted CO₂ came from Volkart et al [32]. Actual permanent storage of CO₂ was not modelled. The limited market share of the technology prevents collection and/or extrapolation of reliable data for the studied context. It must be noted that the reduction potential documented in the technology roadmap was assumed to refer to net reduction. The model for CCS installation and maintenance was nonetheless needed for the other impact categories calculation.

### 2.2.3. Transport

To account for transportation impacts, a knowingly important variable in the environmental attractiveness of crushed concrete [33, 34], two scenarios were built: both newly manufactured concrete and crushed concrete for recycling were assumed to be transported through a distance range of 150–300 km and 30–50 km, respectively. The freight modelling relied on Levova [35].

### 2.3. Dynamic GWP calculation

Once the production cycle of each type of concrete was modelled, it was possible to determine the yearly amounts of main GHGs (CO₂, CO, N₂O, and CH₄) (a) emitted, considering new concrete production and transport and old concrete crushing and transport, and (b) absorbed, considering concrete carbonation during use and end of life, in the two previously described conditions (landfilled and crushed). Emissions associated to material manufacturing from raw material extraction up to production itself are assumed to take place in one year. With that information at hand, the carbon balance associated to concrete structures from 2018 to 2050 was calculated by relying on dynamic characterization factors (DCF) for GWP.

To determine these characterization factors, one can multiply the so-called ‘atmospheric load’ of a GHG by the instantaneous radiative forcing of that same gas. For CO₂, the atmospheric load is given by equation (2), representing the decay of CO₂ in time, after the initial unitary impulse at $t = 0$ [36]. For other GHGs, the decay pattern follows a first order decay equation. Both the atmospheric load and the instantaneous radiative forcing are time dependent, so one can integrate it considering yearly time steps (equation (3), as proposed by Levasseur [13]), to have a DCF for each year in an analysis. The DCF for a given year is then multiplied by the emission and/or uptake of GHG happening at the corresponding year:

$$C_{CO_2}(t) = a_0 + \sum_{i=1}^{3} a_i * e^{-\frac{t}{\tau}}$$  \hspace{1cm} (2)

Where:

$C_{CO_2}(t)$ is the decay pattern of a CO₂ pulse emission (e.g. 1 kg CO₂).

$a_i$ are the coefficients for the calculation of CO₂ fractions remaining in the atmosphere. They have the values: $a_0 = 0.217; a_1 = 0.259; a_2 = 0.338; a_3 = 0.186$.

$\tau_i$ is the perturbation time, $\tau_1 = 172.9$ years, $\tau_2 = 18.5$ years, and $\tau_3 = 1.186$ years:

$$DCF_{inst,CO_2}(t - t_j) = \int_{t_j}^{t} A_{CO_2} \cdot C_{CO_2}(t) \, dt$$ \hspace{1cm} (3)

Where:

$A_{GHG}$ are the specific radiative forcing per unit mass, calculated according to Hartmann et al [37].

$t_j$ is the time of occurrence of a pulse emission or uptake, and $t$ is the time of the effect ($t - t_j$ is the yearly time step).

The calculations described by equations (2) and (3) were performed via an excel based tool [38].

### 2.4. Impact on human health, resources and ecosystem

The LCIA of all carbon-reducing measures within each scenario was modelled using the Recipe 2016 v1.1. method, with characterization factors defined for the endpoint level. The method allows for modelling following three different perspectives according to the Cultural Theory of Thompson [39]: the individualistic, hierarchist, and egalitarian perspectives. Here, the hierarchist perspective was adopted, categorised by the method developers as the ‘default’ approach [40]. The modelling of endpoint indicators still does not allow for a dynamic calculation to be structured as was done here for GHGs.

### 3. Results

#### 3.1. The significance of carbonation as a carbon reduction measure

Figure 2 graphically depicts the potential dynamic Global Warming reductions, alongside
Figure 2. Dynamic GWP of ‘business as usual’ (BAU) concrete manufacturing versus manufacturing with reduction strategies. SCM = supplementary cementitious material, CCS = carbon capture and storage, EoL = end of life, RCA = recycled concrete aggregate, i.e. crushed concrete. To ease interpretation of the boxplots, the median value for the BAU scenario’s GWP is depicted in the horizontal arrows.

The assessment showed that, within the years 2018–2050, the carbonation during the service life of all concrete produced (disregarding crushed concrete) represents approximately 1.6% of the global warming effect of concrete manufacturing (figure 2, orange boxplot’s median), which denotes an almost negligible reduction. If concrete crushed in 2018 and onwards (manufactured in 1968 and onwards) would be landfilled (particles with 100 mm diameter), the percentage of cumulative uptake in the crushed particles would be slightly under 3% of total cumulative GWP (figure 2, purple boxplot’s median). If, on the other hand, 80% of used concrete would be crushed into smaller particles (approx. 20 mm diameter), and 20% would be landfilled, the cumulative proportion in 2050 would increase to over 9% (figure 2, turquoise boxplot’s median).

When compared to roadmap strategies, one notes that the potential uptake due to carbonation of crushed concrete falls within the range of the most promising strategy, namely increase in SCM use (figure 2, green boxplot). In its best modelled scenario, the potential (cumulative) dynamic reduction due to increased SCM use in 2050 is of less than 12%. The best possible scenario for increase in alternative fuel use (figure 2, yellow boxplot), where all alternative fuels would be biobased (no fossil CO\textsubscript{2} emitted), also implies a cumulative reduction of approximately 12%. As for CCS implementation (figure 2, grey boxplot), the net reduction effect for concrete manufacturing would be of less than 3%, which is similar to the reduction due to carbonation in landfilled concrete. It must be noted, however, that the Cement Technology Roadmap’s vision for CCS predicted a percentage capture starting at zero before 2030 and reaching 25%–29% of direct CO\textsubscript{2} emissions generated in cement making at 2050 [9], the final year of analysis. The greatest expected benefit of CCS implementation is, therefore, outside of the set temporal scope. Also, when bringing the reduction in direct emissions into a calculation that considers life cycle indirect emissions not only for cement but the full concrete production cycle, it is expected that its significance will lose strength.

3.2. Environmental trade-offs of low-carbon measures

The sole focus on GHG reducing measures, ignoring their influence on other impact categories, risks shifting the burden to other environmental effects within one or all ‘Areas of Protection’, known as the entities that LCA aims to preserve: human health, natural environment (also called ecosystem quality), and natural resources (also called resource availability) [41, 42]. Analogously to the reduction depiction described in figure 2, the following figures illustrate the potential consequences of implementing...
the evaluated reduction strategies in human health (figure 3), ecosystem quality (figure 4), and resource availability (figure 5). Natural carbonation during concrete’s service life and in landfill are not plotted due to the consideration that no additional environmental loads are generated in those scenarios.

When assessing the trade-off of deploying the mentioned strategies, one concludes that within all three areas of protection, increasing alternative fuel and SCM use does not implicate in added impacts. In fact, for human health and ecosystem quality, their increased use leads to a notable decrease in impact until 2050, shown in the yellow and green boxplots in figures 3 and 4. While the crushing and transporting of used concrete (turquoise boxplot) is not as environmentally attractive as the previous mentioned strategies, it seems to imply in a statistically insignificant increase of these impact categories. CCS implementation (grey boxplot), on the other hand, considerably worsens concrete’s contribution to all three areas of protection. The heat and electricity requirements to capture and pressurize CO₂ are the major drivers of the identified increase in impact. Other technologies for CO₂ capture in

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**Figure 3.** Human health impact of BAU concrete manufacturing versus manufacturing with reduction strategies. The unit DALY means disability adjusted life years and represent the years that are lost or that a person is disabled due to a disease or accident. SCM = supplementary cementitious material, CCS = carbon capture and storage, EoL = end of life, RCA = recycled concrete aggregate, i.e. crushed concrete. To ease interpretation of the boxplots, the median value for the BAU scenario’s human health impact is depicted in the horizontal arrows.

**Figure 4.** Ecosystem impact of BAU concrete manufacturing versus manufacturing with reduction strategies. The unit species.yr refers to a prediction of local species loss integrated over time. SCM = supplementary cementitious material, CCS = carbon capture and storage, EoL = end of life, RCA = recycled concrete aggregate, i.e. crushed concrete. To ease interpretation of the boxplots, the median value for the BAU scenario’s ecosystem depletion impact is depicted in the horizontal arrows.
cement making are currently being investigated by the industry, which might decrease the significant trade-off identified in this study. When the environmental trade-off of CCS implementation (according to the international roadmap) is interpreted side-by-side with the timid reduction potential identified for GWP until 2050 (figure 2), its attractiveness is put to question.

While climate change is undoubtedly the most pressing global environmental threat, expanding the scope of indicators assessed in environmental investigations such as the one depicted here helps establishing sensible goals, which in turn can lead to lower risk investments by industrial stakeholders. Replacing clinker and fossil fuels in cement manufacturing seems to diminish the impact of the industrial activity in all three areas of protection. Investing in these strategies is thus not only effective from a climate change mitigation perspective (figure 2), but would yield a positive preservation effect throughout the pool of natural resources available to mankind (figures 3–5). Crushed concrete carbonation, on the other hand, should be interpreted as a strategy to push the industry towards meeting its carbon reduction targets faster, yet with a very small potential trade-off for other areas of protection (e.g. low risk investment). Finally, CCS implementation would aid climate change mitigation to a much smaller extent than its contribution to worsening the impact on other environmental categories, if following the more conservative estimations of the international roadmap. While this narrative relies on loose investment analogies to illustrate what seems to be the best available strategies for the industry, these findings are not expected to ground investment decisions but to shed light on which areas deserve more research.

4. Discussion and concluding remarks

The estimations confirmed that carbonation after concrete’s use can imply in a cumulative CO₂ uptake as significant as those of the cement technology roadmap strategies. Even when positioning CO₂ uptake against all life cycle CO₂ emissions associated to concrete, moving away from typical comparisons against direct process emissions only [9, 11], the conclusion is clear: crushed concrete carbonation is significant enough to be considered as a carbon-mitigation strategy.

To grasp the magnitude of the potential uptake in relation to all anthropogenic activities, one must consider current carbon budget predictions consistent with remaining below 1.5 °C warming. For a two thirds chance (approximately 66%) to limit global warming below the set threshold, it is suggested that 420 Gt of CO₂ can still be emitted [37], after which humanity must reach a carbon neutrality status. If this remaining amount of CO₂ were to be divided equally between every person on Earth today (considering a value of 8 billion people as an approximation), each individual would be limited to emitting 52.5 tonnes of CO₂ until 2050 (or 1.6 tonnes of CO₂/year). The cumulative uptake of CO₂ due to crushed concrete carbonation from 2018 to 2050, divided by the predicted population in Québec in 2050, represents less than 0.8% of the total budget per person.
It is estimated that boreal forests in Canada absorbed, from 1990 to 2008, around 28 Mt/year of C\textsuperscript{2} [43], or 103 Mt of CO\textsubscript{2}/year. Considering that Québec accounts for 20% of Canada’s boreal forest [44], the potential carbon sink due to cumulative crushed concrete carbonation happening for 32 years (between 2018 and 2050) represents around 20% of the uptake happening yearly in managed boreal forests.

In the current fight against the seemingly inevitable effects of global warming, the small contribution of crushed concrete carbonation in relation to available budget predictions does not erase its significance. With the increase in demand for urbanized areas, and with the growing population in already dense cities, societies need large, solid, and durable infrastructure which require large amounts of concrete. Proposed carbon reduction strategies in the manufacturing stage could be joined with measures to increase CO\textsubscript{2} uptake during concrete’s end of life, increasing the chances of reaching the goals set to limit global warming.

From the industry stakeholders’ perspective, the estimations on climate change mitigation related to different strategies herein presented could be coupled with the magnitude of investments necessary to implement them, to reach a tactical portfolio that will ensure maximum reduction of GHG emissions with minimum environmental and financial tradeoff. While curbing climate change is—and should be—the current major focus, the industry must be kept aware of unintended effects in other areas, to remain prepared for future environmental challenges.

Estimations of this nature embed large uncertainties. The carbonation phenomenon is incredibly variable and complex to accurately predict. Production estimations, as well as any future forecasts on which strategies will be adopted by the cement and concrete industry and how they shall be upscaled to represent future practice, are well-informed expert ‘guesses’. Still, the authors advocate that to properly position carbon reduction strategies among each other, dynamic GWP modelling should be employed to assure proper comparisons between emission pulses and uptake through time.

In that sense, the method applied in this study is adaptable to any other context or geography, upon adjustment of carbonation rates and degrees, as well as trends in cement and concrete manufacturing. Since the presented estimations were based on conservative carbonation rate coefficients (see section 2 and supplementary information), one might argue that an even more significant CO\textsubscript{2} uptake than the result herein depicted would be possible. In fact, with the increasing concentration of CO\textsubscript{2} in the atmosphere, the rate of carbonation is expected to increase in the future. Lagerblad et al. [24] pointed that calculations involving a perspective of 5–10 decades ahead may need to correct all carbonation rates by a factor of 1.5, to take that effect into consideration. While one hopes the implementation of emission-reducing measures throughout all human activities curbs the increase in atmospheric CO\textsubscript{2} concentration, it is realistic to expect that the crushed concrete carbonation phenomenon will become increasingly significant in the decades to come.

Data availability statement

All data that support the findings of this study are included within the article or references.

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