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Dynamic environmental payback of concrete due to carbonation over centuries

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ABSTRACT

This research introduces a dynamic life cycle assessment (LCA) based carbonation impact calculator designed to enhance the environmental evaluation of cement-based construction products. The research emphasizes the limitations of static LCAs which fail to capture the time-dependent nature of carbon sequestration by carbonation.

We provide an easy-to-use spreadsheet-based LCA carbonation model. The model is available in the supplementary information, and includes a suite of changeable parameters for exploring the effect of alternative environmental conditions and concrete block composition on carbonation. The tool enables use of both a static and dynamic LCA method to calculate the production emissions and carbonation sequestration of a concrete block over a 1000-year time horizon.

Carbonation can partially mitigate initial production emissions and adjust radiative forcing over long periods. Using a static attributional LCA approach, carbonation sequesters 6 % of the CO_2 generated from its production emissions. We describe the ratio of carbonation to production emissions as the partial "carbonation payback", and with dynamic LCA show the variation of this ratio over time. Considering time by applying the dynamic LCA approach, we find this partial "carbonation payback" is split between uptake during the 60-year service life (0.13 kg CO_2) and the 940-year end of life period (0.12 kg CO_2) in our baseline case. Further scenario analyses illustrate the significant variability in carbonation payback, driven by environmental factors, cement composition, and the use of supplementary cementitious materials.

The results highlight the critical role of modelling choices in estimating the carbonation payback. The carbonation calculator developed in this study offers a sophisticated yet user-friendly tool, providing both researchers and practitioners with the ability to dynamically model the sequestration potential of concrete, thereby promoting more sustainable construction practices.

1. Introduction

The built environment is the largest source of greenhouse gas (GHG) emissions among human activities, contributing at least 37 % of global GHG emissions (United Nations Environment Programme, 2023). Concrete is among the many carbon intensive materials used widely in the construction of buildings and other structures (Elliot et al., 2024; Göswein et al., 2019) due to the production of Portland cement. This cement not only requires significant energy to manufacture but also involves calcination of calcareous rocks (Monteiro et al., 2012; Shirkhani et al., 2018). This process, responsible for decarbonating the rocks, accounts for 60–70 % of the CO_2 emissions from clinker production (IEA, 2018). As a result, cement is considered one of the most difficult products to decarbonise.

Cementitious materials also undergo carbonation, a chemical reaction between the hardened cement paste and CO_2 from the atmosphere, resulting in the sequestration and storage of CO_2 in cement-containing structures. Carbonation may have significant benefits that, if assessed, could result in smaller carbon footprints of concrete materials during

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Variables and parameters

c	Environmental CO_2 concentration
C_m	CO_2 sequestration potential
C_s	Mass of CO_2 stored in concrete
f_c	Compressive strength
k_0, k_1, k_2, n	Carbonation front coefficients
m	Mass of concrete
R	Carbonation resistance coefficient
SA	Surface area of reference flow
t	Exposure time
V	Volume of concrete
V_c	Volume of carbonated concrete
x	Depth of carbonation front
y	Cementitious material percent replacement
α, β	Carbon sequestration potential coefficients
ϕ_c	Degree of carbonation

and after the service life (Xi et al., 2016). On the one hand, the carbonation reaction goes some way towards sequestering CO_2 emitted during calcination; on the other hand, it weakens the concrete structure over time. Supplementary cementitious materials (SCMs) can be used to partially replace cement in the concrete manufacturing process, and are associated with potentially lower production emissions, but also lower carbonation sequestration (Jungclaus et al., 2024). However, the understanding of the extent and role of carbonation in lowering these carbon footprints is still emerging.

As carbonation fluxes fluctuate over time, they are challenging to model with static attributional LCA models. Much like modelling biogenic carbon, the timeline of these fluxes is relevant to their climate change impact. Monteiro et al. (2012) explained the importance of time of sequestration by carbonation. This was later captured in the mathematical approach published by Souto-Martinez et al. (2017), and subsequently their scenario analyses on various building and environmental conditions (Souto-Martinez et al., 2018). These latter three studies offer highly parameterised methods particularly useful for modelling potential carbonation and the structural and environmental sensitivities therein. Due to the need to consider these time-sensitive parameters, recent studies have described carbonation fluxes using dynamic life cycle assessment (DLCA) (e.g. Hawkins et al., 2021).

DLCA is an approach that considers the timing of emissions in an LCA, as an alternative to the fixed time horizon used in CO_2-equivalent characterisation methods such as GWP100 (Levasseur et al., 2010; Sohn et al., 2020). Rather than adding together the radiative forcing of multiple GHGs over an arbitrary time period (e.g., 100 years) and presenting the results in CO_2-equivalent units, the DLCA approach measures climate change directly in terms of radiative forcing over time. This has the benefit of representing the important difference between a unit of a gas emitted in one pulse compared to the same amount emitted slowly over many smaller pulses. This is especially important when analysing slow and non-linear emissions and sequestration, such as biogenic fluxes (Levasseur et al., 2013; Meyer et al., 2024), and in this case the slow uptake of CO_2 by concrete (Thonemann et al., 2022).

The objectives of this research are to (1) demonstrate the importance of the dynamic LCA approach in understanding mid-to-long-term effects of concrete use; and (2) to provide a carbonation impact calculator for carbon footprints and LCA constraining the necessary parameterisation for specific construction emissions accounting. As such, with our calculator we aim to illustrate that the simplified nature of arbitrary time-horizons in traditional static attributional LCA is not fit for purpose when considering slow and long-run carbon uptake in concrete building products whose life cycles outlive the service life of most buildings in which they are located. Therefore, in this paper we present both static attribution LCA results (Global Warming Potential, GWP100) and dynamic LCA results (radiative forcing, W/m^2) for a case study of a solid concrete block, including a number of scenarios to demonstrate the level of uncertainty when considering the range of factors upon which the carbonation calculation is contingent.

2. Literature review

Understanding of the role of these in carbon footprints is still emerging. A number of review papers have been published in recent years, although we found few research papers that use DLCA to estimate the impact of carbonation.

There are a number of approaches for estimating concrete carbonation, ranging from relational estimates to laboratory-based mathematical models (Possan et al., 2017). Cuéllar-Franca and Azapagic (2015) analysed a range of carbon capture technologies finding that carbonation can provide between 4 and 48 % sequestration. More recently, Ma et al. (2024) calculated the contribution of cement to China's national carbon footprint, finding that between 1930 and 2021 cement offsetting 50.7 % of cement emissions. This long period of time indicates the importance of time-sensitive calculation methods for carbonation. Striking conclusions such as this have led to carbonation gaining attention as a viable negative emission technology in the construction sector (Jeswani et al., 2022).

Several approaches estimate CO_2 uptake as a percentage of CO_2 emitted during concrete production, such as (Jacobsen and Jahren, 2001) who estimated 11 % of production emissions were sequestered due to carbonation in Norway. The Concrete Centre (2016) reported that carbonation can reduce the carbon footprint of a building by 7.5 % over its life in the United Kingdom. This latter estimate assumed 2.5 % of upstream emissions are sequestered during the service life and a further 5 % during end-of-life. Studies such as Jungclaus et al. (2024) explore the effect of varying multiple influencing factors on carbonation uptake as a fraction of production emissions. Estimating carbonation uptake as a percentage of production emissions is attractive but does not allow for differences in the sequestration rate due to different buildings and environmental conditions (Saade et al., 2022). Perhaps more crucially, it is based on specific assumptions about the way in which cement is produced, and these assumptions are expected to be less applicable in the future as cement producers increasingly integrate SCMs and alternative energy sources into their production processes.

Detailed methods have been presented by Engelsen et al. (2016) which included the degree of carbonation. Resch et al. (2022) described an LCA method for the inclusion of carbonation in the service life alone and pooled these credits with results in units of CO_2-equivalent over a 100-year period. They concluded that 0.06 kg of CO_2 would be sequestered by carbonation for every kg of concrete. Such approaches do not acknowledge different building (e.g. surface to volume ratio) or environmental conditions over time. However, Hawkins et al. (2021) did consider time in the life cycle fluxes of concrete, steel, and timber buildings. By estimating concrete carbonation using a dynamic LCA method, Hawkins et al. (2021) demonstrated the advantages of time consideration, and a need to develop more time-specific tools that allow for a broader range of variables.

The variety of methods spans simplified approaches for LCA practitioners on the one hand to complex stoichiometry for cement researchers on the other hand. A knowledge gap exists here for a best-of-both worlds approach that blends both ease of use and sophistication of analysis.

The gap in research on acknowledging time in concrete carbon footprints is also recognised in a number of review papers. In particular, the review by Arehart et al. (2021) emphasised the different timescales relevant to quantifying carbonation, and the wide variety of approaches used, drawing parallels between the temporal challenges found in biogenic carbon storage and carbonation. The recent review paper on LCA of concrete by Olsson et al. (2024) explored current practices of evaluating cement carbon footprints, such as the system boundary, type

of functional unit, and the inclusion of and rationale for carbonation in LCAs. Similar conclusions were drawn a decade earlier by Wu et al. (2014) who emphasised the importance of use and end of life stages in the carbon footprint of concrete. They pointed out that at the time a significant number of studies considered only cradle-to-gate emissions, thereby missing the life cycle stages in which carbonation provides benefits. The review of LCA methods for carbonation by Thonemann et al. (2022) found little harmonisation, and with the growing attention to maximizing carbonation there is increasing need for a carbonation calculator (Huang, 2019; Zhang et al., 2024). Common among this uptick in carbonation-related review papers is the acknowledgement of temporality, and yet the discourse remains dominated by static LCA methods.

This literature review confirms modelling carbonation in carbon footprints is not yet sufficiently harmonized. Moreover, a number of these studies recognize that the rate of carbonation changes over time and is dependent on environmental factors, alluding to the need for an easy-to-use dynamic LCA approach.

3. Methods

3.1. Goal and scope

The reference unit is 1 solid concrete block with dimensions 400 mm by 200 mm by 200 mm. Its volume is 0.016 m^3 and life one surface (0.08 m^2) is exposed while the remaining five faces are not (see Fig. 1). The calculator developed in this work allows the user selecting different reference units by entering different values for a set of parameters such as volume, surface area, and mass. This specific reference unit has been arbitrarily chosen for the case study developed to show what the calculator can do. Another reference unit with different dimensions could have been selected since the objective is not to analyse the absolute results but to demonstrate how it works. Other parameters such as compressive strength can also be varied, and some of these are explored in a sensitivity analysis which is described in Section 3.5.

The system boundary includes the life cycle modules that relate to carbonation. The production of cement, including high temperature calcination of calcareous rocks to produce clinker, is a source of emissions. On the other hand, the service life and end-of-life are periods of potential sequestration by carbonation.

Static LCA and DLCA results are reported. As such, it is important to define the timing of the stages and their activities. The service life is modelled as 60 years (Resch et al., 2022). Production activities occur in year 0, followed by the service life commencing at year 1 and spanning until year 60. After year 60 the concrete block begins its end-of-life at a landfill. In this model, we assume the concrete block is landfilled with the same exposed surface to volume ratio. This stage is measured for 940 years concluding at a time horizon of 1000 years. This long period is chosen due to the ongoing carbonation in the landfill, allowing our model to capture any possible long term carbonation benefits.

Carbonation is measured during the service life and end-of-life periods. The concrete block undergoes carbonation throughout the service life. Later, at the end-of-life, 100 % of the concrete block is sent to landfill, where it continues to undergo carbonation subject to the new exposure conditions.

3.2. The carbonation LCA calculator

The calculator is developed in Microsoft Excel, making it easily accessible and user-friendly for LCA practitioners. The workbook is made up of an inventory worksheet in which basic data such as volume, surface area, and mass of the concrete block are stored, organised by life cycle stages. Many of these parameters can be changed by the user in the worksheet. These are fed into a second part of life cycle inventory which considers these flows at the year in which they occur.

The life cycle inventory is then used to calculate emissions tables for CO_2, CH_4, and N_2O, and these feed into the DLCA calculator developed by Levasseur et al. (2010), which estimates the instantaneous and cumulative radiative forcing curves over time. These, and the conventional static GWP100 LCA results, are displayed on a dashboard, allowing users to see the years when - and rates at - which carbonation occurs, and the contribution to both the GWP100 and radiative forcing over the course of time. Adjacent to the output dashboard is a panel of dropdown menus which allow the user to select different input parameters, such as those we have used in our scenario analyses. For example, the user can select a different SCM and increase or decrease its percentage content, and immediately see the effect of these changes on the results dashboard.

This easy-to-use static and dynamic LCA calculator demystifies the carbonation black box, revealing when and how much carbon is removed by a concrete block subject to a wide set of real-world conditions. The calculator is freely available in the supplementary information and on the Zenodo repository (https://zenodo.org/records/13830331), where any subsequent updated versions can be accessed. The calculator's variables and data are used in the following subsections.

3.3. Process inventory

Emission factors are sourced from the ecoinvent v3.7 cut-off database (Wernet et al., 2016). The emission factors for cement with 20 % fly ash (class F and C) are modelled using the processes “cement, portland

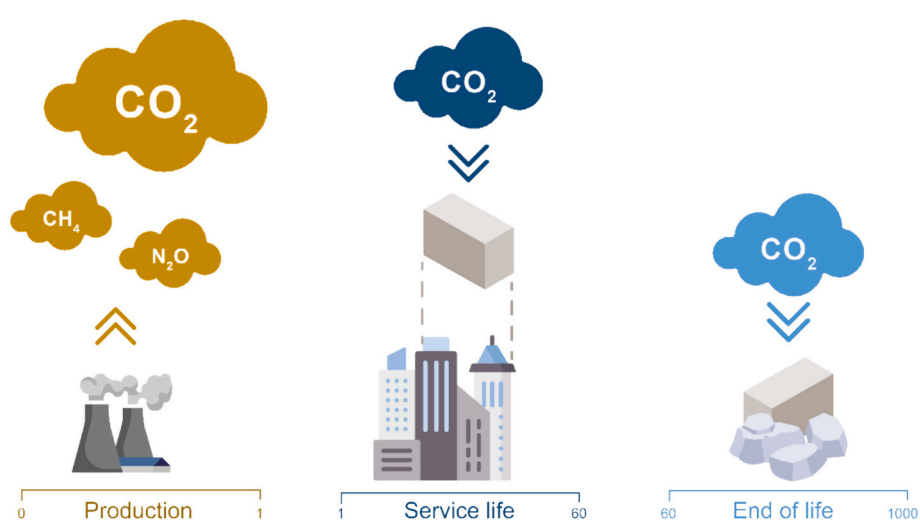

Fig. 1. Stages of carbon flux over time. Production occurs in year 0, service life from year 1 to 60, and the end of life is modelled from 60 to 1000 years.

fly ash cement 6-20%". Cement with 20 % slag is modelled using the process "cement, blast furnace slag 6-20%". These are provided in kg CO_2, CH_4 and N_2O (used in the DLCA), and CO_2-eq (for GWP100) per kg of cement. The production emissions are determined by the proportions and types of SCMs in the cement mixture whose emission profiles are shown in Table 1.

3.4. Carbonation variables

Carbonation depends on the type and constitution of cement in the concrete, the geometry of the concrete, the environmental conditions, and any changes in these variables over time. The following steps are used to estimate the CO_2 flux due to carbonation at each time step. Firstly, the carbonation potential (C_m) is estimated using Eq. (1). To do so, a number of parameters must be known which are determined by the choice of cement type and its SCM content. Ordinary Portland Cement (OPC) Type I is chosen for this case study, which is considered the standard when special properties are not required. However, the analysis can be conducted with Types II, III, IV, V, and white cement.

SCMs can partially substitute OPC, thereby avoiding environmental impacts during the production phase. Common examples of SCMs include fly ash and slag (Kouchaki-Penchah et al., 2024; Souto-Martinez et al., 2017). However, due to differences in chemistry, such as pozzolanic reactivity or aluminum oxide content, SCMs have varying effects on carbon sequestration potential (Von Greve-Dierfeld et al., 2020). The precise impact varies depending on the hydration process initiated by the reaction between cement hydrate, the SCM, and CO_2 species in the pore solution. Literature indicates diverse effects, which are accounted for by adjusting the parameters α and β, as referenced in Souto-Martinez et al. (2017). These are shown in the supplementary information (Table S1). For the baseline scenario, 20 % fly ash (Class F) is used as the SCM. The choice of fly ash as the primary SCM in this scenario is based on its widespread availability, economic feasibility, and established performance in enhancing the durability of concrete (Von Greve-Dierfeld et al., 2020). Additionally, the selection of 20 % substitution is guided by typical industry practices, where this percentage is commonly used to balance mechanical properties and sustainability goals (Souto-Martinez et al., 2017).

The influence of SCM type and classification on carbon sequestration potential is significant due to their different chemical compositions and reactivities. For instance, Class F fly ash has a lower calcium oxide content, which influences its reactivity with CO_2 during the carbonation process, leading to different sequestration outcomes compared to other SCMs like slag (Von Greve-Dierfeld et al., 2020). The choice of a uniform substitution percentage (20 %) across different SCMs, including slag in our sensitivity analysis, allows for a controlled comparison of their effects on carbonation. This approach ensures that the observed differences in carbonation potential can be attributed to the intrinsic properties of the SCMs rather than variations in their dosages. The use of 20 % substitution is also supported by the need to maintain comparable compressive strengths and workability of the concrete mix, which are critical for practical applications (Göswein et al., 2019).

$$C_m = \alpha - \beta \cdot y \tag{1}$$

where:

Table 1
Material production emission factors for static and dynamic calculations in kg per kg cement.

Material	GWP100 (CO_2-eq)	CO_2	CH_4	N_2O
Cement with 20 % fly ash	0.69	0.68	5.25×10^{-4}	4.32×10^{-6}
Cement with 20 % slag	0.71	0.70	5.47×10^{-4}	4.89×10^{-6}

- C_m = carbon sequestration potential, kg CO_2/kg cement
- y = 20 %, percent replacement of OPC by SCM, in decimal
- α = 0.165, carbon sequestration potential coefficients given in the supplementary information (Table S1)
- $\beta = 1.1 \cdot \sigma$, given in the supplementary information (Table S1).

We must calculate the carbonation front's depth from the surface, x. This penetrates more slowly over time, so the depth for each time step (t, year) is relevant (Eq. (2)). As such, this must be calculated for each year t from 0 to 1000. Exposure class determines the parameters k_1 and n (Monteiro et al., 2012). For the baseline scenario, we let the exposure class during both the service life and end of life be XC1 (dry or permanently humid; Table S2). Alternative exposure class assumptions are explored in later scenarios (see Section 3.5 Sensitivity analysis). Together with the environmental CO_2 concentration (c), these factors determine the rate of change of the carbonation front.

$$x = \sqrt{\frac{2 \cdot c \cdot t}{R} \cdot \left(\sqrt{k_0 \cdot k_1 \cdot k_2 \cdot \left(\frac{1}{t}\right)^n} \right)} \tag{2}$$

where:

- c = 0.000814 kg/m^3 environmental CO_2 concentration (Gidden et al., 2019).
- t = exposure time, years
- k_0 = 3 derived from Monteiro et al. (2012)
- k_1 = given in Table S2 (=1 for baseline scenario)
- k_2 = 1 derived from Monteiro et al. (2012)
- $R = 0.0016 \cdot f_c^{3.106}$ kg·year/m^5, carbonation resistance coefficient (=35 for baseline scenario)
- f_c = compressive strength (MPa) (=25 MPa for baseline scenario)
- n = given in Table S2 (=0 for baseline scenario)

The volume of carbonated concrete can be determined at any point on the time continuum (Eq. (3)). The depth of the carbonation front (x) is multiplied by the exposed surface area (SA). Following this, we can calculate the mass of CO_2 stored in concrete (C_S) with constituent cement mass (m, kg/m^3) using Eq. (4).

$$V_c = \begin{cases} SA \bullet x & \text{if } SA \bullet x \le V \\ V & \text{the total volume of concrete, if } SA \bullet x > V \end{cases} \tag{3}$$

$$\phi_c = \text{degree of carbonation}, 0.72.\text{Observed values} \in (0.40, 1)$$

$$C_s = \phi_c C_m \cdot [V_c \cdot m] \tag{4}$$

3.5. Sensitivity analysis

Multiple scenarios are modelled by varying the main parameters from the baseline scenario. These are: the composition of SCMs (S1); the exposure conditions of the concrete during its service life (S2) and its end of life stage (S3); the compressive strength of the concrete (which is linked to the cement content in the concrete) (S4); and the degree of carbonation, Φ (S5). These are defined in Table 2.

The compressive strength determines the composition of the concrete block, and in particular the portion of cementitious material. This influences the production impacts due to the variable upstream processes linked to the material constituents. The baseline scenario with compressive strength 25 MPa contains 381 kg cement per cubic metre of concrete, while the S4 scenarios for 15, 30 and 45 MPa contain 281, 451, and 6421 kg per cubic metre of concrete respectively. These proportions are reproduced from Souto-Martinez et al. (2017), although more recent technology can produce these concrete strengths with lower quantities of cement.

The sensitivity analyses reported in this paper only consider changing one parameter at a time. However, the Calculator described in

Table 2
Scenarios and their definitions with parameter variations. Exposure classes are XC1 (dry or permanently humid), XC2 (humid, rarely dry), XC3 (moderately humid), and XC4 (cyclically humid and dry).

Name	Details	No.
Baseline	Concrete Type I; exposure class is XC1 both during service life and end of life. The compressive strength is 25 MPa with 20 % SCM (fly ash, class F). Degree of carbonation, Φ, is 0.72 (Engelsen et al., 2016)	1
S1	SCM composition changed to fly ash (class C) and slag each at 20 %	2
S2	Exposure class in service life changed to XC2, XC3, XC4	3
S3	Exposure class in end of life changed to XC2, XC3, XC4	3
S4	Compressive strength changed to 15 MPa, 30 MPa, and 45 MPa	3
S5	Degree of carbonation (Φ) changed to a low value 40 % (Fridh and Lagerblad, 2013) and theoretical maximum 100 % (Souto-Martinez et al., 2017).	2

Section 3.2 provides users with the capability to vary multiple or all parameters simultaneously.

4. Results

4.1. Static LCA results

Using the static LCA approach, baseline production impacts contribute 4.26 kg CO_2-eq per reference unit (0.016 m^3 solid concrete block). Around 98 % of this is due to the release of CO_2, most of which can be traced back to calcination for clinker production. Over the thousand-year measurement period, carbonation sequesters 6 % of the CO_2. This is evenly split between uptake during the 60-year service life (0.13 kg CO_2) and the 940-year end-of-life period (0.12 kg CO_2). Together, the carbonation effect reduces the production impacts from 4.26 to 4.02 kg CO_2-eq. These results are shown in Fig. 2.

4.2. Dynamic LCA results

Applying the DLCA approach, the baseline scenario results can be represented as time-differentiated climate impacts. The Global Warming Impact (GWI) of the production stage activities (measured in radiative forcing, W/m^2) decreases over time. This "instantaneous" impact continues, albeit ever diminishing, long after the single pulse emission in year 0 (Fig. 3a). Meanwhile, the GWI of total carbonation (sum of service and end of life stages) begins at year 0 with no contribution, then increasingly offsets the production emissions for approximately the first 200 years. Shortly thereafter, carbonation ceases due to the carbonation front having fully penetrated the depth of the concrete block (Fig. 3b), and the effect of the prior CO_2 removal flat-lines for the remaining eight centuries (c.f. Fig. 3a). The effect of carbonation on CO_2 storage is shown in Fig. 3d, illustrating the rapid uptake in early years, especially during the service life when the carbonation front is nearer the surface and thus more reactive with atmospheric CO_2.

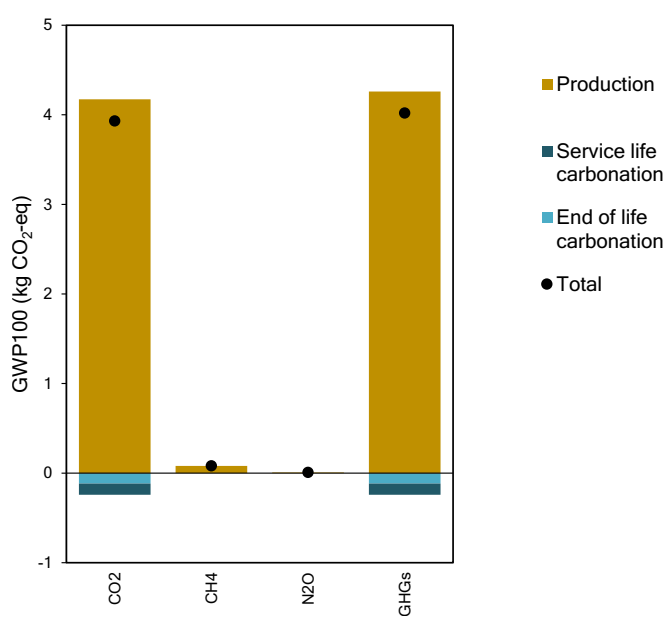

Fig. 2. Baseline Scenario greenhouse gas emissions (GWP100 in kg CO_2-eq).

The cumulative effect of carbonation on net radiative forcing is the real measure of its value. In Fig. 3c we see the cumulative GWI of the production activities (orange line corresponding to orange line in Fig. 3a) and of the carbonation (blue line corresponding to blue line in Fig. 3a). The ratio of these two curves (carbonation to production) is shown in the dashed black line, and this represents the evolution of the partial sequestration payback due to carbonation. Over time the absolute gradient of the orange and blue lines ease, and this multiplicative effect is shown in the sharper decay of the dashed black line.

4.3. Sensitivity analysis

This section investigates the factors affecting carbonation. Some scenarios influence the production emissions, all influence carbonation sequestration, and some influence both. These results are shown in Fig. 4 as differences in GWP100 results, using the static LCA method.

Variations in carbonation can be due to the uptake rate which, if sufficiently slow, can extend beyond the 1000-year time horizon used in this study. The latter are shown by the curves of cumulative carbon storage in Fig. 5.

Starting with S1, carbonation is greatest when class C fly ash is used instead of class F fly ash (in the baseline). To a slightly lesser extent, carbonation is greater with blast furnace slag compared to the baseline class F fly ash. Looking at the secondary y-axis, these higher levels of carbonation cause the ratio of carbonation-related uptake to production emissions to approximately double in the case of class C fly ash (up to 11 %) and increase by around 60 % in the case of slag (up to 9 %). These improvements are marked by the "cross" symbols in Fig. 4 corresponding to the axis on the right.

In S2 and S3 scenarios, minimal variation is seen when varying the exposure conditions during the service life or waste management. This is due to the long time horizon which fully captures the carbonation process, meaning regardless of the type of exposure, which affects the rate of carbonation, the solid concrete block still has time to reach its maximum carbonation potential. Variations are, however, observed between the service life and end-of-life contributions to carbonation under different exposure conditions. Generally, the limiting factor is the exposure during end-of-life due to the longer time period, and this is illustrated by the noticeably lower "cross" for mitigation by carbonation in Fig. 4 for the S3 XC2 result.

With S4 we observe high sensitivity of production impacts due to changing compressive strength. This is expected as compressive strength is directly related to the amount of cement used. Regarding carbonation, owing to the sufficiently long time horizon, carbonation reaches its maximum for the compressive strengths 15 MPa and 30 MPa, and cement is the limiting factor in the carbon removal. It follows that the relationship between production impacts linked to cement and CO_2 removal by carbonation are proportional in these scenarios. This is illustrated by the "cross" symbols on Fig. 4 that remain around 5.7 % mitigation by carbonation for compressive strengths 15 MPa and 30 MPa. The outlier in this regard is the scenario with 45 MPa compressive strength. Due to its higher quantity of cement, the also higher carbonation potential is not fully reached within the 1000-year period, meaning the time horizon truncates the carbon uptake before its maximum level (Fig. 5).

Results for S5 reflect similar trends to S2 and S3. The Degree of

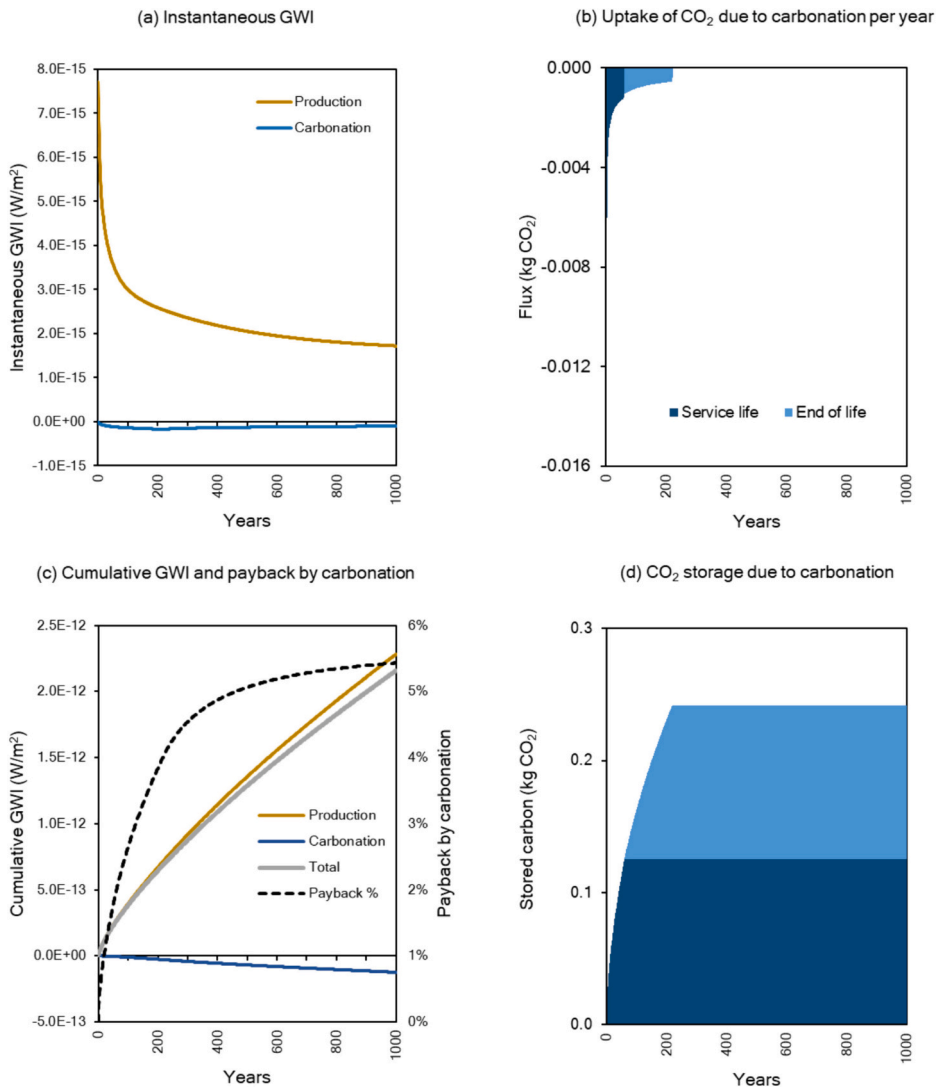

Fig. 3. Baseline DLCA results showing the instantaneous (a) and cumulative (c) GWI over the 1000-year time period and the payback effect of carbonation on it, along with the instantaneous CO_2 uptake (b) and cumulative CO_2 storage (d) by carbonation.

Carbonation variable, Φ, has no influence on production impacts – only on the efficiency of the carbonation reaction. This serves as a scalar limiting the upper bound of carbonation. As such, the total carbon taken up in S5.1, when $\Phi = 0.4$ is 40 % of the carbon taken up in S5.2, when $\Phi = 1$. This affects the ratio of mitigation by carbonation to the same proportions. The rate of carbonation, however, is not influenced by Φ, meaning the carbonation potential reaches its maximum at the same time for both these results (Fig. 5).

5. Discussion

5.1. Static and dynamic LCA

Static LCA has traditionally been used to estimate the carbon footprint of cementitious materials. Our static LCA results of around 6 % carbonation payback are fairly similar to those reported in the literature, such as 7.5 % used by The Concrete Centre (2016) and later Hawkins et al. (2021). The finding that carbonation can mitigate 6 % (and up to 11 %) of the initial production emissions is significant for the concrete industry and broader climate change mitigation strategies. Given the scale of global concrete production, even small percentages of CO_2 sequestration through carbonation could translate into substantial absolute reductions in atmospheric CO_2 concentrations over time. This underscores the potential for integrating carbonation considerations into LCAs of concrete products, promoting the use of materials and designs that maximize and accelerate this natural sequestration process (Scrivener et al., 2018). However, this static relational approach does not account for the progressive and time-dependent nature of carbonation.

In contrast, DLCA provides a more accurate representation of carbonation over time. For example, in the baseline scenario. The maximum carbonation sequestration is not achieved until around 200 years. Over this time, the production emissions persist in the atmosphere and continue to have a warming effect. A static LCA approach fails to capture this difference, instead subtracting the total uptake by carbonation from the production emissions. This is comparable to assuming that full carbonation is achieved at year zero, simultaneous to the emissions from production. This assumption is unrepresentative, as demonstrated by the dashed black line in Fig. 6(a–e), which represents the ratio of carbonation-related CO_2 uptake to production emissions over time for the baseline scenario. The line continues to increase for hundreds of years beyond the pulse production emissions in year 0, showing that the carbonation payback continues well beyond the initial emissions event. The gradual rise in this line reflects the slow and progressive nature of carbonation, emphasizing the need for a time-sensitive approach like DLCA to accurately assess the long-term

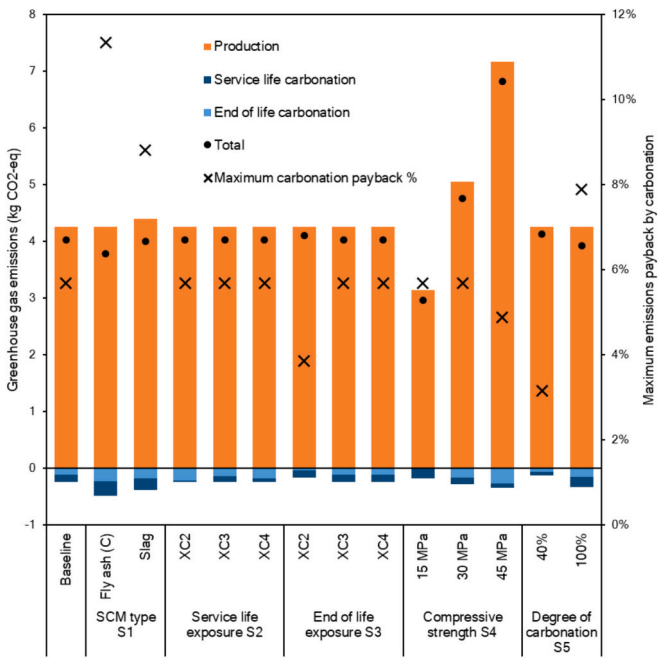

Fig. 4. Static LCA results by scenario and the partial payback effect of carbonation.

climate impacts. The DLCA results show that it takes several decades to reach the first 1 to 2 % payback in terms of radiative forcing, so that carbonation does not really mitigate climate change in the short-term. The static LCA method is unable to show this type of temporal effect. Given the emission reduction targets that we must reach in the next few decades in order to keep global temperature increase below 1.5 or 2 °C (net zero emissions in 2050), differentiating these temporal effects is obviously important.

For the concrete industry, this finding suggests an opportunity to enhance the sustainability of concrete by optimizing mix designs and curing practices to favour carbonation, particularly during the end-of-life phase. Strategies could include designing concrete with higher surface area exposure and controlled environmental conditions that accelerate carbonation without compromising structural integrity (Adu-Amankwah et al., 2017). Additionally, this highlights the need for further research into the development of innovative materials and methods that could amplify this sequestration potential (Gartner and Macphee, 2011). From a climate change accounting perspective, using DLCA to incorporate the carbonation effect into carbon accounting frameworks could encourage a shift towards a more representative understanding of material lifecycles, where the potential for carbon uptake and storage is considered alongside initial emissions over time (Andrew, 2018; Preston and Lehne, 2018).

5.2. Carbonation payback sensitivities

The carbonation payback curves depend most heavily on the choice of Φ, the Degree of Carbonation. This variable is reported in literature between 0.4 (Fridh and Lagerblad, 2013) and 1 (Souto-Martinez et al., 2017), which we used in S5.1 and S5.2 respectively. The carbonation payback is highly sensitive to the choice, illustrated by the wide and widening red trumpet in Fig. 6e. However, the degree of carbonation is not independent of the exposed surface area of the concrete. Future research should aim to converge on more accurate carbonation data, considering the exposed surface area and the extent to which the concrete is broken up. Specifically, experimental studies could focus on long-term monitoring of carbonation in different concrete structures, with varied surface area-to-volume ratios and exposure conditions. For instance, controlled experiments could be designed to measure the carbonation rate in concrete samples with systematically varied degrees of cracking or mechanical break-up, simulating real-world conditions where concrete is subjected to wear and tear over time. Additionally, in-situ carbonation measurements on demolished concrete structures could provide valuable data on how exposure to different environmental conditions (e.g., urban vs. rural settings) affects the carbonation depth and rate. The use of advanced imaging techniques, such as X-ray computed tomography (CT) or microstructural analysis, could also help in understanding the progression of the carbonation front in relation to the porosity and microcracks within the concrete matrix (Thiery, 2005). These efforts would significantly improve the predictive accuracy of carbonation models, leading to better assessments of the long-term carbon sequestration potential of concrete structures. This would lead to a more precise understanding of the long-term climate impact of carbonation, and the opportunity to estimate substitution factors for a suite of concrete products for a range of environmental and life cycle variables as has been achieved for some bio-based products (Cardinal et al., 2024; Gustavsson et al., 2017).

The type of SCM used significantly influences the percentage of production emissions re-sequestered by carbonation, as shown in Fig. 6a. There are two primary mechanisms behind these differences. First, from a production emissions perspective, SCMs are often industrial by-products, and the net emissions associated with their recycling can vary significantly depending on the original process and the resources required for their recovery and processing. This variation in the carbon footprint of SCMs directly affects the overall emissions reduction potential of the concrete mix. Second, the carbon sequestration potential is heavily influenced by the chemical composition and reactivity of the SCMs. For example, Class C fly ash has a higher calcium oxide (CaO) content compared to Class F fly ash, which enhances its reactivity with CO_2 during carbonation. The presence of more calcium hydroxide (CH) in Class C fly ash allows for greater precipitation of calcium carbonate ($CaCO_3$), thus increasing its carbon sequestration potential (Scrivener et al., 2018). In contrast, slag, although lower in CaO content, contains a higher aluminum oxide (Al_2O_3) content. This higher Al_2O_3 concentration facilitates the formation of additional phases such as calcium aluminate hydrates, which also contribute to carbonation, albeit at a slower rate compared to the direct reaction of CH with CO_2 (Alapati, 2019). These differences underscore the need for careful selection of SCMs based on the desired balance between carbon sequestration potential and other performance characteristics.

The differences in carbonation mechanisms among SCMs have important implications for concrete mix design. For instance, using Class C fly ash could be advantageous in applications where rapid early-age carbonation is desirable, such as in eco-friendly concrete products aimed at maximizing carbon uptake. However, the higher reactivity of Class C fly ash with CH might also increase the risk of early-age shrinkage, requiring adjustments in mix design to mitigate cracking risks (Thomas, 2007). On the other hand, slag's slower carbonation rate may be more suitable for applications where long-term durability and gradual carbon sequestration are prioritized. These factors highlight the importance of tailoring concrete mix designs to specific project requirements and environmental conditions. The compressive strength of concrete also plays a critical role in influencing carbonation potential, with the trade-off being that higher compressive strength leads to greater carbon sequestration potential but at a slower rate. This phenomenon can be explained by the chemistry of concrete: higher cement content increases compressive strength by producing more paste and hydration products, including CH. The abundance of CH enhances carbon sequestration potential as it reacts with CO_2 to form $CaCO_3$. However, higher compressive strength is also associated with reduced porosity, which slows down the carbonation process by limiting the diffusion of CO_2 into the concrete (Neville, 1995; Von Greve-Dierfeld et al., 2020).

This trade-off between compressive strength and carbonation rate

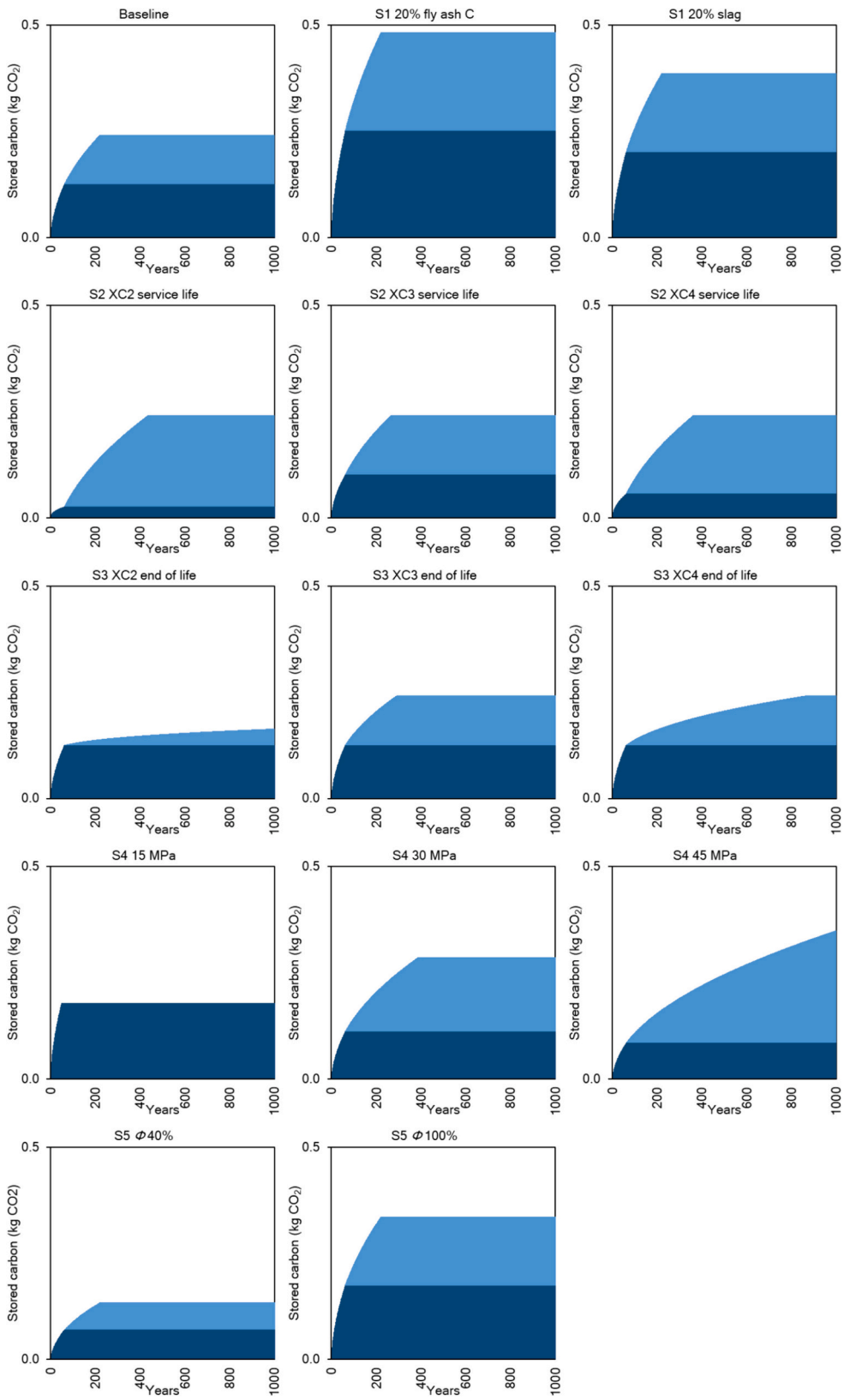

Fig. 5. Cumulative CO_2 storage due to carbonation fluxes in all scenarios. Dark blue indicates CO_2 sequestered during the service life, while light blue indicates CO_2 sequestered during end of life.

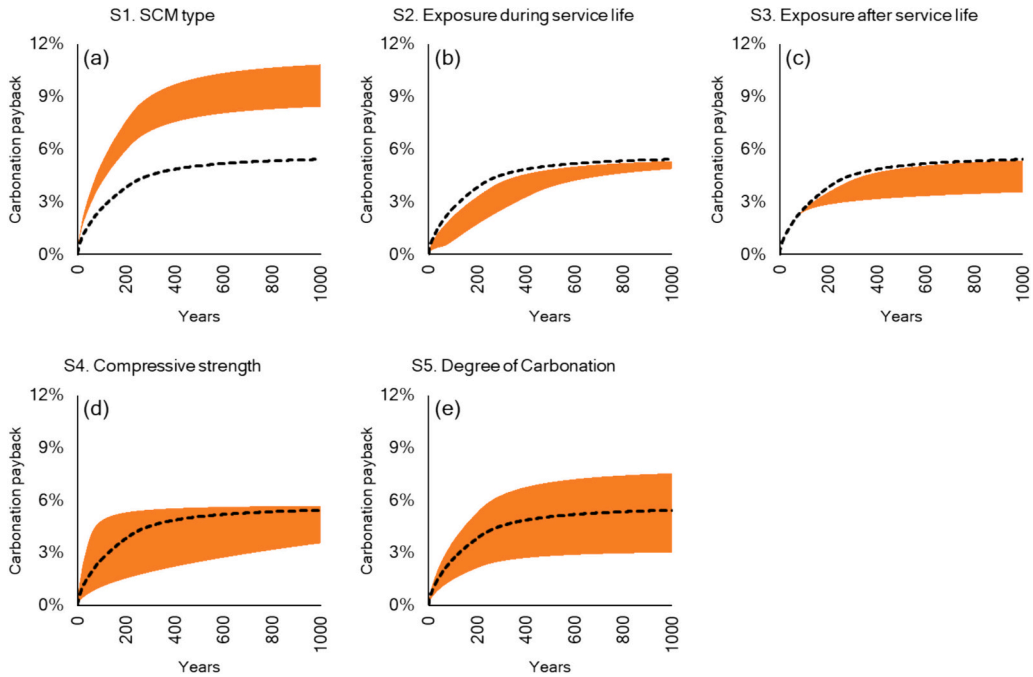

Fig. 6. Sensitivity of radiative forcing (cumulative GWI) mitigated by carbonation as a function of time. The orange band shows the range between upper and lower mitigation in a given scenario. Dashed black line is the Baseline for reference (c.f. Fig. 3c).

has significant practical implications for concrete design. In structures where long-term durability and high strength are critical, such as in high-rise buildings or infrastructure projects, higher compressive strength is often prioritized. However, the slower carbonation rate in these high-strength concretes may delay the realization of environmental benefits from carbon sequestration, potentially reducing their immediate impact on carbon mitigation strategies (Gartner and Macphee, 2011). Conversely, in applications where rapid carbonation is beneficial—such as in precast concrete elements or in the context of early-age carbonation curing—a lower compressive strength concrete may be more appropriate. This would allow for quicker carbon uptake, albeit with trade-offs in structural performance.

Therefore, the choice of compressive strength in concrete mix design should carefully consider the intended application and the desired balance between structural performance, durability and environmental benefits. For instance, lower compressive strengths can achieve their maximum carbonation payback earlier, while also benefiting from lower production-related emissions due to reduced clinker content (see Fig. 6d). Readers are encouraged to experiment with the calculator provided in the supplementary information (and at https://zenodo.org/records/13830331), combining Class C fly ash in lower compressive strength concrete blocks to explore these dynamics further. This yields an earlier and slightly greater carbonation payback (11.44 % over 1000 years).

Lastly, we consider the sensitivity of carbonation payback to environmental changes by varying the exposure conditions during the service life (S2) and end-of-life (S3). Fig. 6b shows that the baseline scenario, XC1, representing 'Dry or permanently humid' conditions, performs the best in terms of magnitude and pace of carbonation payback. This scenario corresponds to real-world conditions where concrete structures are located indoors, away from high humidity areas, or submerged in non-aggressive water, such as in basements or infrastructure with constant water immersion (Monteiro et al., 2012). The performance of XC1 is consistent in both the service life and end-of-life phases, suggesting that concrete used in these relatively stable environments benefits from more effective long-term carbonation.

In contrast, scenarios like XC2, XC3, and XC4 represent more aggressive environments, as defined by the Eurocode standards for exposure classes. XC2 corresponds to environments where concrete is exposed to wetting and drying cycles, such as outdoor structures in urban areas. XC3 refers to structures exposed to moderate humidity, like sheltered outdoor environments, while XC4 covers concrete exposed to frequent contact with water, such as in coastal structures or foundations below groundwater level. These exposure classes influence the carbonation process significantly. For example, in XC2 conditions, the initial carbonation may proceed more rapidly due to the higher moisture levels, but the long-term effectiveness of carbonation payback could be compromised by potential surface degradation (CEN, 2004).

Moreover, the greater sensitivity of the carbonation payback curve under S3 conditions highlights the importance of considering the end-of-life stage, especially in contexts where concrete might be crushed or exposed to different environmental conditions in landfills. The delayed re-sequestration effect, where atmospheric CO_2 persists and continues to have a warming effect over longer periods, emphasizes the critical need for DLCA approaches. DLCA provides a more accurate representation of the timing and impact of carbonation in mitigating the warming effect of cement production emissions, particularly in diverse environmental conditions that concrete may encounter throughout its lifecycle.

The sensitivity analyses show that the DLCA method identifies the impact of changing parameters not only on the total amount of carbon sequestered by concrete through carbonation, but also on the evolution over time of the associated benefits in terms of global warming. Changing certain parameters can lead to slower or faster climate mitigation.

5.3. Limitations and future research

In addition to the intrinsic properties of the concrete mix, environmental conditions play a crucial role in determining the rate and extent of carbonation. The modelling conducted in this study considers various exposure classes (XC1, XC2, XC3, and XC4) as defined by the Eurocode, which represent a range of real-world conditions from dry indoor environments to structures exposed to frequent wetting and drying cycles or constant water immersion. However, it is important to acknowledge that these exposure classes are general categories and that actual climate conditions can vary significantly within each category, influencing the

carbonation process. For instance, in regions with higher humidity and temperature fluctuations, such as tropical or coastal climates, the rate of carbonation may be accelerated due to the increased availability of moisture, which facilitates the diffusion of CO_2 into the concrete matrix (Kosmatka et al., 2002). Conversely, in colder climates, where the concrete may experience freeze-thaw cycles or remain dry for extended periods, carbonation could be slower, potentially limiting the carbon sequestration potential over the same time frame (Dyer, 2014). The variation in CO_2 concentrations in urban versus rural settings also plays a role, with higher urban CO_2 levels potentially enhancing carbonation in structures exposed to such environments (Papadakis et al., 1992).

Temperature is another critical factor influencing the carbonation process in concrete. Higher temperatures generally accelerate the carbonation reaction by increasing the rate at which CO_2diffuses into the concrete matrix and reacts with calcium hydroxide (CH) to form calcium carbonate ($CaCO_3$) (Drouet et al., 2019). This acceleration can lead to more rapid carbon sequestration in warmer climates or during hotter periods. However, the relationship between temperature and carbonation is complex, as very high temperatures can also lead to decreased moisture levels within the concrete, which may slow down the carbonation process due to reduced CO_2 diffusion (Drouet et al., 2019). Moreover, climate change is expected to have a significant impact on the carbonation of concrete. As global temperatures rise, the rate of carbonation in concrete structures may increase, especially in regions that experience more frequent and prolonged heatwaves. This could lead to faster carbon sequestration in some cases, potentially enhancing the long-term environmental benefits of concrete. However, the accompanying changes in humidity, precipitation patterns, and CO_2 concentrations could also alter the carbonation dynamics. For instance, increased CO_2 levels in the atmosphere may enhance carbonation, while shifts in precipitation patterns could either promote or inhibit the carbonation process depending on whether the concrete remains sufficiently moist (Neville, 1995; Stewart et al., 2011).

Given the significant influence of environmental conditions on carbonation, future research should focus on developing a more nuanced understanding of these effects under varying climate scenarios. Long-term experimental studies that simulate different temperature, humidity, and CO_2 concentration profiles across diverse geographic locations would be invaluable. Additionally, incorporating climate change projections into carbonation models could provide insights into how future environmental conditions might alter carbonation rates and carbon sequestration potential. This research could guide the development of adaptive strategies in concrete design, ensuring that concrete structures not only maintain their durability and performance under evolving climate conditions but also maximize their contribution to carbon mitigation efforts. Advanced modelling techniques, coupled with real-world data collection from structures in different climates, could further enhance the predictive accuracy of carbonation processes and support more sustainable concrete practices globally.

The geometric characteristics of the concrete block are also known to play an important role in carbonation rate (Souto-Martinez et al., 2018). For example, a concrete block with two exposed adjacent sides would have a more complex carbonation front, and a steeper uptake curve. Similarly, the concrete block is assumed to be retain the same surface area to volume ratio during the end of life. Our results reflect a conservative time frame for the scenarios that continue undergoing carbonation after 60 years. However, the total potential CO_2 storage would remain the same, so this limitation relates only to the time at which maximum carbonation payback is reached. In fact, including end of life crushing would require a crushing process which is not environmentally benign so would increase the absolute carbon footprint of the concrete block.

6. Conclusions

This research aimed to develop a carbonation impact calculator for carbon footprints of concrete blocks subject to a variety of real-world compositional and environmental conditions. Analysis of the carbonation process over a concrete block's life cycle reveals this carbonation payback effect (the sequestration by carbonation relative to production emissions) plays an important role in reducing the overall production impacts by 6 % in our baseline scenario. This, however, occurs over a very long period of time (around 200 years in the baseline scenario), and is highly sensitive to model parameters. We used the conditional variations to highlight the sensitivity of the results to four main variables (type of SCMs, compressive strength, exposure during and after service life, Degree of Carbonation). In particular, due to the high sensitivity to the Degree of Carbonation variable (Φ), we suggest LCA and carbon footprint studies reporting on the contribution of carbonation include sensitivity analysis until research converges on more accurate carbonation data.

Using a tailored dynamic LCA approach, we showed that the carbonation payback durations occur at different rates over a thousand-year time horizon when the variables are altered. This underscores the need for dynamic LCA to accurately capture the long-term effects. To this end, we provide our user-friendly spreadsheet-based dynamic carbonation LCA calculator for practitioners and academics in the supplementary information (and at https://zenodo.org/records/13830331) of this paper.

The Calculator offers a comprehensive approach to LCA by incorporating dynamic carbonation modelling. Beyond traditional LCA methods, it provides practitioners with actionable insights for reducing concrete's carbon footprint. By accounting for the evolving nature of carbonation and considering the entire product lifecycle, the tool enables informed decision-making, from material selection to construction practices. Additionally, it supports academic research in developing sustainable concrete solutions and educating future professionals.

CRediT authorship contribution statement

Thomas Elliot: Writing – review & editing, Writing – original draft, Visualization, Software, Resources, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Hamed Kouchaki-Penchah:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis. **Victor Brial:** Writing – review & editing, Writing – original draft, Validation. **Annie Levasseur:** Writing – review & editing, Writing – original draft, Validation, Methodology. **Sarah J. McLaren:** Writing – review & editing, Writing – original draft, Resources, Project administration, Methodology, Funding acquisition.

Declaration of competing interest

The authors have no competing interests to declare.

Data availability

The data use in this study and that support the findings of this study are provided in the cited literature and the supplementary information.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.spc.2024.09.009.

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