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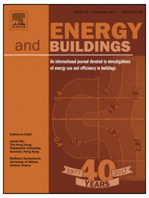

Cradle-to-gate CO_2e emissions vs. *in situ* CO_2 sequestration of structural concrete elements

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ABSTRACT

Life cycle assessment (LCA) is an emerging methodological tool used in building design and construction to quantify the environmental impacts of materials, components, and whole buildings. While calculation of embodied cradle-to-gate greenhouse gas (GHG) emissions is commonplace for ordinary portland cement (OPC) structural concrete elements in building-related LCAs, the carbon dioxide (CO_2) sequestered by *in situ* carbonation of exposed OPC concrete is often neglected because the quantity of sequesterable CO_2 is assumed trivial compared to the initial GHG emissions (kg CO_2e) associated with their manufacture. Using a screening cradle-to-gate LCA and a previously developed and validated CO_2 sequestration model for OPC concrete, this paper quantifies and compares estimates of the initial CO_2e emissions to the CO_2 sequestration potential of several OPC concrete elements at both finite (25 years) and infinite time intervals. The results demonstrate that, depending on cement type, compressive strength, structural geometry, and time, approximately 19%—a non-trivial sum—of initial CO_2e emissions could be recoverable *via* CO_2 sequestration for the concrete elements considered herein. Notably, however, concrete elements that sequester the most CO_2 do not always result in the lowest net CO_2e emissions.

1. Introduction

Ordinary portland cement (OPC) concrete is the most commonly utilized construction material in the world [1]. Consequently, its widespread production, use, and disposal have global environmental consequences. For example, the manufacture of cement alone is responsible for 5–8% of total global carbon dioxide (CO_2) emissions [2]. Approximately 50% of the cradle-to-gate CO_2 emissions associated with cement manufacture is attributable to calcination of calcium carbonate ($CaCO_3$), a predominant mineral in limestone and the primary raw material used in the manufacture of OPC [3]. Calcination occurs by heating $CaCO_3$ in a kiln to temperatures in excess of 1000 °C, resulting in the formation of calcium oxide (CaO) and gaseous CO_2. In addition to chemical calcination, approximately 40% of cradle-to-gate greenhouse gas emissions are attributable to indirect emissions from the combustion of fossil fuels used to heat the kiln to temperatures required for calcination and cement clinkering reactions to occur. Electricity used to power additional equipment and transportation account for the remaining 10% [4].

Experimental studies over the last few decades have identified strategies and best practices to reduce the GHG emissions from OPC concrete manufacture. Such strategies include utilization of recycled aggregates [5,6] in lieu of virgin aggregate and inert mineral fillers [7], like ground limestone, to partially replace cement. Many studies have also investigated the positive environmental benefits of partially replacing cement with supplementary cementitious materials (SCMs) (i.e., fly ash, slag, silica fume, metakaolin) in OPC concrete [8]. In addition to reducing the embodied carbon of straight OPC concrete mixtures, SCMs are silica-rich, which also enhance the strength and durability of cementitious binders [9].

Several tools and methodologies, such as whole-building life cycle assessment (LCA), have emerged to help architects and engineers quantify and report the environmental impacts of building materials and structures. Examples of LCA implementation case studies related to cement and concrete can be found in the literature [10–12]. In the majority of studies that report embodied carbon (i.e., total GHG emissions expressed in kg CO_2e), the *in situ* CO_2 sequestration potential of cement and concrete *via* carbonation is neglected in the environmental accounting. Carbonation is a well-known chemical reaction between hydrated cement paste and atmospheric CO_2. Omissions of sequestered CO_2 is due, in part, to (1) widespread belief that total *in situ* sequesterable CO_2 is trivial compared to initial CO_2e emissions and (2) a lack of familiarity

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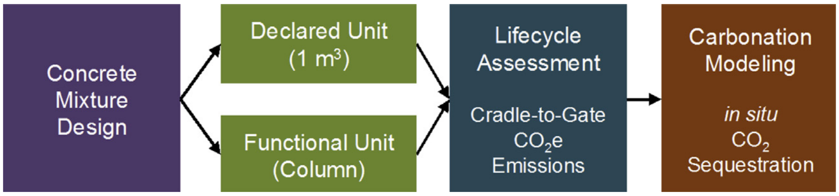

Fig. 1. Overview of modeling methodology.

with simple models to predict sequestered CO_2 over the lifespan of OPC concrete elements. Other barriers include potential uncertainty in the calculations and a lack of standards or methodologies relating to modeling CO_2 uptake.

Only recently have researchers used carbonation data or models to calculate estimates of CO_2 sequestered by exposed concrete elements. Even fewer studies have compared estimates of total sequestered CO_2 with total initial CO_2e emissions [13–18]. While some studies have concluded that the long-term CO_2 sequestration is negligible [18], others claim that 15.5–17% [17], 13–48% [14] or 20–47% [16] of the initial CO_2e emissions of concrete elements can be recovered *via in situ* carbonation of exposed concrete elements. Evidenced by the range of sequestration-to-emissions estimates found in current literature, the cement and concrete community has not yet reached consensus on the triviality or non-triviality of sequesterable CO_2.

Leveraging a simple, yet robust, mathematical model previously developed and validated by the authors [19] that quantifies the total CO_2 sequestration potential of exposed OPC concrete, the goal of this work was to employ LCA to quantify and compare the initial cradle-to-gate CO_2e emissions with total sequesterable CO_2 of exposed OPC concrete elements. A structural column designed using varying OPC concrete mixtures was used as a functional unit to illustrate the effect of geometry (namely surface-area-to-volume), cement type, cement quantity, compressive strength, SCM type, and SCM content on the ratio of initial cradle-to-gate CO_2e emissions versus *in situ* CO_2 sequestration for both finite (25 years) and infinite exposure scenarios.

2. Methodology

Fig. 1 outlines the methodology used in the study. To quantify the initial cradle-to-gate CO_2e emissions and CO_2 sequestration potential of OPC concrete columns, first, 30 candidate OPC concrete mixtures were designed according to the absolute volume method (AVM) specified by the Portland Cement Association (PCA) [20] to estimate total cement content for concretes of different compressive strength. Secondly, a comparative life cycle assessment (LCA) was conducted in accordance with the ISO 14040/14044 [21,22] standards to calculate initial embodied carbon (kg CO_2e), as defined by the Intergovernmental Panel on Climate Change, of structural concrete columns that were designed using the 30 aforementioned concrete mixtures. The LCA employed a consistent declared and functional unit, namely (1) a constant volume of 1 m^3 (declared) and (2) a concrete column with a fixed height of 3 m and a varying cross-sectional geometry required to resist a compressive load of 3000 kN (functional). Finally, the recently validated CO_2 sequestration model [19] was used to calculate total *in situ* sequesterable CO_2 (kg CO_2) of the concrete column elements after (a) finite exposure of 25 years in a 300 ppm CO_2 environment and (b) infinite exposure, assuming full carbonation upon post-use deconstruction, crushing, and subsequent exposure to atmospheric CO_2. Explicit details of the modeling methodology are presented in the following sections.

2.1. Concrete mixture design

Table 1 shows the mixture proportions in kg/m^3 for concrete mixtures varying compressive strengths, namely 15 MPa, 30 MPa, and 45 MPa, designed according to the PCA AVM [20]. For each of these mixtures, additional sample mixtures that incorporated 10% and 20% cement replacement (by weight) with SCMs were also considered to investigate the effect of SCMs on initial carbon emissions and CO_2 sequestration potential. Two of the most common SCMs, fly ash and slag, were included in the analysis for mixtures with cement replacement, thus leading to a total of 15 concrete mixtures—five different concrete mixtures for each of the three compressive strength formulations.

Two types of cement were included in the analysis for all concrete mixtures, Type I cement, one of the most common cement types utilized in construction, and White cement, an architectural cement used most commonly for exposed-surface elements. Theoretical amounts of sequesterable CO_2 for different cement types vary due to their mineral composition [19]. While previous work has shown that White cement exhibits a higher CO_2 sequestration potential than Type I cement due to its lower tetracalcium aluminoferrite (C_4AF) content [19], the embodied carbon of White cement is higher than Type I cement [23]. Therefore, this study necessitated inclusion of Type I and White cement concretes in order to state, with more certainty, a comparison of their net total CO_2e emissions. A total of 30 concrete mixture design formulations were generated.

2.2. Life cycle assessment (LCA) methodology

The LCA in this study was conducted in accordance with ISO 14040/14044 standards and includes lifecycle stages A1–A3 and B1, as specified by EN 15804. Definition of the goal and scope, life cycle inventory (LCI) analysis, critical data review, the CO_2 sequestration model, and limitations of the LCA are discussed in the following sections.

2.2.1. Goal and scope definition

This LCA is a screening LCA that quantifies the net carbon emissions of 15 MPa, 30 MPa and 45 MPa OPC concrete elements that contain either Type 1 or White cement and either 0%, 10%, or 20% replacement of fly ash or slag. This LCA will report values for global warming potential (kg CO_2e), including estimates of CO_2 sequestration assuming both a finite (25 year) and an infinite lifetime for all concrete mixtures listed in Table 1. Results of this LCA will be used to quantitatively compare the use-phase sequesterable CO_2 with initial cradle-to-gate CO_2e emissions to estimate the percent of initial carbon dioxide emissions recoverable *via* CO_2 sequestration.

The system boundaries for initial carbon emissions are cradle-to-gate and include product stages A1–A3. In addition, use phase (B1) is included to quantify the CO_2 sequestered in two time scales – 25 years of exposure and infinite exposure. The use phase (B1) is included in this analysis, because the sequestered carbon is dependent on carbonation depth, which is a function of exposure time

Table 1
Mixture proportions (kg/m^3) for the three concrete design compressive strengths with variations for 0%, 10% and 20% replacement with fly ash or slag.

SCM Content	Compressive strength								
	15 MPa			30 MPa			45 MPa		
	0%	10%	20%	0%	10%	20%	0%	10%	20%
Cement	281	253	225	451	406	361	641	577	513
Water	102	102	102	110	110	110	118	118	118
Coarse aggregate	1013	1013	1013	1013	1013	1013	1013	1013	1013
Fine aggregate	866	866	866	715	715	715	547	547	547
SCM	0	28	56	0	45	90	0	64	128

Table 2
Structural geometry of column functional units.

Column geometry	Compressive strength					
	15 MPa		30 MPa		45 MPa	
	Square	Cross	Square	Cross	Square	Cross
a (m)	0.447	0.128	0.316	0.0905	0.258	0.0739
b (m)	0.447	0.359	0.316	0.2535	0.258	0.207
Cross-Sectional area (m^2)	0.20	0.20	0.10	0.10	0.07	0.07
Surface area (m^2)	5.36	10.14	3.79	7.17	3.10	5.85
Volume (m^3)	0.60	0.60	0.30	0.30	0.20	0.20

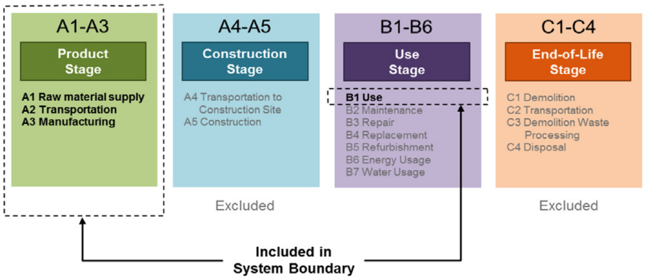

Fig. 2. Lifecycle stages A1–A3 (cradle-to-gate) and B1 (sequestration) (**bold**) included in the system boundary. Per the goal and scope of this LCA, all other stages were excluded.

(see Section 2.2.4). For the infinite exposure condition, total volume of carbonated concrete is assumed equal to total volume of the concrete element. Transportation to the construction site (A4) and construction (A5) are not included. Fig. 2 summarizes the system boundary and lifecycle stages included in the screening LCA.

In addition to analyzing initial cradle-to-gate CO_2e emissions for a declared unit of 1 m^3, the functional unit for this LCA is a three-meter-tall concrete column designed using the candidate concrete mixtures listed in Table 1 to resist a 3000 kN force in simple axial compression (Fig. 3). The required area for each column was computed by dividing the axial compression load by the compressive strength of the concrete. The resulting dimensions for each geometric cross-section (either square- or cross-shaped, according to Fig. 3) were computed and are listed in Table 1. Two column geometries (square- and cross-shaped) were studied to elucidate the effect of surface area-to-volume (SA/V) ratio on total CO_2 sequestration and net CO_2e emissions.

Fig. 3 illustrates how higher compressive strengths result in reduced cross-sectional area for each compressive strength mixture and, thus, less concrete volume. The dimensions, cross-sectional areas, and total volumes of the case study columns (shown in Table 2) were held constant for each compressive strength mixture design. All cross-shaped columns have equal proportionality, namely the ratio between the long and short side of each arm of the cross is held constant at 2.8. While all columns are assumed to be located in a mild CO_2 concentration environment (300 ppm), the concentration only affects CO_2 sequestration potential for short-term (finite) exposure, whereas it does not affect calculations of sequestered CO_2 for the infinite exposure condition [19].

2.2.2. Lifecycle inventory (LCI) data

For consistency, the LCI data were primary data taken from the University of Bath's Inventory of Carbon and Energy (ICE) database, Version 2.0 [24]. The ICE database is an open-access database developed to catalog cradle-to-gate embodied carbon and embodied energy coefficients of materials common in the construction industry. The database contains data regarding the emissions of all GHGs normalized by their relative global warming effect in the atmosphere with respect to CO_2 over a 100-year period. The most common and most significant GHGs emitted in the construction industry are CO_2 (CO_2e of 1.0), methane (CO_2e of 25) and Nitrous Oxide (CO_2e of 298) [25]. The total quantity (i.e., total warming effect of all GHG emissions) is measured and reported in total kg CO_2e.

These coefficients come from peer-reviewed literature reporting both mean values and standard deviations that account for the variance in production methods of materials. While the ICE database is UK-based, it is deemed appropriate for the scope of this screening LCA study because of its consistent methodology for data collection and thoroughness. In instances where embodied carbon coefficients were not available in the ICE Database, primary data were obtained using publicly available environmental product declarations (EPDs) and published literature [23,26,27]. Regardless of source, the data were consistent between analyses. For fly ash and slag, energy from reprocessing is included in the carbon coefficients. Table 3 shows the LCI of embodied carbon coefficients used for this LCA, as well as the comments on the reliability, origin, and references of these data.

In sum, the input parameters for this LCA (see Table 3) include type and quantity of cement, SCMs, water, course aggregate, and fine aggregate to create both the declared unit (1 m^3) and functional unit (a column designed to resist 3000 N). The mass quantities for each unit used in the concrete mix designs are described in Table 1. The output parameters for this LCA are kg CO_2e, which includes both initial GHG emissions in the product stage and recovered emissions via carbonation during the use stage.

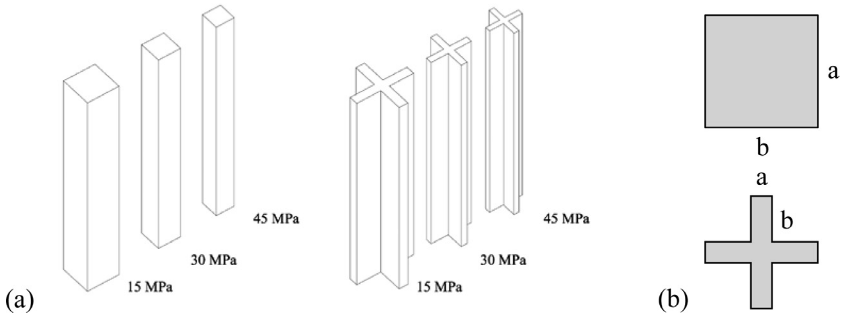

Fig. 3. (a) Square- and cross-shaped column functional units designed with a fixed height (3 m) and compressive load (3000 kN) using concrete mixtures of varying compressive strengths and (b) two representative cross sections. See Table 2 for dimensional details.

Table 3
Embodied carbon coefficients of concrete constituent materials.

Component	kg CO_2e / kg	Reliability	Comments and References
Water	0.0003	High	LCA from US research in accordance with ISO 14044 Standard [27]
Type I cement	0.95	High	BATH ICE Database [24]
White cement	1.07	High	EPD [23]
Fly ash	0.00392	Medium	Data from Danish EPD [24] using by-product allocation
Slag	0.083	Medium	BATH ICE Database [24]
Coarse aggregate	0.0052	Medium	Specific data were difficult to obtain; BATH ICE Database [24]
Fine aggregate	0.0051	Medium	Specific data were difficult to obtain; BATH ICE Database [24]

2.2.3. CO_2 sequestration model

In the use-phase (B1), a mathematical model that predicts the CO_2 sequestration potential of exposed OPC concrete elements was used to quantify the total amount of CO_2 the concrete mixtures would sequester for each of the column geometries and concrete mixtures [19]:

$$C_s = \phi_c \, C_m \cdot [V_c \cdot m] \tag{1}$$

where C_s is the total mass of sequesterable CO_2 (kg CO_2), ϕ_c is the carbonation degree, m is the total mass of cement per unit volume of concrete (kg/m^3) obtained from the concrete mixture proportions (see Table 1), and the quantity in brackets is equal to the total mass of carbonated cement paste. While a theoretical 100% degree of carbonation, $\phi_c = 1.0$, is assumed herein, actual degrees of carbonation ranging from 0.40 to 0.72 have been experimentally obtained by previous researchers [28–32]. The CO_2 sequestration potential, C_m, is calculated according to:

$$C_m = \alpha - \beta \cdot y \tag{2}$$

where C_m is defined as the total mass percentage of sequesterable CO_2 per kg of carbonated cement paste (kg CO_2/kg cement) in the concrete and y is the percent replacement (by mass of cement) by SCMs in decimal form. The α parameter is based on stoichiometric relationships between cement hydration and carbonation reaction chemistries and accounts for variation in cement type. The β parameter is based on the silica content in SCMs and pozzolanic reaction chemistry and accounts for the specific type of SCM [19]. Total carbonated volume is calculated according to:

$$V_c = SA \cdot x \tag{3}$$

where the total carbonation depth, x, is multiplied by the total surface area, SA, of exposed concrete members (see Table 2). An empirical relationship for carbonation depth proposed by Monteiro et al. [33] was used herein:

$$x = \sqrt{\left(\frac{2 \cdot c \cdot t}{R}\right)} \cdot \left[\sqrt{k_0 k_1 k_2}\left(\frac{1}{t}\right)^n\right] \tag{4}$$

where c is the environmental CO_2 concentration (kg/m^3) (Note: 1 kg/m^3 CO_2 = 516 ppb), t is exposure time (years), k_0 is equal to 3.0, k_2 is equal to 1.0 for standard curing, and R is the carbonation resistance coefficient (kg year/m^5) that is calculated for Type I cement according to:

$$R = 0.0016 \cdot f_c^{3.106} \tag{5}$$

and for White cement according to:

$$R = 0.0018 \cdot f_c^{2.862} \tag{6}$$

where f_c is the compressive strength (MPa). The factors k_1 and n, shown in Table 3, are dependent upon exposure classifications as outlined below in Table 4.

The exposure classification used in this analysis was a permanently dry condition ($k_1 = k_2 = 1.0$, $k_3 = 3.0$, $n = 0$) and a CO_2 concentration of 300 ppm (5.81×10^{-4} kg/m^3). See [33,19] for complete mathematical details. All input parameters used in the CO_2 sequestration model are listed in Table 4.

2.2.4. Limitations of the study

There are several limitations in this study that building designers and decision-makers should take into consideration, as some metrics were considered outside of the scope boundary, which affect the findings, recommendations, and results presented herein:

- Variations in compressive strength due to SCM replacement or cement type (Type I vs. White) were not considered and, therefore, an equal compressive strength compared to straight OPC mixtures was assumed for Type I and White cement concretes and SCM-containing mixtures. Given similar calcium silicate mineral contents of Type I and White cement [19], differences in compressive strength were considered negligible.
- Similarly, while SCM additions are well known to provide strength enhancements to OPC concrete, these effects were not considered, since no established, agreed-upon relationships exist to predict such improvements. While a minor simplification in this study, this omission was considered a conservative assumption, since initial carbon emissions of functionally equivalent columns were not underestimated.
- Column cross-sectional areas are calculated for each compressive strength mixture by considering only a vertical load. All serviceability (i.e., deflection) or stability considerations, as well

Table 4
CO_2 sequestration modeling parameters [19].

Parameter	Symbol	Variables	Value	Unit
Cement type	α	Type I	0.165	kg CO_2e / kg cement
		White	0.203	kg CO_2e / kg cement
SCM type	β	Fly ash	0.55	kg CO_2e / kg SCM
		Slag	0.28	kg CO_2e / kg SCM
SCM content	y	10% replacement	0.10	–
		20% replacement	0.20	–
Compressive strength	f_c	Low	15	MPa
		Medium	30	MPa
		High	45	MPa
CO_2 Concentration	c	Low	0.0005831 (300)	kg/m^3 (ppm)

as type and amount of reinforcement, were excluded from the LCA.

- Lifecycle phases outside of A1–A3 and B1, including construction and transportation were not considered within the scope of this study. Including these lifecycle phases for a specific building project would be essential for calculating total cradle-through-construction or cradle-to-grave LCAs.
- The carbonation model employed herein only considers the carbonation potential of portlandite (calcium hydroxide) and does not include potential carbonation of calcium silicates, a limiting but conservative assumption of the model thoroughly discussed in [19]. Furthermore, only one exposure condition ($CO_2 = 300$ ppm) was considered. Higher CO_2 exposures would exacerbate uptake at early ages, affecting results at finite time scale.
- The degree of carbonation used in this study was 1.0, corresponding to the theoretical maximum of potential CH carbonation. While other studies have shown that the actual measured degree of CH carbonation is highly variable and typically less than the theoretical maximum [28–32], the results should be analyzed with an understanding that the theoretical maximum carbonation of only CH is included in the analysis and that true carbonation may be more or less, based on the degree of CH carbonation and the potential (and likely) carbonation of other calcium silicate phases (i.e., calcium-silicate-hydrate).
- Durability assessments were also considered beyond the scope of this study. A negative impact of concrete carbonation is that CO_2, which initially dissolves in water to form carbonic acid (H_2CO_3), can subsequently react with alkalis in the pore solution (e.g., Ca^{2+}) and deplete hydroxide ions (OH^-) from the pore solution chemistry, effectively lowering the pH of the pore solution from approximately 12.5–9.0. This reduction can destabilize the protective oxide layer that forms on the surface of steel reinforcement. Passivation can lead to corrosion in the presence of oxygen and water. Therefore, adequate cover depth is required to protect steel reinforcement, especially in severe exposure conditions. Alternative reinforcement strategies, such as the use of epoxy-coated rebar or glass fiber-reinforced polymer (GFRP) rebar, can increase the service-life of reinforced OPC concrete that may be prone to chemical deterioration *via* carbonation. Therefore, enhanced CO_2 sequestration strategies for concrete (e.g., maximizing surface-area-to-volume ratio) should be thoroughly considered in the context of this potentially negative tradeoff in long-term durability.

3. Results and discussion

Results are first presented for initial cradle-to-gate emissions for both the declared (1 m^3) and functional (column) units. The *in situ* CO_2 sequestered by each declared and functional unit after a finite (25 years) period and infinite period of time. The total net CO_2e emissions are then presented, along with estimations of initial CO_2 emissions that are recoverable through *in situ* carbonation.

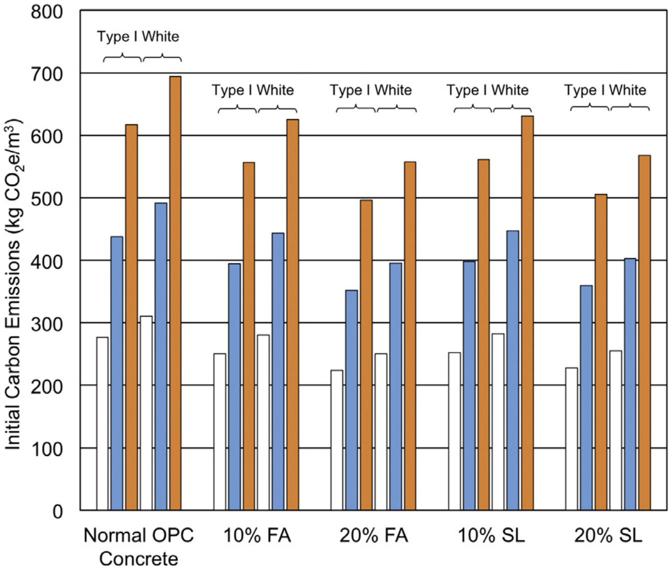

Fig. 4. Estimated embodied carbon (kg CO_2e) for a constant volume (1 m^3) of Type I and White cement 15 MPa (○), 30 MPa (●) and 45 MPa (●) concrete.

3.1. Initial carbon emissions

3.1.1. Declared unit: 1 m^3

The initial cradle-to-gate CO_2e emissions of a unit volume (1 m^3) of the Type I and White cement concrete mixtures investigated herein are presented in Fig. 4. As expected, higher compressive strength concretes resulted in higher per-unit-volume embodied carbon, due to increased cement contents required to achieve high compressive strengths (see Table 1). For example, a cubic meter of a Type I 45 MPa concrete with 0% SCMs would emit approximately 128% more CO_2e than a Type I 15 MPa concrete. As anticipated, this trend is consistent between both Type I and White cement types.

Expectedly, the data also illustrate that addition of SCMs result in a lower per-unit-volume embodied carbon, due to lower embodied carbon coefficients of SCMs compared to that of OPC (see Table 3). When SCMs are utilized in substitution of cement, total unit-volume impacts are reduced by the same percentage of the cement replacement percentage. For instance, when 10% of fly ash is used, emissions from a 15 MPa concrete cubic meter are reduced from 277 kg CO_2e to 250 kg CO_2e, a 9.6% reduction. Unit volumes that contain slag exhibit slightly higher carbon emissions than mixtures that utilize fly ash. For example, a 30 MPa mixture with Type I cement concrete with a 20% cement replacement with fly ash releases 2% less carbon on a per-unit-volume basis than an

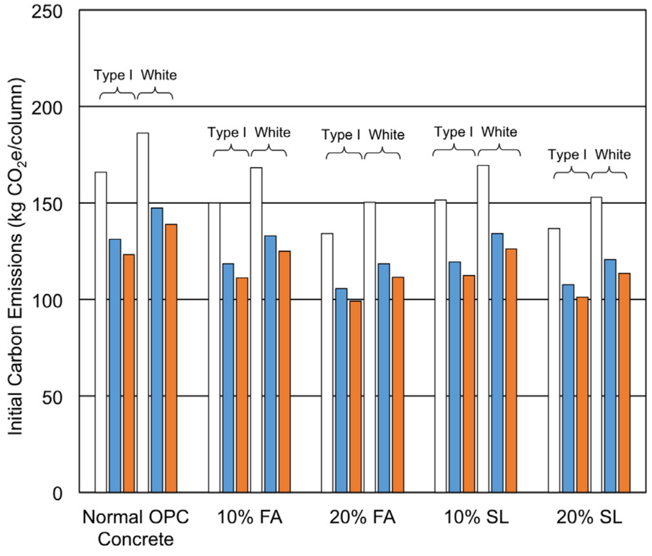

Fig. 5. Initial embodied carbon (kg CO_2e) for the functionally equivalent Type I or White cement 15 MPa (○), 30 MPa (●) and 45 MPa (●) concrete columns.

identical mixture using 20% slag. This result, attributable to the initially higher embodied carbon coefficient of slag compared to that fly ash (see Table 3), can be observed in both cement types and for any compressive strength.

As shown by Kosmata et al. [20], the initial carbon emissions of 1 kg of White cement concrete is approximately 13% higher than Type I cement. Given that Type I cement releases 0.95 kg CO_2e/kg cement and White cement releases 1.07 kg CO_2e/kg cement (see Table 3) [23], the observed increase in unit-volume emissions for White cement concretes is consistent for all compressive strengths and all additions of SCMs, given that the only difference between any two mixtures of identical strength and SCM content is the type of cement. The increase can be explained by the difference in manufacturing of White cement, which limits the total amount of iron and manganese oxides to ensure a white, instead of grey-brown, color [20].

The high- and low-carbon-emitting mixtures on a per-volume basis are evident in Fig. 4. Maximum initial embodied carbon observed for 1 m^3 of concrete was 694 kg CO_2e for a high compressive strength (45 MPa) White cement concrete mixture with no SCM replacement. Contrastingly, a low compressive strength (15 MPa) Type I cement concrete with 20% fly ash replacement (228 kg CO_2e) exhibited the minimum initial embodied carbon for 1 m^3. While initial cradle-to-gate carbon emissions per unit volume can be more than triple for different structural concretes designs and proportions, finite conclusions regarding total life cycle carbon emissions for structural concrete elements are limited without (1) considering structural functional or (2) including estimates, if non-trivial, of sequesterable CO_2.

3.1.2. Functional unit: concrete column

The cradle-to-gate embodied carbon emissions of concrete columns designed using all Type I and White cement concrete mixtures are presented in Fig. 5. The results illustrate that, contrary to trends for unit volume, higher compressive strength concretes correspond to decreased initial embodied carbon, which is attributable to lower volumes of high-strength concrete required to perform the same structural function as low-strength concrete. For example, a Type I 15 MPa concrete column ($V = 0.6$ m^3) with no SCMs emits 24.5% more CO_2e during manufacture than a Type I 45 MPa concrete column ($V = 0.2$ m^3). Similar to the declared unit (1 m^3) results, however, initial emissions of the White cement concrete columns are consistently higher than Type I cement concrete columns of identical compressive strength for reasons previously explicated. In addition, the data in Fig. 5 further illustrate the anticipated reductions in initial embodied carbon of the functionally equivalent concrete columns when using SCMs. For instance, emissions from a Type I 15 MPa concrete column is reduced from 166 kg CO_2e to 150 kg CO_2e or 151 kg CO_2e for a 10% replacement with fly ash or slag, respectively.

In analyzing the functionally equivalent columns, the minimum initial cradle-to-gate embodied energy corresponds to a Type I 45 MPa concrete column with 20% fly ash replacement, while the maximum corresponds to a White 15 MPa column with no SCM replacement. These findings quantitatively demonstrate that, depending on material choice (i.e., concrete mixture design), the initial embodied carbon emissions for the functionally equivalent structural element analyzed herein can range from 99.2 to 186 kgCO_2e. Therefore, similar to findings by other studies that investigated initial carbon emissions of structural concrete [34–36], non-trivial reductions in initial cradle-to-gate carbon emissions can be achieved *via* judicious materials selection decisions during the design development phase.

3.2. CO_2 sequestration

3.2.1. Finite exposure (25 years) of functionally equivalent columns

In contrast to initial CO_2e emissions, Fig. 6 presents the quantitative predictions of *in situ* CO_2 sequestration after 25 years of exposure using the mathematical model presented in Section 2 for the square- and cross-shaped columns. Given that all columns designed with identical compressive strengths have equivalent volumes, the data in Fig. 6 elucidate the enhancing effects of increasing surface area on total sequesterable CO_2 after a finite exposure period for columns of equal compressive strength. For the cross-shaped geometries, for example, the surface area of the concrete column increases, and, consequently, the total carbonated volume is higher compared to the square-column geometry. This effect is observable in Fig. 6, in which a Type I 45 MPa cross-shaped column sequesters 90% more CO_2 than a Type I 45 MPa square-shaped column after 25 years of exposure.

The concrete columns designed with a strength of 15 MPa show identical carbonation potential regardless of geometry (square vs. cross). This result is due to both columns fully carbonating after 25 years. Unlike higher-strength columns, 15 MPa columns have a higher diffusion coefficient resulting in faster carbonation depth. Thus, for these low-strength concretes, while the higher surface area geometry likely enhanced early-age sequestration, after 25 years, both columns have reached full carbonation.

An increase in total CO_2 sequestration after finite times of exposure is observed with these low-strength concrete mixtures, as well as mixtures that contain White cement and mixtures that do not contain SCMs in comparison to all other mixtures. Lower compressive strengths increase the total CO_2 sequestered at early ages due to more porous cement pastes and higher overall gas permeability. Previous research has also shown that the use of low C_4AF cements, like White cement, enhances the CO_2 sequestration potential of exposed OPC concrete. As discussed, the color of White cement is preserved by limiting amounts of metals (e.g., iron) in raw material precursors and, consequently, low C_4AF content in the cement clinker [20]. During cement hydration, C_4AF consumes available calcium hydroxide (CH), which reduces the CO_2 sequestration potential of cements that contain it. In comparison to a Type I 30 MPa concrete column, for example, a White cement 30 MPa column with 0% SCMs can sequester 73% more CO_2. Similarly, as evident in Fig. 6, the addition of siliceous SCMs causes a reduction in the CO_2 sequestration potential of hydrated portland cement paste by reacting with CH to produce calcium-silicate-

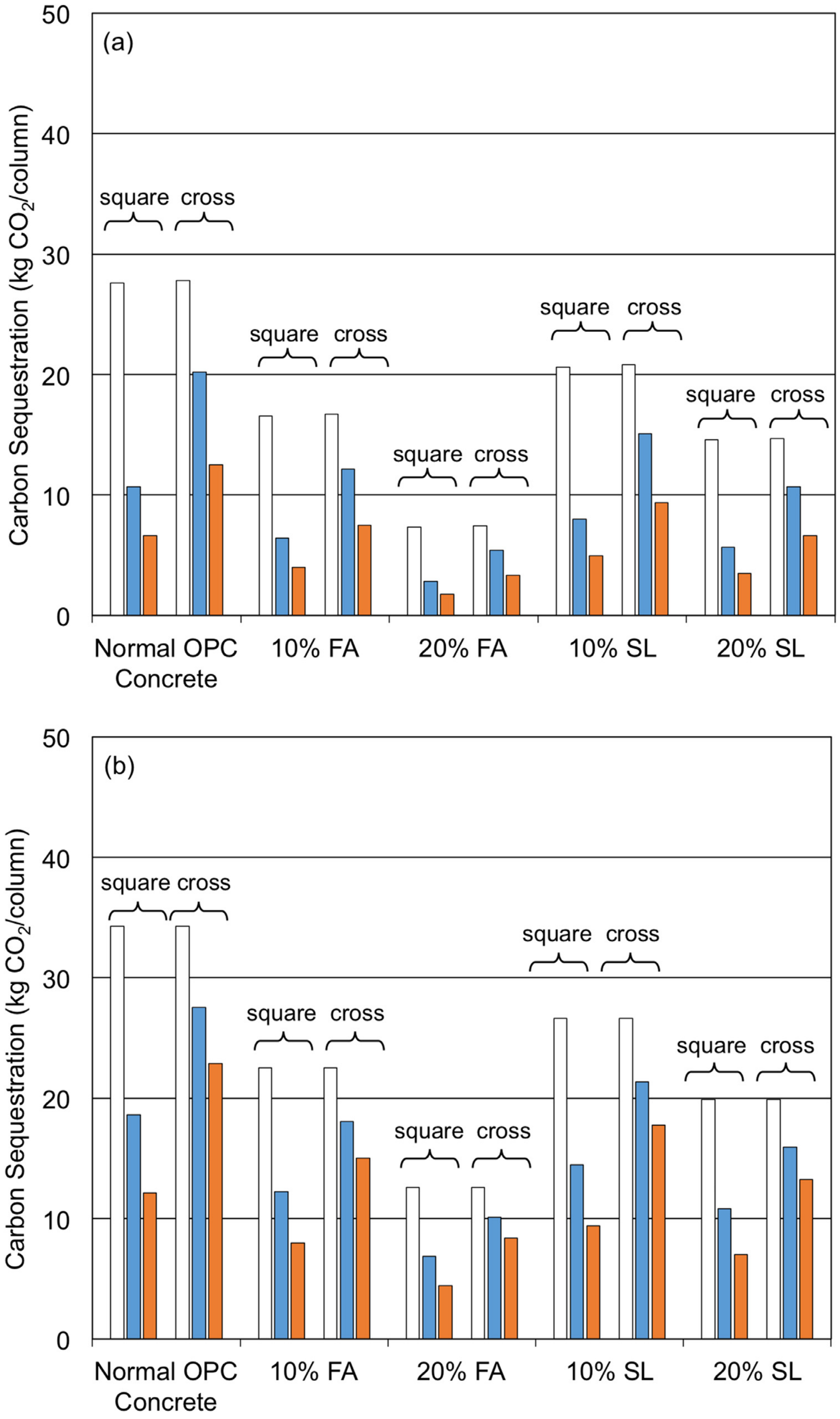

Fig. 6. Estimated sequestered CO_2 after 25 years of exposure by functionally equivalent square- and cross-shaped columns designed with either (a) Type I or (b) White cement 15 MPa (○), 30 MPa (●) and 45 MPa (●) concrete.

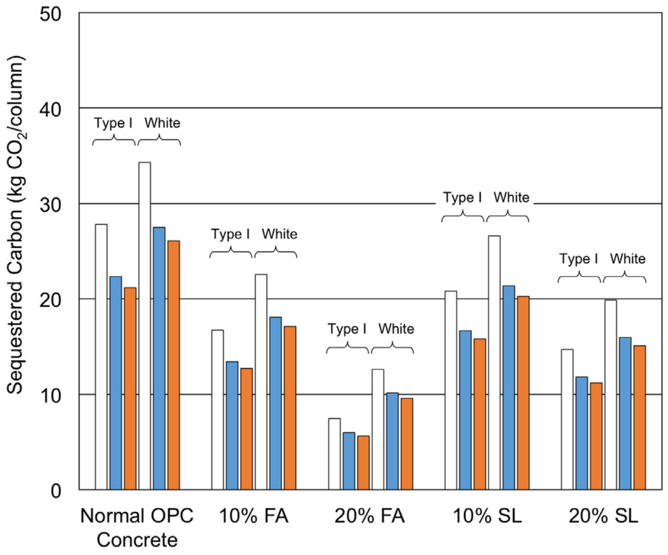

Fig. 7. Estimated sequestered CO_2 after infinite exposure by functionally equivalent columns designed with either Type I or White cement 15 MPa (○), 30 MPa (●) and 45 MPa (●●) concrete.

hydrate (CSH). This pozzolanic reaction renders less CH available for CO_2 sequestration [37].

The CO_2 sequestration model effectively reduces the CO_2 sequestration potential of cement paste accordingly *via* the β factor, which varies based on the silica content of the SCM. When fly ash replaces a percentage of cement, for example, lower quantities of CO_2 are sequestered in comparison with concretes that replace the same percentage of cement with slag. For instance, a Type I 15 MPa square column sequesters 27.6 kg CO_2 after 25 years of exposure, while the same column with 10% or 20% slag replacement reduces this amount to 20.6 and 14.6 kg CO_2, respectively. If fly ash were used instead, these quantities are reduced to 16.7 kg CO_2 and 7.4 kg CO_2. This difference is attributable to the lower silica content of slag, represented by β the factor for SCMs listed in Table 4.

3.2.2. Infinite exposure of functionally equivalent columns

Concrete columns of equal compressive strength, regardless of geometry (square- or cross-shaped), yield equivalent volumes of concrete (see Table 2). Therefore, total sequesterable CO_2 after infinite exposure is equal for both geometries. Fig. 7 shows the theoretical maximum sequesterable carbon emissions of the concrete columns investigated herein.

Furthermore, higher strength concretes exhibit less CO_2 sequestration compared to lower strength concretes. This reduction is evident for both Type I and White cement concrete columns with 0%, 10%, and 20% SCM replacement. Past research has shown that, for equivalent volumes of concrete, higher compressive strength concretes sequester higher amounts of CO_2 due to higher cement contents [38]. However, this result is mathematically obtained, given that the reduced volumes of the high-strength concrete columns correspond to lower total amounts of cement required to ensure functional equivalency of those columns and, in turn, corresponds to an overall reduction in CO_2 sequestration potential.

3.3. Net lifecycle carbon emissions

To quantify net lifecycle CO_2e emissions, estimates of sequestered CO_2 discussed in Section 3.2 were subtracted from initial CO_2e emissions that were quantified *via* the cradle-to-gate LCA and presented in Section 3.1.2. Results are shown in Fig. 8 for square- and cross-shaped columns after 25 years of exposure, respectively, and Fig. 9 after infinite exposure to CO_2.

As shown in Figs. 8 and 9, higher compressive strength concretes result in lower total lifecycle carbon emissions. Since a unit volume of high compressive strength concretes emit higher emissions and were shown to sequester lower amounts of CO_2, the lower lifecycle CO_2e emissions can be attributed to smaller volumes of concrete required for the structural column.

As discussed, when SCMs are used as a cement replacement, lower initial carbon emissions are observed during manufacture but result in lower *in situ* CO_2 sequestration. When both phases are included in the LCA (Fig. 7), total impacts are lower for higher strength concretes with higher additions of SCMs. For instance, after 25 years, the net emissions of a Type I 15 MPa square concrete column are 138 kg CO_2e. However, for identical concretes with 10% and 20% FA replacement, this amount decreases to 134 kg CO_2e and 127 kg CO_2e, respectively. In cases were slag is used, the total impacts for the concrete columns investigated herein are lower, despite initially higher embodied carbon. These findings indicate that, while the use of SCMs reduce CO_2 sequestration potential, their initial reductions in emissions result in total net emissions lower than any other concrete mixture formulation, even when estimates of CO_2 sequestration are included in the LCA.

Net carbon emissions after infinite exposure are shown in Fig. 9. Similar to the results of the finite exposure analysis, higher strength and Type I cement concretes with SCM addition exhibit the lowest total carbon emissions. This trend is similar to initial carbon emissions (see Section 3.1) of functionally equivalent columns. Maximum total carbon emissions correspond to a White cement 15 MPa column with no SCM replacement (152 kg CO_2e), while minimum total impacts correspond to a Type I 45 MPa column with 20% slag replacement (90 kg CO_2e). Differences in total emissions between mixtures that use fly ash or slag are approximately 3 to 4 kg CO_2e, which accounts only for a 2% of total emissions. These findings suggest that, while White cement concretes exhibited higher CO_2 sequestration potential, the amount of sequestered carbon was insufficient to reduce total net carbon below that of the Type I cement concrete columns analyzed herein. In addition, these results further demonstrate that net total emissions can be reduced up to 40% by mixture constituent selection and proportioning in concrete mixture design and that inclusion of SCMs and utilization of high-strength concretes are the best strategy for reducing overall net CO_2e emissions.

3.4. Recoverable carbon

The data in Table 5 show that, while the total amount of initial CO_2e emissions recovered *via* CO_2 sequestration increases with exposure time, high-strength concretes, low SCM replacement, and low-C_4AF cements (i.e., White cement), these trends are reversed at early exposure ages. When using SCMs, the percentage of emissions recovered by sequestration is reduced by approximately 50% when an additional 10% SCM replacement is specified. Slag-containing concretes recover a slightly higher percentage of initial carbon emissions than fly ash concretes, which can also be attributed to the lower silica content of slag. For example, after infinite time, a 20% slag replacement can recover up to 9% of initial carbon emissions, while a 20% fly ash replacement can recover up to 4.5%.

In addition, according to Table 5, the data indicate that up to 18.8% of initial carbon emissions can be recovered when CO_2 sequestration is included in the scope of the LCA for the concrete columns investigated in this analysis. These results align well with the percentages previously determined by Yang et al. [17]. However, García-Segura et al. [16] and Collins [14] reported higher percentages (up to 48%) of carbon could be recovered. Differences in these percentages likely arise from variability of system boundaries, assumptions (e.g., degree of carbonation), geography, and

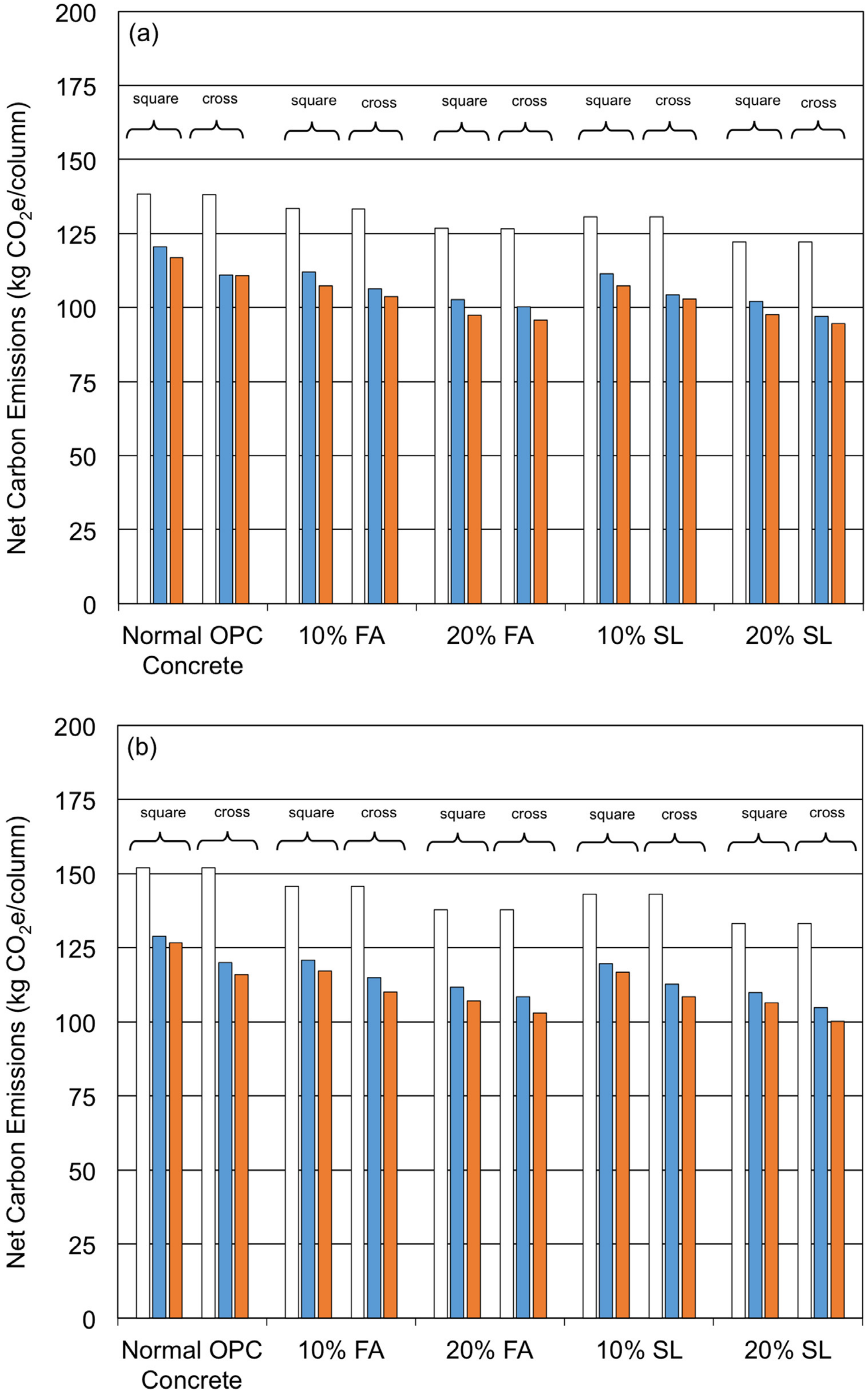

Fig. 8. Total net carbon emissions (kg CO_2e/column) after 25 years of exposure of functionally equivalent square- and cross-shaped columns designed with either (a) Type I or (b) White cement 15 MPa (○), 30 MPa (●) and 45 MPa (●) concrete.

more simplified models used to predict CO_2 sequestration. For instance, García-Segura et al. [16], who based the study in Valencia, Spain, calculated lower initial emissions during concrete manufacture, which, given similar predictions of sequesterable CO_2 as obtained in this study, resulted in a higher percentage in recoverable carbon.

The maximum recoverable carbon shown in Table 5 corresponds to emissions of a White cement 15 MPa column with no SCM replacement, which equal 186.2 kg CO_2e when CO_2 sequestration is not considered and 151.9 kg CO_2e when sequestration is included in the LCA. However, the total net emissions, as elucidated in Fig. 9, illustrate that, while the total recoverable carbon

Table 5
Percentage of carbon emissions recovered *via* CO_2 sequestration for the functionally equivalent column.

	Compressive strength					
	15 MPa		30 MPa		45 MPa	
	Type I	White	Type I	White	Type I	White
Finite exposure (25 Years)						
Normal OPC concrete	16.6%	18.4%	8.2%	12.6%	5.4%	8.7%
10%FA	11.0%	13.4%	4.9%	8.3%	3.2%	5.7%
20%FA	5.5%	8.4%	2.2%	4.6%	1.4%	3.2%
10%SL	13.6%	15.7%	6.1%	9.8%	4.0%	6.8%
20%SL	10.7%	13.0%	4.3%	7.3%	2.8%	5.1%
Infinite exposure						
Normal OPC concrete	16.8%	18.4%	17.0%	18.7%	17.1%	18.8%
10%FA	11.1%	13.4%	11.3%	13.6%	11.4%	13.7%
20%FA	5.5%	8.4%	5.6%	8.5%	5.7%	8.6%
10%SL	13.7%	15.7%	14.0%	15.9%	14.1%	16.0%
20%SL	10.7%	13.0%	10.9%	13.2%	11.0%	13.3%

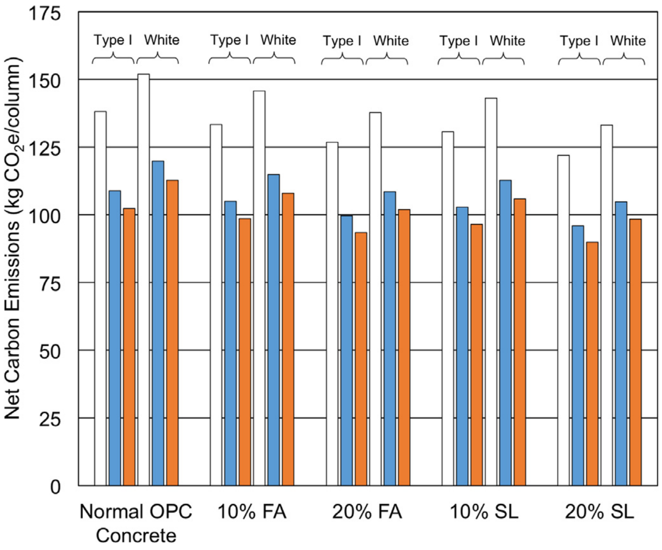

Fig. 9. Total net carbon emissions (kg CO_2e/column) after infinite exposure of a column designed with either Type I or White cement 15 MPa (○), 30 MPa (●) and 45 MPa (●) concrete.

is highest for this concrete column, the overall net emissions are higher than any other functionally equivalent concrete column investigated in this study. These data substantiate that, while the total mass of CO_2 sequestered for certain concrete mixtures and applications is certainly non-trivial, the ultimate magnitude of the total net carbon emissions is of greatest importance rather than the percentage of initial carbon emissions recoverable *via* sequestration.

4. Conclusion

This paper quantified and compared the initial carbon dioxide equivalent (CO_2e) emissions with estimates of sequesterable CO_2 for several OPC concrete columns at finite and infinite time intervals. The following were key insights elucidated in this work:

- Higher compressive strength concretes exhibited lower total emissions when CO_2 sequestration was included in the lifecycle assessment (LCA). While high-strength concretes contain higher amounts of carbon-intensive cement, less volume of cement is required to ensure functional equivalency in a simple structural application.
- When SCMs were used as a cement replacement, total embodied carbon was lower for all cases, despite lower estimates of CO_2 sequestration potential.
- While fly ash-containing concretes exhibited the lowest initial (cradle-to-gate) embodied carbon, slag-containing concretes were observed to exhibit the lowest net CO_2e emissions when CO_2 sequestration was included in the scope of the LCA.
- Both initial CO_2e emissions (a negative environmental benefit) and the CO_2 sequestration potential (a positive environmental benefit) of White cement concrete were highest; however, the total net CO_2e emissions of White cement concretes were higher than all other concrete mixture formulations investigated herein. This result elucidated the necessity and importance of quantifying net CO_2e emissions in tandem with total *in situ* CO_2 sequestration.
- LCA studies that negate *in situ* CO_2 sequestration may be overestimating concrete emissions up to approximately 19%, a non-trivial sum, but the overall net magnitude of CO_2e emissions is more important than the ratio of sequesterable CO_2 versus initial CO_2e emissions.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.enbuild.2018.02.042.

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