

# Reducing greenhouse gas emissions for prescribed concrete compressive strength

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## HIGHLIGHTS

- Equations are developed for green engineering of concrete with specified strength.
- Higher compressive strength typically results in higher GHG emissions in concrete.
- Optimal use of SCMs to reduce GHG emissions varies depending on their properties.

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## ABSTRACT

Often, when proportioning “green” concrete mixtures, the use of Supplementary Cementitious Materials (SCMs) is deemed to be favorable. While appearing more in the literature, it is still not commonplace that design strength is considered in assessments of environmental impacts. When mechanical or material properties are incorporated into environmental impact assessments, comparisons are drawn based on the property attainable for any given mixture, most commonly, compressive strength. However, in structural applications, compressive strength is typically specified in the design and there are currently no means for specifying the best concrete constituents for a set compressive strength to reduce greenhouse gas emissions from production. In this research, concrete mixture proportioning based on water-to-binder content and supplementary material-to-Ordinary Portland Cement ratios are examined. For these parameters, mathematical models are developed to perform optimization of GHG emissions for four groups of concrete (those containing varying levels of fly ash, ground granulated blast furnace slag, natural pozzolans, and limestone filler as a means to reduce Ordinary Portland Cement content). For the particular binary blended binders examined in this work, optimal ratios of supplementary material use were highly dependent on the type of alternative material and were consistently below the highest replacement level considered. The equations developed will facilitate the green engineering of concrete when a specified strength is required.

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## 1. Introduction

Growing urban populations and the demand for infrastructure are resulting in a growth of cement consumption globally [1,2]. The production of hydraulic cement has seen a steep incline within the past two decades with global production from 1950 to 2000 growing by 1.5 gigatonnes (Gt) and production in the subsequent 15 years, 2001–2015, growing by over 5 times the rate seen in the previous 5 decades with an increase in production of 2.4 Gt [3,4]. This high cement production produces notable greenhouse gas (GHG) emissions, estimated at 8.6% of total anthropogenic

GHG emissions in 2012 [5]. This growing demand for a material that is resulting in substantial GHG emissions highlights the need to develop means to produce a structural material that can meet the needs of society while limiting its burden on the environment.

While there are many ways to reduce the emissions associated with the production of concrete, including use of more efficient equipment, alternative fuels, and carbon capture and storage (e.g., [6,7]), one of the most prevalent means to reduce emissions is to reduce the demand for Ordinary Portland Cement (OPC). OPC is one the most prevalent binders in concrete and current production methods involve using high levels of clinker, a kilned and quenched material, that requires high energy input during production, resulting in GHG emissions, as well as produces material-derived CO<sub>2</sub> emissions through processes such as the chemical

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### Nomenclature

$b$	binder content by weight ( $\text{kg/m}^3$ )	$k_C$	constants which depend on the manufacturing of supplementary cementitious materials
$c$	ordinary Portland Cement content by weight ( $\text{kg/m}^3$ )	$L$	limestone
$f_c$	compressive strength of concrete (MPa)	$NP$	natural pozzolans
$FA$	fly ash	$OPC$	ordinary Portland Cement
$i$	volumetric environmental impact (environmental impact/ $\text{m}^3$ )	$s$	supplementary cementitious material or filler content by weight ( $\text{kg/m}^3$ )
$GGBS$	ground granulated blast furnace slag	$w$	water content by weight ( $\text{kg/m}^3$ )
$k_{1,2}$	constants which depend on the concrete materials used	$\alpha, \beta_{1,2}, \gamma_{1,2}, \xi_{1,2}$	empirically derived parameters to relate constants $k_{1,2}$ to ratio of supplementary cementitious materials or filler to Ordinary Portland Cement content
$k_{3,A}$	constants which depend on cement manufacturing		
$k_{4,B,D}$	constants which depend on materials and processing used		

reaction in which limestone is converted to lime. One way to reduce the quantity of OPC necessary is increased use of Supplementary Cementitious Materials (SCMs) or increased use of well-dispersed inert fillers (e.g., [8]). SCMs can include a wide range of materials including natural pozzolans (e.g., diatomite), thermally treated clays (e.g., calcined clay), and industrial byproducts (e.g., fly ash from coal combustion and slag from metal refinement). These materials not only offer potential benefits in terms of reducing GHG emissions from the production of concrete, but also offer benefits to certain material and durability properties [9–11]. However, while the environmental impacts of inclusion of SCMs in concrete has been examined, how to use them most efficiently while meeting material property requirements has not been well studied.

## 2. Background

The use of SCMs and other means to develop “green” concretes have been thoroughly examined in the literature. Among the most prevalent means of evaluation is the use of life cycle assessment (LCA) methodology to quantify relative environmental impacts associated with concrete production and the influence of changing mixture proportions [12]. Many such studies have examined the role of improved equipment efficiency, use of alternative fuels, and increased use of industrial byproducts, for example fly ash and blast furnace slag, among others, to reduce clinker content in cement or to reduce use of clinker-intensive cement (e.g., [6,13–15]). Research has expanded further to assess individual mixtures based on production in a certain location to assess benefits or drawbacks to concrete constituent selection when trying to reduce environmental impacts (e.g., [16–20]). Due to the prevalence of industrial byproducts as a means to reduce the demand for clinker in concrete, the role of allocation of environmental impacts from initial production has also been a growing field. Rather than modeling these byproducts as having no associated impact from production, but only refinement, when required, and transportation prior to use, the impacts from production are considered at a certain degree using allocation methods (e.g., [21–23]).

Most commonly the investigation of life cycle impacts is based on a constant volume or mass of material produced (either concrete or cement) (e.g., [12]). However, as has been discussed by several authors, the role of material properties or longevity in an application will directly influence the volume of material or maintenance and replacement necessary (e.g., [24–26]). Recent efforts to change the units of comparison used in the assessment of concrete have attempted to overcome these factors. The most common methods implemented are comparisons using application of case studies (e.g., [16,27]) and using either emissions or binder per cubic meter of concrete as a fraction relative to compressive strength achieved (e.g., [28–30]). More recently, the contributions

of concrete design and durability have started to be included in comparisons of mixtures (e.g., [31,32]). While these methods mark an improvement to common volume- or mass-based comparisons, they have the potential to counter design specifications; for example, it is possible that these new comparison techniques could suggest a very low strength or very high strength concrete that would be undesirable from a structural engineering or construction viewpoint. As such, there is a gap in understanding how concrete mixtures should be best proportioned to reduce environmental impacts while meeting specific design requirements.

The objective of this research is to develop a set of mathematical equations that facilitate finding a minimum environmental impact ( $i$ ), in this case GHG emissions, for a specified constant compressive strength of concrete ( $f_c$ ) and to dictate the necessary concrete mixture proportions, as are given in this work in terms of water-to-binder ( $w/b$ ) and supplementary material to OPC ( $s/c$ ) ratios, to get an optimized environmental impact.

## 3. Materials

To investigate applicability of equations developed for a variety of concrete mixtures, four groups of concrete mixtures containing different SCMs or fillers were examined. For this research, binary blends were considered to reduce complexity of equations developed; however, principles can be extended to other SCMs and blended binders. Specifically, this research considers concrete mixtures containing Ordinary Portland Cement (OPC) with natural pozzolans (NP), limestone fillers, ground granulated blast furnace slag (GGBS), or Class F fly ash (FA). While there are many viable SCMs that could have been considered, to present the methods discussed in this research, this subset of SCMs facilitates the visualization of the work presented. To facilitate equation development, 20 concrete mixtures with NP contents ranging from 0 to 0.82 NP:OPC, 25 mixtures with limestone ranging from 0 to 0.82 L:OPC, 32 mixtures with GGBS ranging from 0 to 1.57 GGBS:OPC, and 28 mixtures with FA ranging from 0 to 0.58 FA:OPC were used. Data for mixture proportions and compressive strength were from Meddah [33], Meddah et al. [34], Oner and Akyuz [35], and Oner et al. [36]. Equations developed by Yi et al. [37] were used to account for variations in specimen dimensions. Table 1 shows the nomenclature used for mixtures assessed in this project. Each group of concrete mixtures examined in this research was considered using 28-day compressive strength data with varying  $w/b$  and  $s/c$  ratios; mixture proportions and properties used can be found in the Appendix. Limited data sources were used to provide visualization of methods while improving uniformity in data. However, for any set of comparable data, the methods developed in this research could be applied.

**Table 1**  
Mixture nomenclature.

Name	Description	Data Source
NP-OPC	Concrete mixtures containing Ordinary Portland Cement and varying levels of natural pozzolans (0–0.82 NP:OPC)	Meddah [33]
L-OPC	Concrete mixtures containing Ordinary Portland Cement and varying levels of limestone filler (0–0.82 L:OPC)	Meddah et al. [34]
GGBS-OPC	Concrete mixtures containing Ordinary Portland Cement and varying levels of ground granulated blast furnace slag as the binder (0–1.57 GGBS:OPC)	Oner and Akyuz [35]
FA-OPC	Concrete mixtures containing Ordinary Portland Cement and varying levels of fly ash as the binder (0–0.58 FA:OPC)	Oner et al. [36]

#### 4. Life cycle assessments

To calculate environmental impacts used in this research, an adaptation of life cycle assessment (LCA) methodology, was implemented. LCA methodology quantifies material, energy, emissions, and waste flows during every life stage of a material or product to facilitate social, economic, or environmental impact assessment. In this research, the method was used for cradle-to-gate, i.e., from raw material acquisition through production of concrete on a cubic meter basis, to understand greenhouse gas (GHG) emissions for the mixtures examined. To determine the GHG emissions for these concrete mixtures, the GreenConcrete LCA tool, developed by Gursel and Horvath [38], was applied.

GHG emissions associated with extraction of raw materials, including quarrying and/or crushing of excavated materials, such as those used in the production of cement, NPs, limestone, and aggregates, were assessed. For the production of OPC, beyond the raw material acquisition and refinement, GHG emissions from pre-homogenization, grinding, blending, pyroprocessing of clinker, cooling and finishing of clinker, as well as blending with gypsum, were analyzed. For the other extracted materials, e.g., aggregates, NP, and limestone fillers, further preparation processes, such as sieving and grinding, were considered. FA and GGBS are co-product materials associated with the combustion of coal for energy sources and the production of iron and steel, respectively. The quantification of emissions associated with the production of such materials is under debate as to whether only emissions associated with collection, refinement when required, and transportation should be included when considering them as SCMs (e.g., [17]) or whether some emissions from the primary process (either coal combustion or iron/steel production) should be allocated to these materials (e.g., [21]). For the baseline scenario in this work, FA and GGBS are considered without emissions allocated from production; only emissions associated with processing post-production and transportation were assessed. To examine the difference in results, a secondary scenario in which mass-based allocation (based on calculations by Miller [39]) are applied; these values from allocation were added to those used in the baseline for post-production processing. Transportation of materials, energy mixes, and equipment efficiency were included for each concrete constituent as well as for concrete batching. For these values, production of concrete was assumed to occur in the San Francisco Bay Area as described by Celik et al. [18]. One location was selected to perform LCAs to normalize effects of energy mixes, equipment efficiency, and transportation, which would be expected to vary based on location; however, the method presented in the subsequent section could be conducted for any location of interest.

#### 5. Mixture proportioning for specified strength

To develop a system of equations to facilitate the determination of mixture proportions that will result in reduced GHG emissions for a specified concrete strength, mixture design relationships were analyzed. The equations developed are primarily applicable

to concrete mixtures with negligible influences from chemical admixtures and one or no mineral admixtures, but they offer an initial step in defining a means to strategically reduce GHG emissions for prescriptive concrete design in which strength is pre-specified.

##### 5.1. Formulas for relationship between concrete constituents and compressive strength

To correlate concrete compressive strength with constituents used, Abram's relationship was applied. This relationship that classically associates compressive strength water-to-cement ratio, building from empirical knowledge regarding paste volume and concrete compressive strength, is applied in this work in a modified form. Namely, water-to-binder ratio ( $w/b$ ), that is the ratio between the water content to the OPC and SCMs combined, was applied. Additionally, a weight based ratio for  $w/b$  is used as opposed to a volume. Extensions such as these have been successfully applied in past work (e.g., [40,41]). This relationship gives:

$$f_c = \frac{k_1}{k_2^{w/b}} \quad (1)$$

where  $f_c$  is the compressive strength, and  $k_1$  and  $k_2$  are constants determined through fitting to experimental datasets. Because the binder can consist of more than one material, this relationship is extended to incorporate factors for quantifying the extent to which the binder is formed of a particular SCM. In this work, the extension is examined through defining the parameters  $k_1$  and  $k_2$  as functions of the supplementary material to OPC ratio ( $s/c$ ). To redefine  $k_1$  and  $k_2$ , simple polynomial curve fitting was applied giving the following equations:

$$k_1 = \alpha(s/c)^3 + \beta_1(s/c)^2 + \gamma_1(s/c) + \xi_1 \quad (2)$$

$$k_2 = \beta_2(s/c)^2 + \gamma_2(s/c) + \xi_2 \quad (3)$$

where  $\alpha$ ,  $\beta_1$ ,  $\beta_2$ ,  $\gamma_1$ ,  $\gamma_2$ ,  $\xi_1$ , and  $\xi_2$  are empirically derived constants determined from the datasets used that allow  $k_1$  and  $k_2$  to be defined as functions of  $s/c$ . In the mixtures examined in this work, water content was held constant for each group of concrete mixtures. As a result, a varying  $w/b$  ratio reflected an increase in binder as opposed to a constant binder content, for which varying  $w/b$  ratios would reflect changes in water content. With the water content held constant and by defining the binder content by weight ( $\text{kg}/\text{m}^3$ ),  $b$ , as the sum of the supplementary material by weight ( $\text{kg}/\text{m}^3$ ),  $s$ , and the OPC content by weight ( $\text{kg}/\text{m}^3$ ),  $c$ :  $b = s + c$ , Abram's relationship can be rewritten using Eqs. (2) and (3) in terms of the supplementary material content and cement content:

$$f_c = \frac{\alpha(s/c)^3 + \beta_1(s/c)^2 + \gamma_1(s/c) + \xi_1}{(\beta_2(s/c)^2 + \gamma_2(s/c) + \xi_2)^{\frac{w}{s+c}}} \quad (4)$$

This equation only contains two variables, supplementary materials content ( $s$ ) and cement content ( $c$ ), which are used in subsequent sections to relate GHG emissions from concrete pro-

duction to compressive strength for each group of concrete mixtures. Further, it can be noted that the OPC content and the SCM content can be defined in terms of  $k_1$ ,  $k_2$ ,  $f_c$ , and the  $s/c$  ratio by rewriting Abram's relationship, Eq. (1), and again applying the relationship  $b = s + c$ :

$$c = \frac{w}{1 + \frac{s}{c}} \cdot \frac{\ln k_2}{\ln \frac{k_1}{f_c}} \quad (5)$$

$$s = \frac{w}{1 + \frac{s}{c}} \cdot \frac{\ln k_2}{\ln \frac{k_1}{f_c}} \quad (6)$$

where again,  $s/c$  is the replacement ratio of SCM or filler to OPC content by weight,  $w$  is the water content by weight, and all other terms are the same as previously defined. This equation is based on two conditions: 1)  $k_1/f_c$  must not be equal to 1; and 2)  $f_c$ ,  $k_1$  and  $k_2$  should be greater than zero.

## 5.2. Mathematical relationships for GHG emissions and concrete constituents

The previous equations allow for the determination of compressive strength as functions of  $c$  and  $s$ , which can be used to provide  $w/b$  and  $s/c$  ratios. The environmental impact, which is GHG emissions for this work, must be defined in similar terms to the compressive strength. For several environmental impacts, including GHG emissions, the environmental impact is nearly a linear function of the OPC content ( $c$ ) if little to no environmental impacts are attributed to the other concrete constituents or production processes. As such, a linear equation can be developed that defines environmental impact,  $i$ , as a function of  $c$  [40]:

$$i = k_3c + k_4 \quad (7)$$

where  $k_3$  and  $k_4$  are constants as defined based on the emissions associated with the production of cement and the remaining emissions from the production of concrete, respectively. This function, or ones similar to it, is a relatively common method of defining GHG emissions for concrete, but if high SCM replacement ratios (i.e., high ratios of  $s/c$ ) are used or if any allocation is given to the SCMs, it can become inaccurate. Therefore, in this work it is proposed to use system of equations that follow:

$$i_1 = k_Ac + k_B \quad (8)$$

$$i_2 = k_Cs + k_D \quad (9)$$

$$i = i_1 + i_2 \quad (10)$$

where  $k_A$  and  $k_C$  are constants reflecting GHG emissions associated with cement production and SCM production, respectively,  $k_B$  and  $k_D$  are constants reflecting remaining GHG emissions associated with concrete production, and  $i_1$ ,  $i_2$  are the contribution to total volumetric environmental impact,  $i$ , (i.e., the environmental impact for the production of a set volume). Note, constant  $k_D$  reflects changes to remaining concrete production associated with increased SCM use (e.g., reduced aggregate use associated with a higher SCM content and larger binder volume). Using these relationships and the formula developed that defines compressive strength as a function of  $c$  and  $s$ , for a constant compressive strength, one can solve for  $i$ .

Applying the definitions provided in Eqs. (5), (6), (8) and (9), Eq. (10) can be rewritten as:

$$i = k_A \frac{w}{1 + \frac{s}{c}} \cdot \frac{\ln k_2}{\ln \frac{k_1}{f_c}} + k_C \cdot \frac{w}{1 + \frac{s}{c}} \cdot \frac{\ln k_2}{\ln \frac{k_1}{f_c}} + k_B + k_D \quad (11)$$

It must be noted that this equation is only applicable for  $s/c$  in the domain for which the equations have been experimentally validated. Further, based on common mixture proportions and the

likely range to achieve appropriate hydration as well as consistency, the range for the  $w/b$  ratio is limited. Thus, two important boundaries are applied:

$$\begin{cases} 0.25 \leq \frac{w}{b} \leq 1 \\ 0 \leq \frac{s}{c} \leq \text{maximum} \end{cases} \quad (12)$$

Note that all parameters can be empirically derived for groups of concrete mixtures (e.g.,  $k_A$ ,  $k_B$ ,  $k_C$ ,  $k_D$ ) and/or defined in terms of mixture proportioning parameters,  $w/b$  and  $s/c$ , (e.g.,  $k_1$ ,  $k_2$ ). Because  $k_1$  and  $k_2$  can be rewritten in terms of  $s/c$ , as shown in Eqs. (2), (3), and (4), the environmental impact for a volume of concrete,  $i$ , can be written as a function of empirical constants, the  $w/b$  and  $s/c$  ratios, and the compressive strength. Using this new formula, which is a function of  $c$  and  $s$  for a set  $f_c$ , derivatives can be taken to find the local minima with respect to  $w/b$  and  $s/c$  to provide the lowest  $i$  for a set  $f_c$ .

## 6. Results

### 6.1. Parameters developed and representativeness

Applying the equations developed in Section 5 to the four groups of materials analyzed in this research, parameter fitting was conducted. For each of the four groups, namely, FA-OPC, GGBS-OPC, NP-OPC, and L-OPC, the compressive strength data provided by the literature and environmental impact assessment derived values for GHG emissions from production of each of the mixtures assessed, facilitated determination of parameters defined in this research. The values of constants in the equations developed for each group of concrete and the associated  $R^2$ -values are presented in Table 2; constants for fitting Abram's relationship (Eq. (1)) at each  $s/c$  ratio considered for the concrete mixtures and the associated  $R^2$ -values are presented in the Appendix.

To visualize the ability of the mathematical models developed in this work to capture compressive strength and GHG emissions from production, these factors were plotted relative to water-to-binder ratio (Fig. 1). Specifically, using the models developed, GHG emissions for the production of one cubic meter of concrete relative to the compressive strength of said concrete (i.e., in units of kg CO<sub>2</sub>-eq/m<sup>3</sup>/MPa) was plotted against the water-to-binder ratio at each  $s/c$  ratio for which there was experimental data for the four groups of concrete. The empirical values, that is, the values of calculated GHG emissions using LCA methodology and the experimentally derived strength values, were overlaid as data points (See Fig. 1).

From this assessment of GHG emissions relative to compressive strength, the ability of the mathematical models to capture both mechanical properties and environmental impact can be examined. For all mixtures assessed, the models are more accurate for lower water-to-binder ratios. For the FA-OPC, GGBS-OPC, and NP-OPC mixtures, the models capture the empirical data points well (percentage root mean square error of 3–6% for the FA mixtures, 2–13% for the GGBS mixtures, and 2–5% for the NP mixtures). For the limestone mixtures, the correlations are weaker (percentage root mean square error of 8–24%). The greater range in fit for the L-OPC mixtures is likely a reflection of the mixtures and methods selected. This assessment method assumes the material replacing clinker-based OPC has cementitious or pozzolanic properties. While the experimental data and the models developed in this work assess the limestone filler as a replacement of cement, both represent an increase in limestone filler content as a correlated decrease in OPC, the filler itself does not behave as an SCM. The reflection of the SCMs as a replacement for clinker-based OPC as opposed to additives in the production of cement is in part a function of the sources of experimental data used. However, the



method developed in this research could be applied more broadly to other forms of replacement.

## 6.2. Greenhouse gas emissions for specified strengths

The four groups of concrete mixtures are examined at six specified compressive strengths and predicted GHG emissions are calculated from production as a function of supplementary material to OPC content ( $s/c$ ) for these mixtures (shown in Fig. 2). The trends in GHG emissions from concrete production as a function of  $s/c$  vary in several aspects for each group of concrete analyzed. One difference is the FA-OPC and GGBS-OPC typically have lower GHG emissions from production than that of solely OPC as the binder for each of the concrete strengths examined. For the NP-OPC mixtures, while high  $s/c$  ratios (over 0.25) show a decrease from the OPC-only binder mixtures, low  $s/c$  ratios (0.25 and below) display an increase in GHG emissions for the production of concrete containing NP as an OPC replacement for each of the six compressive strengths modeled. This increase suggests the benefits associated with a reduced GHG emitting binder are lost when a certain compressive strength is being sought if only low levels of replacement are used. For the L-OPC mixtures, while the inclusion of limestone with an associated reduction in OPC consistently shows a decrease in GHG emissions for each of the compressive strengths modeled for the range of  $s/c$  ratios considered, it is clear that the greatest benefit of this method for reducing OPC is at low  $s/c$  ratios, with a minimum hovering around 0.2.

Looking individually at each group of concrete and strength, a minimum for GHG emissions from production is noted for each at an  $s/c$  ratio greater than 0. That is to say, for each of the mixtures, the use of SCMs or fillers consistently can offer a potential means to lower GHG emissions relative to a concrete containing OPC as the only binder; however, when these minima are achieved is dependent upon type of alternative material (including influences of properties within classifications of SCMs or fineness, which were not examined in this work), water content, and strength desired. For the mixtures assessed, it can be seen for the GGBS-OPC (Fig. 2b), there is a decline in GHG emissions of 55–125 kg CO<sub>2</sub>-eq/m<sup>3</sup> when the  $s/c$  ratio increases from 0 to 0.5. Yet, when the  $s/c$  ratio increases from 0.5 to 1.57, the drop in GHG emissions is at most 2–30 kg CO<sub>2</sub>-eq/m<sup>3</sup>, with the greatest change for the highest strength concrete. Further, for this rise from 0.5 to 1.57 in the  $s/c$  ratio, the GGBS-OPC mixtures reach an optimal point of low GHG emissions. For the low strength concrete mixtures, the change from 0.5 to 1.57 in the  $s/c$  ratio could lead to an increase in GHG emissions. For FA-OPC (Fig. 2a), similar to GGBS-OPC, there is a steep initial decline in GHG emissions, between 40 and 80 kg CO<sub>2</sub>-eq/m<sup>3</sup> as the  $s/c$  ratio increases from 0 to 0.35, with minima between 0.38 and 0.44 depending on compressive strength specified. The minimum GHG emissions from production using NP-OPC, lies

around 0.7–0.75  $s/c$  ratio, depending on compressive strength; unlike the other mixtures for which any inclusion of an alternative material results in a decrease in GHG emissions, for the NP-OPC the maximum GHG emissions lies around 0.1  $s/c$  ratio. For the L-OPC mixtures, while the use of the filler does not result in an increase in GHG emissions for the mixtures considered, there is clearly a greater benefit to using the limestone at levels below 0.4  $s/c$  ratio, with minimum GHG emissions typically around 0.2  $s/c$  ratio for each of the compressive strengths. The optimal concrete mixture proportions to achieve the minimum GHG emissions and corresponding  $s/c$  and  $w/b$  ratios are shown in Table 3.

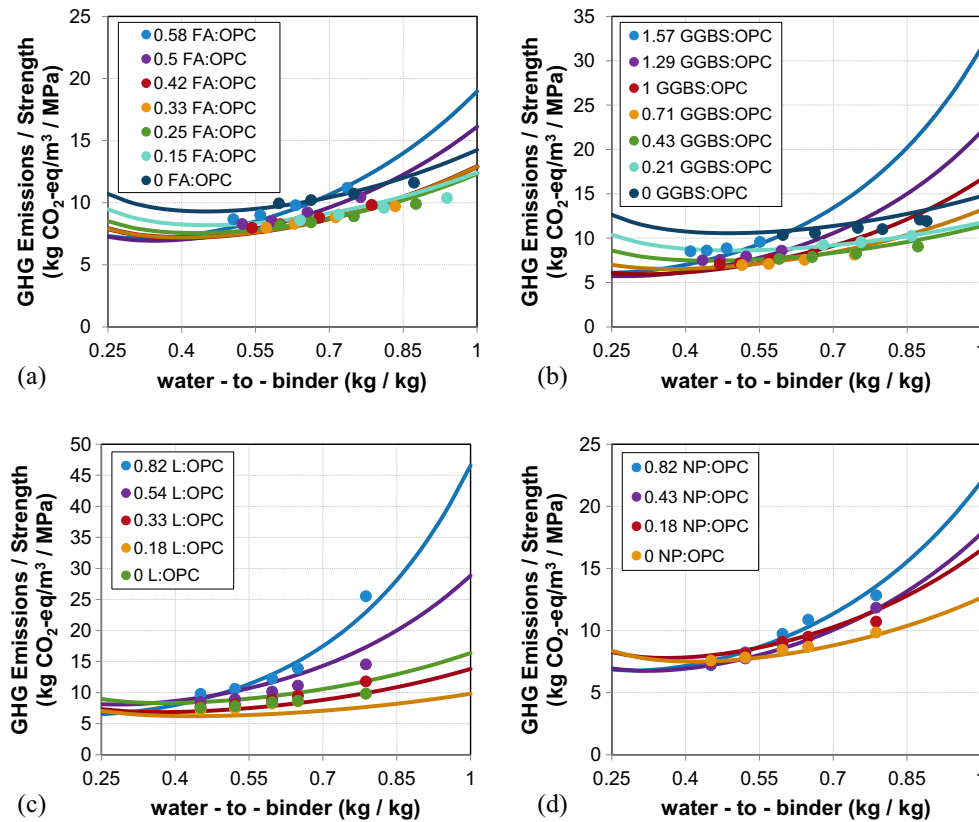
This behavior is to be expected for these SCMs and filler; the GGBS has cementitious properties, so for the 28-day strength properties used in this analysis, the GGBS would have significant contribution to strength and low replacement levels would be expected to have high benefits in reducing GHG emissions for specified strengths. For the limestone filler, while it itself is not cementitious, strategic use of cement grade limestone and dispersants has been shown to be able to be incorporated in binder up to 10% without significant loss in strength [42] and greater levels of replacement should be viable, but because of water reduction, the net benefit to cutting GHG emissions might be lower than the clinker fraction replaced [8]. For the pozzolans, Class F FA and the NP, their influence on hydration reactions and ultimate strength are favored at longer curing periods. If 56-day strength were examined rather than 28-day, higher  $s/c$  ratios would be expected to be favorable and the peak in the NP-OPC plot less pronounced.

While there are clear divergences among certain trends for the groups of concrete examined, there are also some similarities in these four plots. Higher specified compressive strength corresponds to higher GHG emissions from production for any set  $s/c$  ratio within a group of concrete mixtures. Higher compressive strength is based on a lower  $w/b$  ratio; because water content is constant for the equations developed, the higher strength concrete has higher binder content and higher corresponding GHG emissions. Also at higher compressive strength, the GHG emissions are more susceptible to the changes of  $s/c$  because the binder content itself is greater. As shown in Table 3, there is a corresponding increase in GHG emissions from the production of concrete and a decrease in  $w/b$  with higher specified compressive strengths. For the FA- and GGBS-OPC mixtures, higher  $s/c$  ratios are noted as favorable with increasing strength, yet, the opposite trend is present for the NP-OPC concrete mixtures and approximately the same  $s/c$  ratio is specified for each of the L-OPC mixtures, regardless of compressive strength.

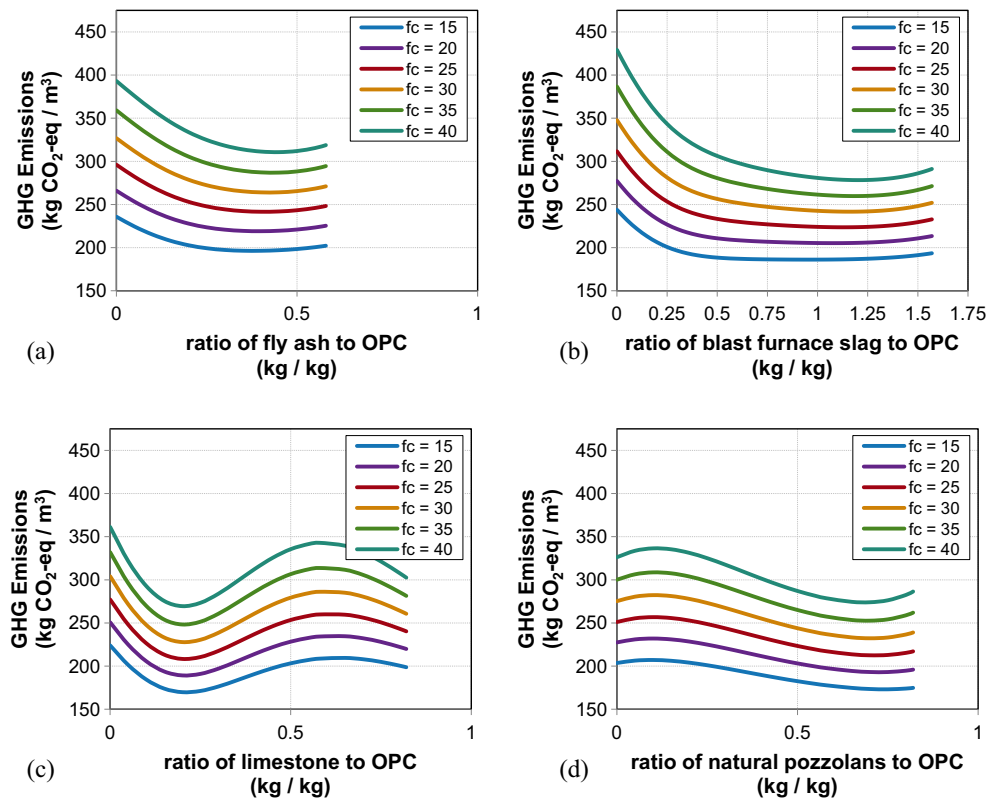
While the mixtures analyzed in this research are not representative of all mixtures containing FA-, GGBS-, NP-, or L-OPC mixtures, to further exemplify how the models developed could be applied, plots of expected GHG emissions for the production of

**Table 2**  
Values of constants in each class of concrete by polynomial fitting.

Concrete Type	PC-FA	R <sup>2</sup>	PC-GGBS	R <sup>2</sup>	PC-L	R <sup>2</sup>	PC-NP	R <sup>2</sup>
$k_A$	0.96	1	0.96	1	0.97	1	0.97	1
$k_B$	27.2		34.3		32.8		32.8	
$k_C$	0.037	0.99	0.14	1	0.017	0.92	0.31	0.97
$k_D$	−0.17		0.19		0.025		−0.66	
$\alpha$	−742	0.98	−84.8	0.99	459	0.98	−437	1
$\beta_1$	678		178		−378		470	
$\gamma_1$	−96.9		−21.5		44.0		−113	
$\xi_1$	147		121		157		157	
$\beta_2$	47.2	1	18.5	1	138	0.97	−8.26	0.99
$\gamma_2$	−8.77		−5.90		−46.3		23.4	
$\xi_2$	8.30		6.78		12.0		9.35	



**Fig. 1.** Predicted ratio of GHG emissions per cubic meter of concrete to concrete compressive strength as a function of water-to-binder ratio with empirical data plotted as points and models displayed as lines for (a) FA-OPC mixtures, (b) GGBS-OPC mixtures, (c) L-OPC mixtures, and (d) NP-OPC mixtures.



**Fig. 2.** Comparison of GHG emissions per cubic meter of concrete as a function of s/c ratio for six compressive strengths for (a) FA-OPC mixtures, (b) GGBS-OPC mixtures, (c) L-OPC mixtures, and (d) NP-OPC mixtures.

15 MPa and 35 MPa concretes relative to  $s/c$  ratio are shown for these mixtures concurrently (Fig. 3). The  $s/c$  ratio from 0 to 0.58 was chosen for these plots, as this was the largest range for which all groups of mixtures considered had experimental data. The compressive strengths are chosen for a low strength (15 MPa) and a high strength (35 MPa) to aid in visualization of differences present.

Fig. 3 shows that the trends of predicted GHG emissions for concrete mixtures with limestone are notably different from other classes of concrete mixtures at lower  $s/c$  ratios. Until approximately 0.04  $s/c$  ratio, the use of NP-OPC offers the lowest GHG emissions of the mixtures analyzed despite its growth in GHG emissions between 0 and 0.2  $s/c$  ratio relative to OPC as the sole binder. Past this point, the use of limestone as a replacement for OPC results in the lowest GHG emissions until approximately 0.38  $s/c$  ratio, at which point the NP-OPC mixtures become favorable again. The margin at which mixtures are desirable is greater for the higher strength concretes. It must be noted that the NP- and L-OPC mixtures contain a water content approximately 50 kg/m<sup>3</sup> lower than that of the FA- and GGBS-OPC mixtures. As such, the FA- and GGBS-OPC mixtures have inherently higher binder content to have similar water-to-binder ratios. This higher binder content results in higher GHG emissions as the binder is the main contributor to such emissions. This factor exhibits the degree to which certain parameter selection could influence goals to reduce GHG emissions and how concrete mixture proportioning should be specified based on multiple factors. It must be noted that the differences in binder content and the minor approximations made by the models result in some differences in expected GHG emissions for concrete mixtures with no SCM or filler at a specified strength. With more comparable datasets, this difference would diminish and more direct comparisons between supplementary material alternatives could be drawn.

### 6.3. Global minimum for each class of concrete via 3-D visualization

By applying the same equations developed in Section 5, minimum GHG emissions for the binary blended binder concrete mixtures can be determined without specified strength. In addition to the previous discussion of the applicability of these equations for prescribed concrete strengths, the relationship between GHG emissions from production,  $s/c$  ratio and compressive strength can be plotted (see Fig. 4). These plots show some of the phenomena previously discussed, such as when the compressive strength increases, the GHG emissions for the production of the concrete (shown as both the vertical axis and the color gradation in Fig. 4) increase. Similarly, as previously noted, the effects of using alterna-

tive materials as replacement of OPC have a more dramatic effect on the GHG emissions for concrete production at higher strengths; again, this phenomenon is pronounced due to the water content being held constant for these assessments and increased strength corresponding to increased binder content. For all mixtures assessed, the global minimum of GHG emissions, that is the minimum possible GHG emissions that can be achieved for each of the groups of concrete, occurs at the lowest strength considered. This trend shows that benefits achieved through use of supplementary binders and fillers do not outweigh prescribed properties. As such, the design and application of concrete is expected to play a significant role in the emissions associated with producing the material by dictating the required properties.

### 6.4. Assessment for environmental impact with allocations

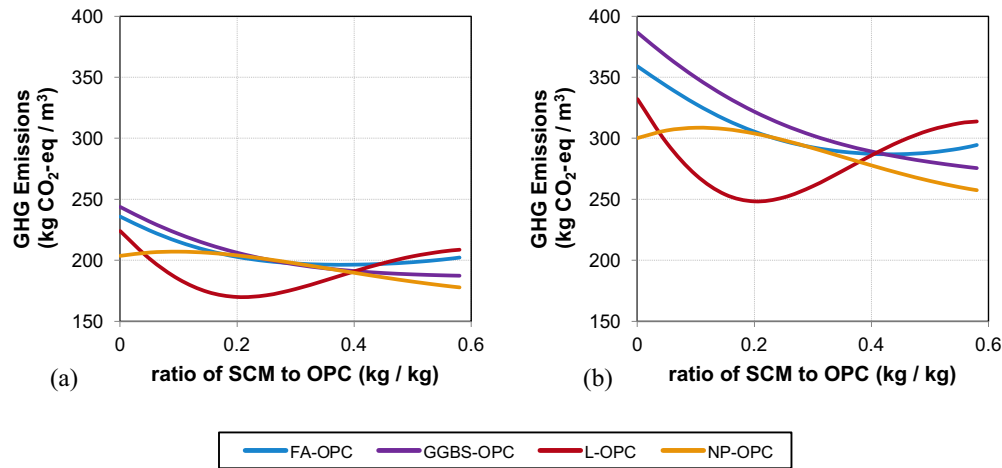
In the baseline GHG emissions calculations, GHG emissions for FA and GGBS were based solely on post-production processing requirements. In this second case, mass-based allocations considering production of FA and GGBS, as well as processing, are obtained through the procedures presented in Miller [39], which provides the GHG emissions formulas for these SCMs. Using these mass-based allocations in the assessment of emissions for FA and GGBS-OPC concrete mixtures, the parameters used in Eqs. (8)–(11) were recalculated and these new constants are listed in Table 4. Compared to the initial values of these constants, which are presented in Table 2, the constants  $k_A$ ,  $k_B$  and  $k_D$  are not changed while the constant  $k_C$  is increased approximately twofold for each FA and GGBS. These differences are because the changes are only applied to the GHG emissions associated with use of SCMs and the GHG emissions from the other concrete constituents production are not changed.

The use of allocation shifts the GHG emissions associated with use of FA and GGBS such that increased use of these materials may not be as desirable as the baseline scenario previously discussed. Noting the shift in GHG emissions relative to  $s/c$  ratio for the same concrete compressive strengths previously examined, it is seen that the use of allocation changes the concrete mixture proportions associated with the minimum GHG emissions (see Fig. 5). Specifically, the optimal ratio of  $s/c$  at which GHG emissions is a minimum drops for FA- and GGBS-OPC mixtures at all strengths considered. The decrease in  $s/c$  ratio ranges from approximately 8–23% for the FA-OPC mixtures and 18–48% in the GGBS-OPC mixtures; parallel to these changes, the GHG emissions at the optimal mixture proportions rises by approximately 6–7% for the FA-OPC mixtures and 8–12% for the GGBS-OPC mixtures. The optimal GHG emissions for each group of concrete mixtures including allo-

**Table 3**  
Optimal mixture proportions for specified concrete compressive strengths and the associated GHG emissions for production.

Compressive Strength (MPa)		15	20	25	30	35	40
FA-OPC	$s/c$ (kg/kg)	0.38	0.40	0.41	0.42	0.43	0.44
	$w/b$ (kg/kg)	0.98	0.85	0.76	0.68	0.61	0.56
	$i$ (kg CO <sub>2</sub> -eq/m <sup>3</sup> )	196	219	242	264	287	311
GGBS-OPC	$s/c$ (kg/kg)	0.96	1.08	1.13	1.16	1.19	1.20
	$w/b$ (kg/kg)	0.88	0.74	0.66	0.59	0.54	0.50
	$i$ (kg CO <sub>2</sub> -eq/m <sup>3</sup> )	186	205	224	242	260	278
L-OPC	$s/c$ (kg/kg)	0.04*	0.21	0.21	0.21	0.21	0.20
	$w/b$ (kg/kg)	1.00*	0.96	0.85	0.77	0.70	0.64
	$i$ (kg CO <sub>2</sub> -eq/m <sup>3</sup> )	207*	189	208	228	248	270
NP-OPC	$s/c$ (kg/kg)	0.74	0.73	0.71	0.70	0.69	0.69
	$w/b$ (kg/kg)	0.75	0.66	0.60	0.54	0.49	0.45
	$i$ (kg CO <sub>2</sub> -eq/m <sup>3</sup> )	173	193	212	232	253	274

\* Note: for this case, the mathematical models noted a minima would be expected at a higher  $s/c$  ratio and higher  $w/b$  ratio, but the data are presented considering a maximum  $w/b$  of 1.00.



**Fig. 3.** Comparison of GHG emissions per cubic meter of concrete as a function of s/c ratio for the groups of concrete examined for (a) 15 MPa compressive strength and (b) 35 MPa compressive strength.

cations can be found in Table 5. These shifts suggest that the method used to conduct the environmental impact assessments can be a strong driver in how constituents are selected and the method should be uniform and inclusive across platforms such that consistent decisions can be derived.

## 7. Discussion

### 7.1. Potential influence of optimal constituent selection: A California-based example

The role optimal constituent selection could have on GHG emissions can better be understood through a simplified case-study. The mixtures assessed in this research were modeled as being batched in the San Francisco Bay Area and, if used as a surrogate for approximate emissions for the production of concrete in California, production statistics can be applied to understand the potential mitigation of GHG emissions associated with improved concrete proportioning. Using statistics from the European Ready Mixed Concrete Organization for the quantity of each of four classes of concrete strength used in the United States (namely, <16 MPa, 16–20 MPa, 25–30 MPa, and  $\geq 35$  MPa) [43] and data for the quantities of FA, GGBS, limestone, and NP used in the United States (based on [44]), national statistics for concrete and SCM demand can be scaled to production in California, which is responsible for approximately 10% of the United States consumption [1]. From these statistics and the GHG emissions associated with producing concrete mixtures of varying strength and OPC replacement levels, a baseline for expected GHG emissions for 1 m<sup>3</sup> of concrete in California can be approximated. Further, by examining the optimal mixture proportions for each of the four OPC replacement materials considered in this research, the use of each of these materials as well as increased use of all of the materials at the same relative ratio of use to one another can be understood (see Fig. 6).

Based on the required strengths from national averages, increased use of most supplementary materials considered in this research result in reduced GHG emissions. Namely, increased use of GGBS, NP, and limestone result in reduction of GHG emissions by 4, 10, and 11% from concrete production, respectively. Using the same ratio of FA/GGBS/NP/limestone as is currently implemented, but applying the optimal amounts there is a decrease in GHG emissions from concrete production by 6%. It is interesting to note, due to different physiochemical properties of the alternative materials from OPC and their role in hydration reactions, many

of these material alternatives result in an increase mass of binder: FA-OPC – increase in mass by 16%; GGBS-OPC – increase in mass by 34%; NP-OPC – increase in mass by 18%; use of all materials at the same ratio as currently used – increase in mass by 12%.

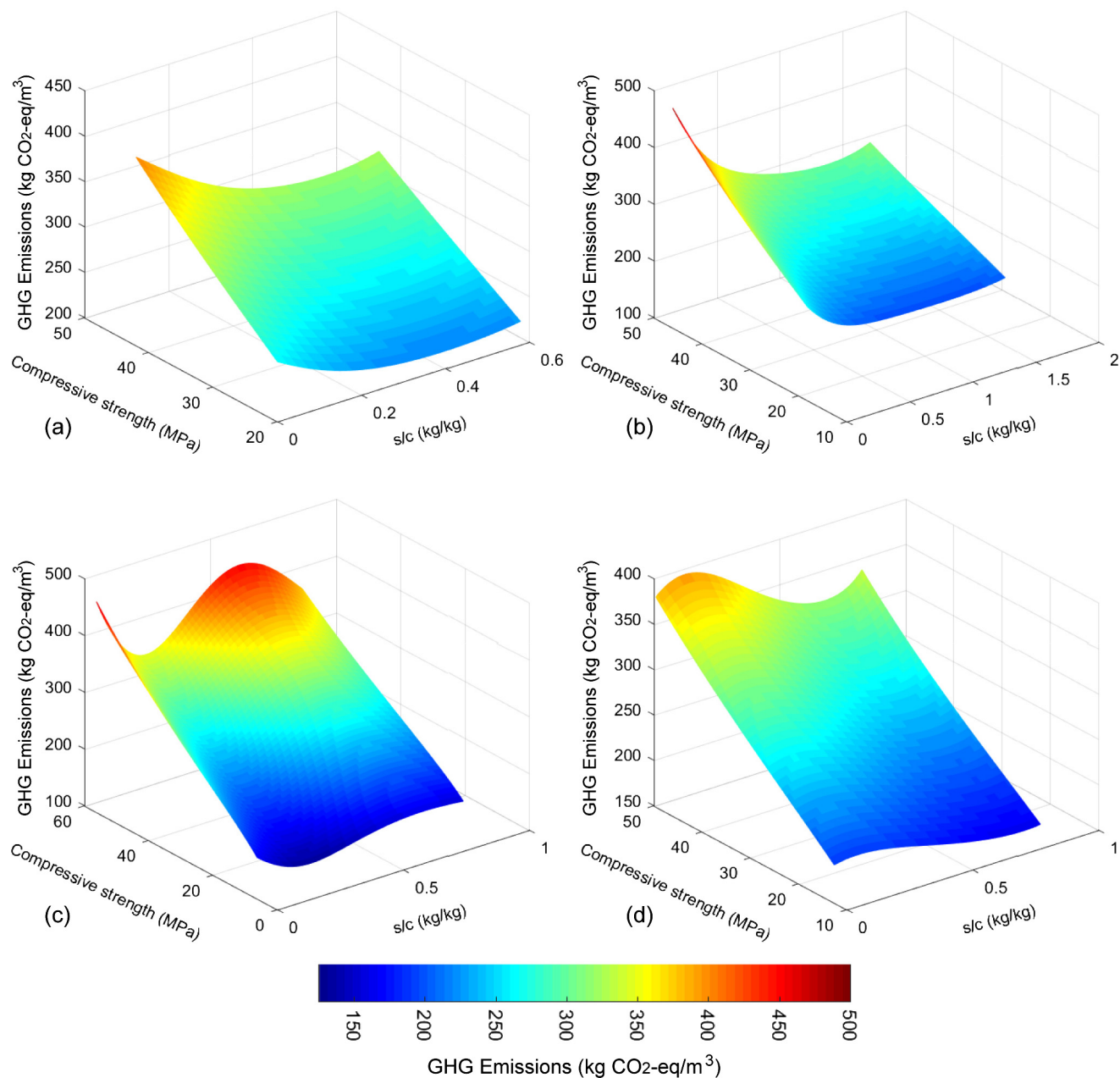
Considering that California used 7.4 Mt of hydraulic cement in 2012, which corresponds to approximately 24 m<sup>3</sup> of concrete (based on Miller et al. [5]), if we assume the production of concrete considered in this research is representative of California, then around 0.6 Mt of GHG emissions could be cut in the state. That is, through strategic use of the SCMs and fillers considered in this analysis 0.6% of industrial emissions and 0.13% of total GHG emissions for the state could be alleviated (based on comparisons to documented use [45]). Keeping in mind that the state reduced GHG emissions from 2000 to 2012 by 1.6% [45], the contribution that better proportioning concrete mixtures would have been approximately 8% of the reduction actually achieved. While the mixtures examined in this research scratch the surface of the thousands of potential mixtures that could be produced, the concept of how improved concrete mixture proportioning could have a strong influence is well exemplified in California.

### 7.2. Other parameters for consideration

While this work focused on GHG emissions and strength for concrete mixture design, other properties such as thermal properties, elastic moduli, and durability should be taken into account as well. Changes in material longevity can have a profound effect on the environmental impacts associated with the concrete structure [26]. Further, as has been shown by several authors (e.g., [24,31]) the specified design will drive the material property under consideration and the volume of material necessary to serve a given application. As such, in certain cases, if improved longevity or improvements to another material property are achieved, it may be desirable to sacrifice upfront GHG emissions for cumulative emission reductions (taking into account time-dependent factors associated with GHG emissions).

In cases where relationships between a small number of concrete constituents, environmental impacts, and material properties of interest can be drawn, as is the case for GHG emissions and compressive strength, the methods presented in this work can be applied. However, for more complex systems, methods need to be developed to facilitate balancing the other properties and environmental impacts in concrete mixture design. Additionally, consideration should be made for the role of both CO<sub>2</sub> emissions





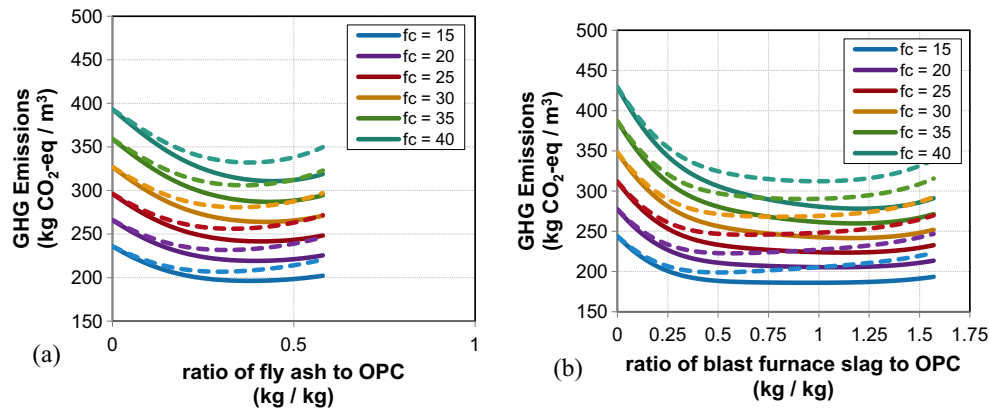
**Fig. 4.** 3-Dimensional plots of the relationship between strength, SCM to OPC ratio (*s/c*), and GHG emissions associated with the production of concrete for (a) FA-OPC concrete, (b) GGBS-OPC concrete, (c) L-OPC concrete, and (d) NP-OPC concrete mixtures.

**Table 4**  
Values of constants in each class of concrete by polynomial fitting for environmental impact parameters.

Concrete Type	FA-OPC	R <sup>2</sup>	GGBS-OPC	R <sup>2</sup>
<i>k<sub>A</sub></i>	0.96	1	0.96	1
<i>k<sub>B</sub></i>	27.2		34.3	
<i>k<sub>C</sub></i>	0.22	0.99	0.28	1
<i>k<sub>D</sub></i>	−0.17		0.19	

during the concrete production as well as CO<sub>2</sub> that is taken back up in the carbonation reaction of concrete structures (e.g., [46]). If durability of concrete structures were considered in concrete design, the calculation of GHG emissions for concrete would be revised as well. Additionally, this research focused on binary blends in the concrete binder and use of ternary or quaternary blends and/or chemical additives, fibers, and other additives could

be used to further enhance and alter properties. As such, further research should be conducted in the assessment of incorporating other material properties, durability, and other concrete constituents. Of course, another parameter for consideration would be the effects of mixture proportioning on financial costs. Due to varying costs associated with different concrete constituents often based on market and spatiotemporal parameters, a quantification



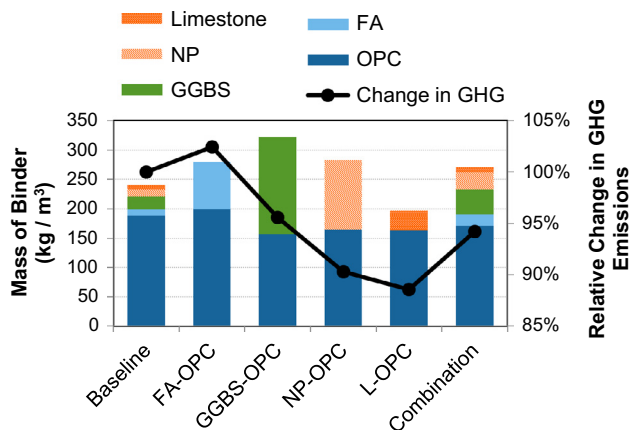
**Fig. 5.** Comparison of GHG emissions per cubic meter of concrete as a function of  $s/c$  ratio for six compressive strengths for (a) FA-OPC mixtures, (b) GGBS-OPC mixtures with and without allocation considered in calculation of GHG emissions, where dashed lines represent the use of allocation.

**Table 5**

Optimal mixture proportions for specified concrete compressive strengths when allocation is used in LCA and the associated GHG emissions for production.

Compressive Strength (MPa)		15	20	25	30	35	40
FA-OPC	$s/c$ (kg/kg)	0.35*	0.31	0.33	0.35	0.36	0.38
	$w/b$ (kg/kg)	1.00*	0.90	0.79	0.71	0.64	0.58
	$i$ (kg CO <sub>2</sub> -eq/m <sup>3</sup> )	207*	232	256	281	306	332
GGBS-OPC	$s/c$ (kg/kg)	0.64*	0.56	0.65	0.80	0.92	0.99
	$w/b$ (kg/kg)	1.00*	0.90	0.78	0.67	0.59	0.53
	$i$ (kg CO <sub>2</sub> -eq/m <sup>3</sup> )	200*	223	246	268	290	312

\* Note: for this case, the mathematical models noted a minima would be expected at a higher  $s/c$  ratio and higher  $w/b$  ratio, but the data are presented considering a maximum  $w/b$  of 1.00.



**Fig. 6.** Potential role of using optimal mixtures for reducing GHG emissions from concrete production.

of this nature was not included in this work; however, for individual cases in which costs are known, these parameters could also be incorporated.

## 8. Conclusions

This research presents equations for greenhouse gas emissions from the production of concrete as correlated to concrete mixture proportions and compressive strength. By applying the life cycle assessment approach, the calculation of greenhouse gas emissions per cubic meter of concrete mixtures are divided into two parts, one of which is caused by production of cement, and the other is attributed to supplementary cementitious materials. Some key findings from this work are:

- Higher compressive strength results in higher GHG emissions in each group of concrete mixtures in a specified  $s/c$ .
- The trends for GHG emissions with increased levels of FA or GGBS in the binder decreased with increasing SCM contents until reaching a local minima; yet, the inclusion of natural pozzolans did not follow the same patterns, suggesting properties of SCMs and concrete mixture proportions could be a strong driver in the role GHG emission reduction from increased SCM use.
- Within the scope of  $s/c$  examined and for the same compressive strength requirement, limestone filler can be used to lower 15–20% of GHG emissions from the production of concrete.
- The influence of environmental impact assessment scope, such as inclusion of allocation methods, can drive the optimal binder constituents for reduced environmental impact at different compressive strengths.

Depending on the scope of the analysis and production methods employed, the equations can be extended to other supplementary cementitious materials to find analytic solutions. Future directions for research should include development of equations to assess other material properties and the whole lifespan of concrete mixtures including service-life and end-of-life.

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## Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <https://doi.org/10.1016/j.conbuildmat.2018.02.092>.

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