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Optimizing supplementary cementitious material replacement to minimize the environmental impacts of concrete

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ABSTRACT

With growing environmental consequences from material consumption, there is increased urgency to decarbonize the production of materials we consume frequently, including concrete. It is common to use supplementary cementitious materials (SCMs) to limit the clinker content of Portland cement and reduce greenhouse gas (GHG) emissions in concrete production. However, over-utilization of SCMs can degrade material performance and increase other environmental impacts. Here we derive quantitative methods to determine the optimal SCM to Ordinary Portland Cement (OPC) ratio (s/c) for 7 different SCMs to minimize 11 environmental impact categories while maintaining compressive strength. We find that optimal replacement levels are heavily dependent on the SCM. Notably, lower s/c in many cases lowered emissions (e.g., s/c of 0.17 kg/kg for limestone leads to \sim 1.6x lower GHG emissions than a s/c of 0.42 kg/kg in 30 MPa concrete). This work demonstrates a systematic means to effectively utilize limited SCM resources to mitigate environmental impacts from concrete production.

1. Introduction

Unprecedented demand for construction material causes has caused environmental burdens [1-3]. Concrete and other cement-based materials have recently been under scrutiny for their contribution to anthropogenic greenhouse gas (GHG) emissions [4,5]. The notable GHG emissions from concrete production are a function of enormous amounts of concrete consumed annually and are the primary focus of environmental research on concrete [6]. Yet, the large consumption of concrete also contributes significantly to other anthropogenic environmental emissions including: ~8% of nitrogen oxides emissions [7], ~9% of mercury emissions [8], \sim 5% of sulfur oxide emissions [7], \sim 8% of CO₂ emissions [9], and \sim 5% of particulate matter smaller than 10 μ m (PM₁₀) emissions [7]. With consumption of concrete expected to grow by \sim 23% by 2050 [10], emissions from concrete production will continue increasing. In this work, we demonstrate an approach to mixture design using performance metrics and environmental impacts to identify optimal SCM content.

Anthropogenic GHG emissions contribute to climate change impacts with global implications [11], and mitigating these impacts is important. However, regional environmental impacts, beyond those from

GHG emissions, are less-often considered in evaluating the sustainability of concrete [12]. These other categories typically have more localized impacts to the environment and human health, and they can acutely impact minority and lower income populations [13-15]. Already over 80% of global populations live in areas where particulate matter (PM) emissions exceed World Health Organization guidelines [16], and exposure to elevated PM emissions in communities near cement production facilities has led to adverse respiratory effects and lower lung function [17]. Like PM emissions, heavy metal exposure from cement production can lead to adverse health effects in exposed populations [18]. Heavy metals from cement production may include toxins that pose significant health hazards to those exposed to them [18,19]. Water scarcity, while not an environmental emission, is also of growing concern globally, in that 75% of water demand is expected in regions that will experience water scarcity by 2025 [20]. The non-GHG emissions and water use from concrete production continues to be an understudied aspect of the regional sustainability of concrete. These impacts should be considered in sustainable concrete mixture design to avoid unintended consequences from alternative materials.

The use of supplementary cementitious materials (SCMs) is commonplace in the concrete industry. SCMs can both reduce GHG

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emissions, by reducing cement clinker content [21,22], and can contribute to improved strength and durability properties of concretes [23–25]. Improved strength can decrease impacts by reducing the required concrete volume in a structure [26]. Likewise, increased durability can improve the longevity of a structure and can also lead to reduced environmental impacts [27]. Many SCMs are often byproducts of other industries (e.g., fly ash from coal combustion, ground blast furnace slag from pig iron production), and thus their supply is restricted by the demand for the primary product [28]. This constricted supply has led to regional scarcity of some SCMs [29–31] and necessitates the efficient and effective use of SCMs to maximize environmental impact reductions, even with limited supply [32].

Traditionally, optimizing SCM contents to use SCMs most efficiently has not focused on the environmental impacts, and when environmental impacts are used, the focus has been predominantly on GHG emissions. Conventional SCM optimization is largely dependent on material performance indicators [33]. Quantifying the effects of SCM to cement ratio allows for selecting mixtures the best mitigate negative environmental impacts. Tushar et al. [34] considered mechanical performance and a harmonized sustainability metric to identify the optimal fly ash and ground granulated blast furnace slag (GGBS) to decrease environmental damage. Naseri et al. [35] also applied sustainability criterion for selecting mixtures of fly ash, silica fume, and GGBS that are machine-optimized using material performance models. While combined impact categories simplifies such calculations, it could obscure the effects of specific impact categories, such as localized impacts to populations near production areas. When considering GHG emissions per unit-volume for concretes with SCM replacement, Fan and Miller [36] show that optimal replacement levels are less than the maximum replacement. Similar conclusions have been drawn in other work showing excessive SCM replacement does not necessarily minimize GHG emissions [32]. These trends occur because of trade-offs that occur in material property development and factors driving environmental impacts. On a mass-basis, a replacement of a high Portland clinker cement with low GHG-emitting mineral additives will reduce the GHG emissions of the blend. However, if those mineral additives do not contribute to strength or other performance characteristics beyond a certain replacement level, then to achieve desired performance, more Portland clinker may be needed, thus limiting the efficacy of the environmental benefits from replacement. Further analysis of optimal SCM replacement that considers additional impact categories and SCMs is necessary for a holistic approach to sustainability that avoids unintended environmental impacts.

While the emphasis on sustainable materials is growing, there is still always a need to ensure safety, strength, and durability when it comes to structural materials. In this work we address how mechanical performance and multiple environmental impact categories can be concurrently assessed to reduce the environmental impacts of structures during the design of concrete materials. Process-based lifecycle assessment (LCA) methodologies are commonly used to quantify the environmental impacts from concrete and cement production [37]. This study uses the OpenConcrete Tool [12] for concrete constituent life cycle inventories and impact modeling. This tool allows users to provide information about a concrete mixture, energy sources, and transportation to quantify the cradle-to-gate environmental impacts across multiple impact categories.

Herein, we demonstrate a method for identifying the optimal SCM content by evaluating concrete mixtures with one of seven different SCM types and quantifying the environmental impacts for 11 different impact categories: GHG emissions, nitrogen oxides (NO_X) emissions, sulfur oxides (SO_X), particulate matter under 10 μm in diameter (PM₁₀) emissions, particulate matter under 2.5 μm in diamete (PM_{2.5}) emissions, volatile organic compound (VOC) emissions, carbon monoxide (CO) emissions, lead (Pb) emissions, water consumption, water withdrawal, and energy demand (MJ). Using compressive strength data as an indicator for material performance, we identify how the optimal SCM to

cement ratio can be determined to minimize environmental impacts while still meeting the compressive strength requirements for concrete.

2. Materials and methods

2.1. Concrete mixtures

To implement the optimization methods derived in this work, data were collected from the literature. To streamline calculations, we selected concrete mixtures containing binary blended cements, focusing on Ordinary Portland Cement (OPC) alone or with one of seven different SCMs: limestone filler (LS), natural pozzolans (NP), shale ash (SA), calcined clay (here, examining metakaolin (MK)), silica fume (SF), fly ash (FA), and ground granulated blast furnace slag (GGBS). The mixtures selected ranged in 28-day strength from 6 to 80 MPa, which was used to facilitate assessment of the role of varying optimal mineral additive use to minimize environmental impacts and to achieve different strengths. To limit variability in inputs, we focused on mixtures and properties where water contents were held relatively constant for varying concrete compressive strengths (see summary Table 1). For the purposes of this research, a fixed water content was assumed; in cases where there was minor variability in water content from the literature used, average water contents were implemented. Further, in mixtures utilizing plasticizers/superplasticizers, average quantities were used in equations developed. These concrete mixtures were used to represent the linkage between SCM mixture proportions, compressive strengths, and environmental impacts; however, we note that the equations derived can be applied more broadly.

To utilize these concrete mixtures in environmental impact comparisons, unit conversions were needed to compare kg of constituents required per m³ of concrete produced. Here, we assumed the following densities for each material constituent (Table 2).

The complete table of all the scaled mixtures proportions and compressive strengths are found in Supplemental Materials 1.

2.2. Environmental impacts

Cradle-to-gate assessments of 11 environmental impacts (emissions of GHGs, NO_X, SO_X, PM₁₀, PM_{2.5}, VOCs, and Pb, as well as water consumption, water withdrawal, and energy demand) were performed using the OpenConcrete tool [12], as noted previously. To guide inputs into this tool, we model the production of concrete in the San Francisco Bay Area of California. OPC, NP, MK, and LS were all assumed to be produced in California. Because of negligible coal electricity generation in California, FA was assumed to be produced in Wyoming. Similarly, SA was assumed to be produced in Wyoming. SF and GGBS were produced in Pennsylvania. FA and GGBS are byproducts of other industries; here we consider no allocation of impacts from the primary products to impacts resulting from their initial production. However, any additional processing and transportation needed after initial generation of these byproducts for their use in concrete was considered. Fine and coarse aggregate were assumed to be quarried in California. All constituents assumed to be manufactured in California utilized the California average electricity mix (note: we use electricity inputs by US State as available in OpenConcrete [12]). The transportation distances are based on a report

Table 1Concrete mixture proportions and strength sources.

| Supplementary cementitious material | Source | | | |
|---|---------------------|--|--|--|
| Limestone filler (LS) | Meddah et al. [39] | | | |
| Natural pozzolans (NP) | Meddah et al. [40] | | | |
| Shale ash (SA) | Meddah et al. [40] | | | |
| Metakaolin (MK) | Meddah et al. [41] | | | |
| Silica fume (SF) | Meddah et al. [41] | | | |
| Fly ash (FA) | Oner et al. [42] | | | |
| Ground granulated blast furnace slag (GGBS) | Oner and Akyuz [43] | | | |

Table 2Concrete constituent densities (based on [39–43]).

| Constituent | Density (kg/m³) |
|--------------------------------------|-----------------|
| Ordinary Portland Cement | 3140 |
| Limestone | 2700 |
| Natural Pozzolans | 2480 |
| Shale Ash | 2670 |
| Calcined Clay | 2590 |
| Silica Fume | 2200 |
| Fly Ash | 2090 |
| Ground Granulated Blast Furnace Slag | 2870 |
| Fine Aggregates | 2680 |
| Coarse Aggregates | 2700 |
| Superplasticizer | 1006 |
| Water | 997 |

from the Portland Cement Association, which reports the distances resources are moved for concrete production [44]. Namely, over 95% of cement and SCMs are moved approximately 150 km in the United States, so we modeled PC, LS, NP, and MK as being transported this distance. As described above, SA, SF, FA, and GGBS were produced outside of California, and the transportation distance used in this model for each was 2000 km by train. To capture the approximate distance for superplasticizers, we modeled the distance from the location where a significant fraction of petroleum and chemical processing occurs in the United States (the Gulf of Mexico) to the location where these materials were being batched (California), which is approximately 3000 km. Transportation of fine aggregate and coarse aggregate was assumed to be 50 km by truck [44]. OpenConcrete facilitates data retrieval for both concrete mixtures as well as for individual constituents, and in this work, we utilize the outputs of impact per constituent to model the role of changing SCM content.

3. Theory

3.1. Relating mixture proportions and environmental impacts

To derive functions to relate the mixture proportions and environmental impacts, we first determine the linear relationship between the volume of SCM, the volume of cement, and the volume of fine aggregates; namely, an equation for the linear correlation between the increase in SCM content and the corresponding decrease in fine aggregate was determined for each SCM type. A linear relationship also exists between the volume of coarse aggregate and the rest of the constituents. These relationships were used in a system of equations to relate mixture proportion inputs necessary for both material strength (as stipulated below) and for environmental impact assessments. To determine environmental impacts, relative weights of constituents per cubic meter of concrete were used (leveraging densities in Table 2) and factored by emissions per kg constituent (as discussed in the prior section).

3.2. Compressive strength of the concrete mixtures

Expanding from a methodology introduced by Fan and Miller [36], the quantitative optimization method begins with Abram's law (Eq. (1)) in order to relate compressive strength to the concrete constituents. Abram's law gives the following relationship:

$$f_c = \frac{k_1}{k_2^{w/b}}$$
 Eq. 1

where f_c is compressive strength, w/b is the water to binder ratio, and k_1 and k_2 are constants that are determined by fitting to experimental data. Here, we use k_1 and k_2 as defined by Fan and Miller [36], namely, with the following equations:

$$k_1 = \alpha(s/c)^3 + \beta_1(s/c)^2 + \Upsilon_1(s/c) + \varsigma_1$$
 Eq. 2

$$k_2 = \beta_2 (s/c)^2 + 2(s/c) + \varsigma_2$$
 Eq. 3

where α , β_1 , β_2 , Υ_1 , Υ_2 , ς_1 , ς_2 are empirically derived constants that were determined from the datasets discussed in Section 2.1. These relationships used that allow k_1 and k_2 to be defined as functions of the SCM to OPC ratio (s/c). From Eq. (1), binder, b, can be defined as s + c in terms of content by weight (kg/m³). Using this definition of the cementitious binder and the prior equations, f_c can be defined as:

$$f_c = \frac{\alpha(s/c)^3 + \beta_1(s/c)^2 + \gamma_1(s/c) + \varsigma_1}{\left(\beta_2(s/c)^2 + 2(s/c) + \varsigma_2\right)^{\frac{w}{s+c}}}$$
 Eq. 4

Again, using the cementitious binder defined as s+c, Abram's law (Eq. (1)) can be written as:

$$\frac{\ln(f_c) - \ln(k_1)}{\ln(k_2)} = \frac{w}{s + c}$$
 Eq. 5

Here, this rewriting of Abram's law facilitates an additional simplification as the concrete mixtures used in this work have constant or near constant water content. Namely, in order to change the water-to-binder ratio for these mixtures, the cementitious binder content must change. As a result, we can define the mass of s and c as functions of the fitting parameters used to determine strength, the average water content, and the concrete compressive strength:

$$s = \frac{w}{\frac{\ln(f_c) - \ln(k_1)}{\ln(f_c)}} - c$$
 Eq. 6

$$c = \frac{w}{\frac{\ln(f_c) - \ln(k_1)}{\ln(k_2)}} - s$$
 Eq. 7

As such, these relationships allow us to determine the SCM content and cement content for a known water content. To address aggregate content, a linear relationship between the volume of $\mathbf{s}+\mathbf{c}$ and the volume of fine aggregate was determined. As volumes were needed to assess the quantity of material replaced, we used a functional unit of comparison of 1 m³ and the densities of constituents presented in Table 2. With these parameters, we define the volume of the fine aggregate as:

Volume of Fine Aggregate =
$$\lambda_1(Volume \ of \ s+c) + \varphi_1$$
 Eq. 8

where λ_I and φ_I are empirically derived constants from the datasets used that allow the volume of fine aggregate to be determined based on the relationship to the volume of SCM plus OPC (s + c), and all other terms are as previously defined. Similar to Equation (8), the volume of coarse aggregate can be determined using the linear relationship between the volume of coarse aggregate and the volume of all other constituents in the concrete mixture:

Volume of Coarse Aggregate =
$$\lambda_2(Volume \ of \ Constituents) + \varphi_2$$
 Eq. 9

where λ_2 and ϕ_2 are empirically derived constants from the datasets used that allow the volume of coarse aggregate to be determined based on the relationship to the volume of all other constituents (fine aggregate, water, plasticizer, and SCM plus OPC (s + c)), and all other terms are as previously defined and all other terms are as previously defined. In this work, to exemplify the application of these equations, we leverage the mixtures discussed in the prior section. The parameters determined for each SCM type are presented in Tables 3 and 4.

3.3. Environmental impacts

Using the equations derived in Section 3.2, the mass of each constituent can be specified for a set volume of concrete mixture with a specified strength, given some experimental data to determine fitting parameters and constituent densities. Using these constituent masses,

Table 3
Coefficients for Eqs. (2) and (3)

| 28 Day Strength | | | | | | | | | | | |
|-----------------|---------|----------|--------|--------|----------------|--------|--------|-------|----------------|--|--|
| | k1 | | | | | k2 | | | | | |
| SCM | α | β1 | Υ1 | ς1 | R ² | β2 | Υ2 | ς2 | R ² | | |
| LS | 412.73 | -340.11 | 39.55 | 140.70 | 0.9823 | 138.18 | -46.31 | 12.04 | 0.9740 | | |
| NP | 749.69 | -1203.50 | 550.62 | 70.69 | 0.3400 | -26.83 | 43.73 | 4.99 | 0.8445 | | |
| SA | 228.60 | -259.92 | 48.13 | 140.64 | 0.9533 | 2.11 | 9.17 | 9.53 | 0.9877 | | |
| MK | 495.68 | -1056.99 | 526.62 | 142.43 | 0.9891 | -56.24 | 34.01 | 9.86 | 0.9891 | | |
| SF | 7568.93 | -4229.90 | 883.78 | 141.00 | 0.9985 | -66.43 | 27.41 | 9.90 | 0.9452 | | |
| FA | -693.69 | 634.53 | -90.60 | 137.50 | 0.9825 | 47.17 | -8.78 | 8.30 | 0.9977 | | |
| GGBS | -79.27 | 166.27 | -20.11 | 113.25 | 0.9946 | 18.49 | -5.90 | 6.78 | 0.9998 | | |
| 60 Day Str | ength | | | | | | | | | | |
| | k1 | | | | | k2 | | | | | |
| SCM | α | β1 | Υ1 | ۲1 | R^2 | β2 | Υ2 | ς2 | \mathbb{R}^2 | | |
| LS | 270.86 | -161.39 | -8.29 | 141.46 | 0.9862 | 131.76 | -43.12 | 10.37 | 0.9795 | | |
| NP | 794.93 | -1238.95 | 555.60 | 71.04 | 0.3438 | -21.08 | 35.34 | 4.30 | 0.8329 | | |
| SA | 300.15 | -322.37 | 77.62 | 141.07 | 0.6961 | 3.04 | 7.85 | 8.28 | 0.9865 | | |
| MK | 654.73 | -1122.08 | 523.30 | 143.11 | 0.9902 | -48.74 | 29.66 | 8.51 | 0.9825 | | |
| SF | 5712.97 | -3733.07 | 815.67 | 141.63 | 0.9968 | -78.19 | 26.39 | 8.49 | 0.9604 | | |
| FA | -669.42 | 622.30 | -85.85 | 142.43 | 0.9889 | 48.49 | -9.39 | 8.36 | 0.9984 | | |
| GGBS | -96.18 | 199.35 | -30.75 | 115.35 | 0.9756 | 12.58 | -3.23 | 6.38 | 0.9968 | | |
| 180 Day St | rength | | | | | | | | | | |
| | k1 | | | | | k2 | | | | | |
| SCM | α | β1 | Υ1 | ۲1 | \mathbb{R}^2 | β2 | Υ2 | ς2 | \mathbb{R}^2 | | |
| LS | 311.36 | -270.43 | 32.15 | 148.19 | 0.9896 | 96.53 | -25.26 | 9.29 | 0.9836 | | |
| NP | 1131.46 | -1595.79 | 628.52 | 74.25 | 0.3227 | -14.10 | 24.05 | 4.22 | 0.7163 | | |
| SA | -65.61 | -18.28 | 30.16 | 147.95 | 0.9431 | -2.86 | 10.37 | 7.57 | 0.9777 | | |
| MK | -878.26 | -278.29 | 395.21 | 147.84 | 0.9811 | -44.48 | 29.14 | 7.69 | 0.9732 | | |
| SF | 2082.82 | -2150.72 | 641.21 | 147.91 | 0.9945 | -46.03 | 19.00 | 7.89 | 0.9895 | | |
| FA | -577.32 | 575.73 | -67.96 | 160.91 | 0.9974 | 52.66 | -11.32 | 8.52 | 0.9992 | | |
| GGBS | -49.97 | 96.77 | 33.26 | 126.96 | 0.9920 | 13.72 | -6.06 | 7.21 | 0.9959 | | |

Table 4Coefficients for fine and coarse aggregate equations.

| | Fine Aggre | egate | | Coarse Aggregate | | | | |
|------|-------------|----------------|--|------------------|----------------|---|--|--|
| SCM | λ_1 | φ ₁ | $\begin{array}{c} \phi_1 & \text{Vol of Fine} \\ \text{Agg-R}^2 \end{array}$ | | φ ₂ | Vol of Coarse Agg- R ² | | |
| LS | -0.6135 | 0.3171 | 0.7127 | -0.0069 | 0.4482 | 0.0000 | | |
| NP | -0.6337 | 0.3185 | 0.7338 | -0.0171 | 0.4538 | 0.0003 | | |
| SA | -0.6118 | 0.3171 | 0.7100 | -0.0216 | 0.4562 | 0.0005 | | |
| MK | -0.6054 | 0.3168 | 0.7022 | -0.0223 | 0.4566 | 0.0006 | | |
| SF | -0.6175 | 0.3174 | 0.7168 | -0.0365 | 0.4643 | 0.0016 | | |
| FA | -0.9508 | 0.5557 | 0.9980 | -1.0250 | 1.0029 | 0.9899 | | |
| GGBS | -0.5726 | 0.3201 | 0.9985 | -0.9962 | 0.9825 | 1.0000 | | |

environmental impacts can be determined by using cradle-to-gate inputs for constituents and batching.

$$I = i_b + \sum_{i} \left(m_j \times i_j \right)$$
 Eq. 10

where I is the environmental impact per unit volume of the concrete mixture, i_b is the environmental impact from batching the mixture, m is the mass of each constituent, i is the environmental impact of each constituent (including requisite transportation), and j refers to the constituents used in the mixture. Namely, here we use inputs for the impacts from batching and for each of the concrete constituents from OpenConcrete, and we are able to define 11 environmental impacts for concrete mixtures. While there are many ways to implement the series of equations defined, we use them to perform multi-objective optimization. Namely, for each type of SCM studied, we examine what replacement rate would concurrently drive down each of the 11 environmental impacts for a series of specified compressive strengths (for all SCMs and strengths, environmental impacts were plotted against s/c ratios see Supplemental Materials 3). The partial derivative of each function of environmental impact vs s/c ratio was taken to find the optimal s/c ratio

that lowers each impact for each strength and SCM mixture.

Optimal
$$s / c$$
 ratio = $\frac{df}{ds/c} = \lim_{h \to 0} \frac{f(s/c + h, I) - f(s/c, I)}{h}$ Eq. 11

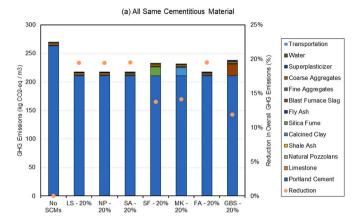
where f is the calculated function of I as a relation to s/c ratio, h is representative of the slope of the changing function, and all other variables are defined previously. For the examples we use in this work, the calculated impacts are limited by the s/c ratio experimental values such that strength and impacts could be compared to input values. The optimal s/c ratios can be found in Supplemental Materials 2.

4. Results

4.1. Environmental impacts of concrete mixtures

Conventionally, increased utilization of SCMs is presumed to lower GHG emissions from concrete mixtures. This assumes that reducing clinker content in concrete will inherently lower its environmental burdens [45]. While on a per volume basis, this holds true since a reduction in the clinker content, the primary contributor to GHG emissions, would lower GHG emissions, but when material performance is integrated, these results can shift [46]. A loss in desired properties, such as compressive strength, can outweigh benefits from reduced emissions per unit volume of concrete if more concrete must be used to overcome the loss in performance [47].

Our initial environmental impact results reflect these points. In Fig. 1 (part a), we show GHG emissions per cubic meter of concrete for simulated mixtures containing the same cementitious content, but with either no SCM use or with 20% OPC replacement using each of the 7 SCMs considered in this work (note: these are reflective of s/c ratios ranging from 0.17 to 0.23, a function of the differences in SCM densities). Herein, emissions from transportation and batching are considered. Our results indicate that up to 20% of emissions could be reduced by use of SCMs when assessing concrete on a per volume basis



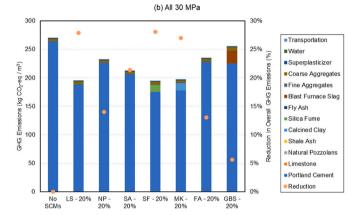


Fig. 1. Comparison of GHG emissions per cubic meter of concrete for simulated mixtures using models derived for (a) the same cementitious content per unit volume and for (b) the same strength concrete mixtures (30 MPa). (Note: these are reflective of s/c ratios ranging from 0.17 to 0.23, reflecting the differences in SCM densities).

(reduction ranges from 12% to 20%, with the lowest reduction from use of GGBS and the greatest reduction from use of FA and SA). Similar trends in environmental impact reductions are noted for Pb, VOC, SO_X , and NO_X emissions, which result from these emissions being primarily driven by clinker production (see Supplemental Materials 3). Namely, the resources used for the thermal energy used in the cement kilns lead to the majority of these emissions. However, these trends are not consistent for $PM_{2.5}$, PM_{10} , water consumption, or water withdrawal. This lack of consistency is a function of other concrete constituents contributing notably to these environmental impacts; as a result, a reduction of OPC, which inherently is replaced by another material for a unit volume to remain consistent, does not necessarily result in reduced impact for these other categories.

When addressing material strength (Fig. 1, part b), we show concrete mixtures modeled with the same SCM replacement ratio (20%) as well as a mixture with no SCM replacement. Here, however, we use our models to simulate each mixture as achieving the same compressive strength, 30 MPa, instead of the same cementitious content. While the use of SCMs still reduces GHG emissions at this replacement level, we now see that reductions range from 6% to 28%, with the lowest reduction from use of GGBS and the greatest reduction from use of LS and SF. While clinker is still the primary driver of GHG emissions, here the inclusion of strength in the comparisons reflects how a reduction in this binder could compromise performance. For the mixtures we simulate in this work, water content is held constant. As such, to overcome lower strength, more cementitious material would be needed, thus increasing the OPC content. This increase can in turn limit the benefits that would have been noted on a per-unit-volume basis.

It should be noted that the experimental data for FA and GGBS was taken from a different set of authors than the other SCMs, and as a result, are not as directly comparable. We were unable to find this set of SCMs tested by exclusively one author, but these two sets of authors performed similar experimental assessments and held relatively consistent water content while varying cementitious content to change compressive strength. For the FA and GGBS mixtures, the experimental data varied constituents such as coarse aggregate and, to a much lesser extent, water; these two constituents remained constant with other SCM mixtures. In order to utilize the calculations for this study, an average water amount was applied. The variability contributed to higher coefficients of determination among the relationships between the volume of fine aggregate to binder and volume constituents to volume of coarse aggregate. Also, the data for the FA and GGBS mixtures used higher cementitious and water contents than the authors presenting the other concrete mixtures. As such, comparisons should not be drawn across these different SCMs. However, these results show how methods we derived can be applied across multiple materials to determine desired mixture proportions to mitigate environmental impacts.

4.2. Optimizing mixture proportions to reduce environmental impacts

The primary goal of this work was to derive equations that would facilitate determining the optimal s/c mass ratio to minimize environmental impacts for a specified concrete strength. The environmental impacts calculated through the equations above are plotted relative to the s/c ratio for each SCM. Here, we note that rate of strength development can vary with SCMs, so these values were plotted for each impact at 28 d, 60 d, and 180 d compressive strengths (additional plots are shown in Supplemental Materials 3). The optimal s/c ratio for compressive strengths ranging from 15 MPa to 45 MPa to minimize each of the 11 environmental impact categories are shown in Fig. 2 (note: this figure presents 28 d strength. 60 d and 180 d strengths are presented in Supplemental Materials 3).

The greatest reduction in environmental impacts possible through optimization of SCM replacement rate varies by SCM type and by environmental impact, the latter of which is a function of the varying drivers in environmental impacts as noted in the previous section.

For LS, the optimal s/c ratio ranges between 0.13 and 0.21 (i.e., a replacement rate of 12-18%). Lower GHG emissions, SO_X, NO_X, VOC, CO, and Pb emissions occur at higher replacement rates, as does energy demand. However, lower PM₁₀ and PM_{2.5} emissions occur at the lower end of this ratio range. Minimizing water consumption and withdrawal occurs at an s/c ratio of ~0.18 at 28 d strength. For many of the environmental impact categories, desired s/c varies with the age of concrete, where if strength development is allowed to occur, a lower s/c ratio is desired (~0.18 at 28 d and ~0.15 at 180 d). For many of the impact categories examined, at higher specified strength, a lower s/c ratio may be desired to mitigate emissions, but over 0.13 s/c consistently reduced impacts relative to a pure OPC binder. For the mixtures simulated here to conduct optimization, the lowest GHG emissions achieved using LS resulted in a reduction of 27% relative to the mixture with no LS, occurring at an s/c of 0.21. This s/c ratio results in a 2%-28% reduction of the other environmental impact categories examined.

When NPs as an SCM were examined, there was a more consistent optimal s/c ratio to lower multiple environmental impacts. The optimal s/c ratio to lower GHG, NO_X , SO_X , VOC, CO, Pb, and energy demand was 0.82 (kg/kg) and was constant across all strengths and testing age. The optimal ratios for $PM_{2.5}$, PM_{10} , water withdrawal, and water consumption vary, but the optimal ratio increased as strength increased. For PM_{10} emissions, this ratio ranged between 0 and 0.25 kg/kg. The optimal s/c ratio for $PM_{2.5}$ emissions ranged between 0 and 0.22 kg/kg. For water consumption and water withdrawal, the optimal s/c ratio ranged between 0 and 0.27 kg/kg and 0–0.29, respectively. Fig. 3 shows the environmental impacts as a function of s/c for several specified concrete strengths at 28 d and varying local minima in impacts achieved. For the

| | | 28 days | | | | | | |
|------|--|---------|--------|--------|--------|--------|--------|--------|
| | | 15 MPa | 20 MPa | 25 MPa | 30 MPa | 35 MPa | 40 MPa | 45 MPa |
| LS | GHG/NOX/SOX/VOC/CO/Pb/Energy Demand | 0.21 | 0.21 | 0.21 | 0.21 | 0.20 | 0.20 | 0.20 |
| | PM10 | 0.18 | 0.18 | 0.18 | 0.17 | 0.17 | 0.17 | 0.17 |
| | PM2.5 | 0.17 | 0.17 | 0.16 | 0.16 | 0.16 | 0.16 | 0.16 |
| | Water Consumption | 0.18 | 0.18 | 0.18 | 0.18 | 0.18 | 0.18 | 0.18 |
| | Water Withdrawal | 0.19 | 0.19 | 0.19 | 0.19 | 0.18 | 0.18 | 0.18 |
| | GHG/NOX/SOX/VOC/CO/Pb/Energy Demand | 0.82 | 0.82 | 0.82 | 0.82 | 0.82 | 0.82 | 0.82 |
| | PM10 | 0.00 | 0.12 | 0.10 | 0.21 | 0.22 | 0.24 | 0.25 |
| IP. | PM10 PM2.5 | 0.00 | 0.13 | 0.18 | | 0.23 | 0.24 | |
| | Water Consumption | 0.00 | 0.07 | 0.12 | 0.16 | 0.18 | 0.20 | 0.22 |
| | Water Withdrawal | | 0.18 | 0.22 | 0.24 | 0.25 | | |
| | GHG/NOX/SOX/PM10/PM2.5/VOC/ | 0.17 | 0.23 | 0.25 | 0.27 | 0.28 | 0.28 | 0.29 |
| A | CO/Pb/Enery Demand | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 | 0.67 |
| A. | Water Consumption | 0.04 | 0.04 | 0.05 | 0.06 | 0.06 | 0.06 | 0.07 |
| | Water Withdrawal | 0.67 | 0.67 | 0.67 | 0.30 | 0.28 | 0.26 | 0.24 |
| | GHG/SOX/NOX/VOC/CO/Pb/Water Consumption/Water | 0.33 | 0.33 | 0.33 | 0.33 | 0.33 | 0.33 | 0.33 |
| ик | Withdraw al/Enery Demand | 0.55 | 0.55 | 0.55 | 0.55 | 0.55 | 0.55 | 0.55 |
| K | PM10 | 0.00 | 0.00 | 0.04 | 0.09 | 0.12 | 0.15 | 0.17 |
| | PM2.5 | 0.00 | 0.00 | 0.00 | 0.03 | 0.06 | 0.09 | 0.11 |
| SF | GHG/SOX/PM10/PM2.5/VOC/CO/Pb /Water Consumption/Energy Demand | | 0.25 | 0.25 | 0.25 | 0.25 | 0.25 | 0.25 |
| | NOX | 0.06 | 0.07 | 0.09 | 0.10 | 0.25 | 0.25 | 0.25 |
| | Water Withdrawal | 0.00 | 0.00 | 0.01 | 0.02 | 0.03 | 0.03 | 0.04 |
| | GHG/NOX/SOX | 0.40 | 0.42 | 0.43 | 0.44 | 0.45 | 0.46 | 0.47 |
| | PM10 | 0.42 | 0.44 | 0.45 | 0.46 | 0.46 | 0.47 | 0.48 |
| | PM2.5 | 0.43 | 0.44 | 0.45 | 0.46 | 0.47 | 0.48 | 0.48 |
| A | VOC/CO/Pb | 0.40 | 0.42 | 0.43 | 0.44 | 0.45 | 0.46 | 0.47 |
| | Water Consumption | 0.19 | 0.20 | 0.21 | 0.23 | 0.24 | 0.26 | 0.28 |
| | Water Withdrawal | 0.31 | 0.32 | 0.34 | 0.36 | 0.38 | 0.39 | 0.40 |
| | Energy Demand | 0.35 | 0.37 | 0.39 | 0.40 | 0.42 | 0.43 | 0.44 |
| | GHG | 0.42 | 0.46 | 0.50 | 0.57 | 0.68 | 0.81 | 0.90 |
| | NOX | 0.05 | 0.04 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| GGBS | SOX | 0.44 | 0.48 | 0.54 | 0.63 | 0.76 | 0.88 | 0.96 |
| | PM10/PM2.5 | 1.57 | 1.57 | 1.57 | 1.57 | 1.57 | 1.57 | 1.57 |
| | VOC | 0.32 | 0.35 | 0.37 | 0.41 | 0.45 | 0.51 | 0.60 |
| | со | 0.69 | 0.94 | 1.04 | 1.10 | 1.13 | 1.16 | 1.18 |
| | Pb | 1.41 | 1.40 | 1.39 | 1.38 | 1.38 | 1.38 | 1.37 |
| | Water Consumption | 0.31 | 0.33 | 0.36 | 0.39 | 0.43 | 0.48 | 0.56 |
| | Water Withdrawal | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| | Energy Demand | 0.30 | 0.32 | 0.34 | 0.37 | 0.41 | 0.46 | 0.53 |

Fig. 2. Heat map of calculated optimal supplementary cementitious materials to cement (s/c) ratio to reduce environmental impacts given strength at 28 days (note: for each SCM, green represents higher optimal s/c ratios and red represents lower optimal s/c ratios to lower the respective environmental impact(s)).

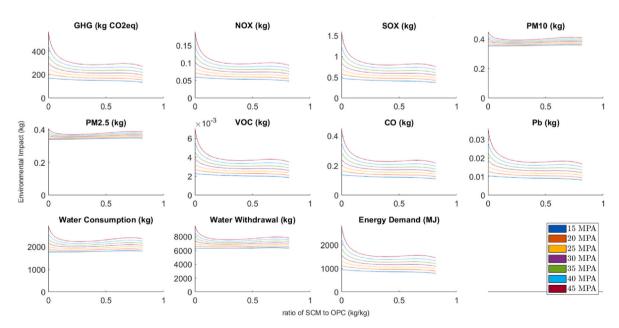


Fig. 3. Natural Pozzolan Environmental Impact (kg) vs ratio of SCM to OPC (kg/kg) (28 Day Strength).

mixtures simulated here to conduct optimization, the lowest GHG emissions achieved using NP resulted in a reduction of 35% relative to the mixture with no NP, occurring at s/c of 0.82. This s/c ratio results in a 1%–36% reduction of the other environmental impact categories

examined, but a negligible increase in PM_{2.5}.

For SA as an SCM, with the exception of water consumption, there was a consistent desired s/c across all impacts at 28 d and 60 d, 0.67 kg/kg. At higher ages, though, shifts occurred in the trends (see Fig. 4). For

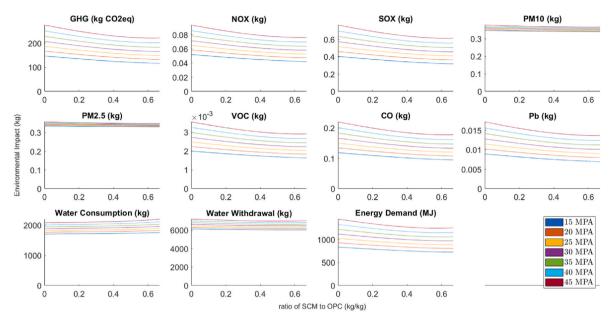


Fig. 4. Shale Ash Environmental Impact (kg) vs ratio of SCM to OPC (kg/kg) (180 Day Strength).

180 d, the optimal s/c ratio to lower most impact categories dropped slightly (to 0.57–0.65 kg/kg); a 3–15% overall drop in the optimal ratio that was mostly consistent across the 15–45 MPa strengths and across the 28 d–180 d range. Water withdrawal had similar results aside from 30 to 45 MPa at 28 d, where the optimal s/c range drops by approximately 50%. Water consumption was affected differently by changing the s/c ratio than the other impacts, where an optimal ratio to lower water consumption ranged between 0.0 and 0.10 kg/kg at 28 d, 60 d, and 180 d.

The calcined clay examined in this study was MK (see impact trends in Fig. 5), and an s/c ratio of 0.33 minimized GHG, SO_X , CO, and Pb emissions for all strengths and tests. The optimal s/c ratio to minimize NO_X emissions was the same as these other emissions (0.33 kg/kg) until 180 d; at this higher age, minimal emissions for 25–45 MPa concrete occurred at an s/c ratio of 0.32 kg/kg and for 15 MPa at 0 kg/kg. Similarly, for water withdrawal and VOC emissions, the optimal ratio was 0.33 kg/kg, but decreases slightly at 180 d design strength. For

energy demand and water consumption, again, 0.33 kg/kg was optimal at 28 d and 60 d, but for low strength concrete at 180 d, negligible use of the MK was desired. To lower PM $_{10}$ at 28 d, the optimal s/c ratio increased from 0 to 0.15 kg/kg at 45 MPa. The s/c ratios to lower PM $_{2.5}$ were similar to those of PM $_{10}$ where the optimal ratio increased from 0. to 0.09 kg/kg for 15–45 MPa at the same age (28 d). To lower PM $_{2.5}$ and PM $_{10}$ emissions at greater ages, lower s/c ratios were desirable. For MK, the optimal s/c ratio remained generally consistent between 0.28 and 0.33 kg/kg, except for particulate matter where the optimal ratio was typically lower, between 0 and 0.15 kg/kg to reduce impacts.

Use of SF had very consistent results across strengths and testing ages. The optimal s/c ratio to lower all impacts, excluding NO_X and water withdrawal, was 0.25 kg/kg for all strengths and testing ages. At higher strengths and earlier testing, 0.25 kg/kg was an optimal ratio, but at 180 d and lower strengths between 0.04 and 0.10 kg/kg was desirable. Minimal use of SF would minimize water withdrawal. For the mixtures simulated here to conduct optimization, the lowest GHG

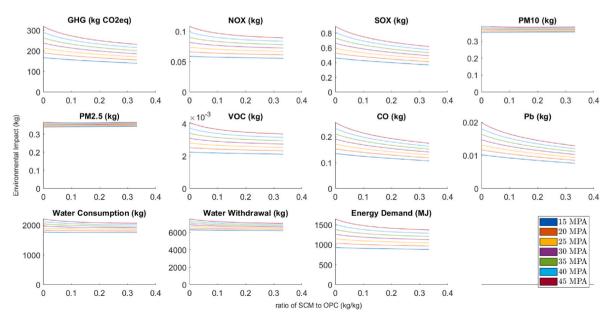


Fig. 5. Metakaolin Environmental Impact (kg) vs ratio of SCM to OPC (kg/kg) (28 Day Strength).

emissions achieved using SF resulted in a reduction of 23% relative to the mixture with no SF, occurring at s/c of 0.25. This s/c ratio resulted in a 3%–26% reduction of the other environmental impact categories examined, except water withdrawal which increased by 6% at this s/c ratio

For FA as an SCM, the optimal s/c ratio to lower GHG, NO_X , SO_X , VOC, CO, and Pb were between 0.37 and 0.49 depending on strength and age; the optimal s/c ratios to lower water withdrawal, water consumption, energy demand, and particulate matter were between 0.28 and 0.40, 0.19–0.28, 0.35–0.44, 0.40–0.52, respectively, depending on strength and testing age. The s/c ratio to minimize all impacts increased as strength increased. Additionally, there were slight increases in the s/c ratio to minimize impacts at 180 d compared to 28 d and 60 d. These trends were expected as a function of the contributions of FA to the formation of hydrate minerals and their dependency on time.

Relative to the other mineral admixtures studied, the optimal ratios of GGBS to OPC had a wide range to drive down environmental impacts. This greater variation likely reflects the cementitious characteristics of this SCM. Additionally, we note that data for higher replacement levels were available for GGBS than for the other SCMs, again, a function of it having cementitious characteristics. The optimal s/c ratio to lower GHG emissions was between 0.42 and 0.90 kg/kg, 0.59-0.99 kg/kg, and 0.55-0.77 kg/kg for 28 d, 60 d, and 180 d, respectively. The ratio increased as strength increased, ~2.2 fold at 28 d, ~1.7 fold at 60d, and ~1.4 fold at 180 d. At 28 d, low levels of GGBS replacement reduced NO_X emissions (0-0.05 kg/kg), but at 180 d, higher replacement levels were favorable (0.22-0.36 kg/kg). To minimize SO_X emissions, 0.4-1.02 kg/kg s/c ratios were desired with the optimal ratio increasing as strength increases. The optimal s/c ratios to lower VOC, CO, water consumption, and energy demand trend similarly to GHG and SOX emissions. The optimal s/c ratio to lower CO was 0.69-1.18 kg/kg increasing with strength and slightly varying with age. The optimal s/c ratios to lower PM₁₀ emissions, PM_{2.5} emissions, and water withdrawal were the same across all strengths and testing ages: the highest reduction of OPC to lower PM emissions and no OPC replacement to lower water withdrawal. For Pb emissions, as these primarily are driven by fuels used, high s/c ratios were typically favorable to reduce emissions. For the mixtures simulated here to conduct optimization, the lowest GHG emissions achieved using GGBS resulted in a reduction of 20% relative to the mixture with no GGBS, occurring at s/c of 0.57. This s/c ratio resulted in a 3%-36% reduction of the other environmental impact categories examined, but a 12% and 20% increase in NO_X and water withdrawal, respectively.

5. Discussion

The use of SCMs to drive down GHG emissions has become a frequent point of discussion in the cement and concrete industries [48]. Yet, the resources used can be globally or locally scarce [49]. In order to best utilize the resources available, their application should be implemented to gain maximum benefit. In the case of SCMs being used to lower environmental burdens, using appropriate replacement ratios can both reduce environmental impact and reduce consumption pressures that can lead to scarcities. Our findings emphasize the need to optimize the amounts of SCM used in concrete mixtures. Notably, our results for optimal SCM ratios are generally greater than what have been suggested to drive increases in strength. To show this trend, we present the optimal replacement levels and s/c ratios from the experimental studies used in this work to maximize compressive strength (see Table 5). While optimal s/c ratios vary by impact category and with increased concrete age, we note how these trends based on increasing compressive strength alone differ from the trends presented in Fig. 2, where results were more conservative [39-43]. Our work investigates methods to minimize multiple environmental impacts concurrently and not just to obtain required strength.

Since SCMs are limited resources, ensuring the best application of

Table 5Recommended replacement amounts from experimental literature [39–43].

| | Optimal s/c Ratio |
|--------------------------|-------------------|
| Limestone Filler | 0.176 |
| Natural Pozzolan | 0.176-0.25 |
| Shale Ash | 0.176-0.25 |
| Calcined Clay/Metakaolin | 0.25 |
| Silica Fume | 0.25 |
| Fly Ash | 0.667 |
| GGBS | 1.22–1.49 |

them in concrete mixtures is crucial. The best way to use them is to determine the strength needed, allowable age to reach desired strength, and the location in which they are to be used, then integrate the environmental impacts associated with production of the mixture. Considering multiple impacts allows for a more holistic analysis and can help mitigate local scarcities and/or localized air pollutants.

6. Conclusions

In this work, we found desired SCM to OPC ratios to minimize 11 environmental impacts by deriving a set of equations between strength, impacts, and concrete constituents. The key findings of this work are:

- The optimal s/c ratios to lower impacts vary depending on the impact, concrete strength, and age.
- With limestone filler, the optimal s/c ratios increase as strength increases. To lower GHG, NO_X , SO_X , VOC, CO, and Pb emissions, as well as energy demand, an s/c ratio of 0.21 kg/kg performed the best. To lower PM emissions, an s/c ratio less than 0.17 kg/kg was optimal. To lower water withdrawal and water consumption, an s/c ratio less than 0.19 kg/kg performed the best.
- With natural pozzolans, between 0 and 0.29 kg/kg resulted in lower PM emissions, water consumption, and water withdrawal. Higher amounts of NP lower GHG, NO_X, SO_X, VOC, CO, and Pb emissions; an s/c ratio of 0.82 kg/kg was desirable.
- With shale ash, an s/c ratio between 0.50 and 0.67 kg/kg lowered all impacts aside from water consumption, where no more than 0.10 kg/kg was ideal.
- With metakaolin, the s/c ratio to lower all impacts generally decreased as strength and age increased, except for PM emissions.
- With silica fume, the optimal s/c ratio was relatively consistent at ~ 0.25 kg/kg.
- With fly ash, the optimal s/c ratio to lower GHG, NO_X, SO_X, VOC, CO, and Pb emissions was between 0.38 and 0.49 depending on strength and age. The optimal s/c ratios to lower water withdrawal, water consumption, energy demand, and particulate matter were between 0.22 and 0.33, 0.31–0.42, 0.37–0.48, 0.42–0.51, respectively, depending on strength and age.
- With ground granulated blast furnace slag, higher optimal ratios are desirable at higher strengths. Optimal s/c ratios to lower PM₁₀, PM_{2.5}, and CO were consistently above 1, between 1.33 and 1.57 kg/ kg. Negligible use of GGBS is desirable to lower water withdrawal.

The results of this study indicate that optimizing the amount of SCM in concrete mixtures is crucial in order to prevent an increase in environmental impacts other than GHG emissions. Required performance, such as compressive strength in design, needs to be accounted for when determining a viable concrete mix. Further, depending on the SCM used the maximum s/c ratio may not be the optimum amount needed to reduce all environmental impacts. The concrete industry needs to consider what increasing SCM amounts in their mixtures will do on a local level, in order to prevent other detrimental consequences (e.g., harm to human health from increased particulate matter or water scarcity from increased water consumption).

Herein, this study focused on deriving a novel methodology that can be applied more broadly to decarbonize the cement and concrete industries, while limiting unintended consequences to other environmental burdens. In future research, a wider range of experimental inputs should be considered. These additional data can support robust comparisons across SCM types and better inform optimal mixtures for practice. Additionally, optimizing s/c ratios considering other material properties or functional units should be investigated.

Author contributions

Kelli A. Knight: Data curation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing. *Patrick R. Cunningham:* Writing – original draft, Writing – review & editing. *Sabbie A. Miller:* Conceptualization, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cemconcomp.2023.105049.

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Further reading

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