Statistical Variation in the Embodied Carbon of Concrete Mixtures

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Abstract

This study presents a refined calculation of the embodied carbon of concrete mixtures *via* life cycle assessment (LCA) with an explicit focus on three innovations. First, probability distributions that represent process-related variability in the embodied carbon of concrete are calculated using a variety of life cycle inventory data sources. Second, the traditional concrete LCA system boundary (*i.e.,* cradle-to-gate) is expanded to incorporate estimates of *in situ* carbon sequestration *via* concrete carbonation. Third, we analyze the impact of different transportation scenarios on the utility of using fly ash to reduce the embodied carbon of concrete. We use these data to heuristically determine the breakeven transportation distance for fly ash *via* trucking to be 2655 km for domestic sources of fly ash. However, when fly ash is imported from international sources, reductions to embodied carbon attributed to fly ash replacement can be negligible. The calculated breakeven maritime shipping distance for fly ash equals 15,110 km—beyond which, the anticipated embodied carbon reductions due to fly ash use in concrete are compromised due to transportation. The advancements described herein enable improved scenario-based decision-making for understanding, quantifying, and reducing the embodied carbon of concrete mixtures. In addition, the results highlight the importance of accounting for international transportation of fly ash in LCAs, especially given that domestic sources of quality fly ash are expected to continue to decline and imports are expected to increase in many parts of the world over the next few decades.

Keywords: concrete; life cycle assessment; embodied carbon; variability; fly ash; transportation

1. Introduction

The manufacturing of cement, the most carbon-intensive component of concrete, contributes about 7% of global carbon dioxide CO_2 emissions [1]. Due to the ubiquity of concrete and its contribution to anthropogenic CO_2 emissions, a large body of literature exists that aims to understand, quantify, and subsequently reduce the embodied carbon of concrete. Multiple comparative life cycle assessment (LCAs) of alternative concretes or concrete materials that compare the embodied carbon of different concrete mixtures have been published [2]–[11]. In addition to a myriad of LCAs that report environmental impacts of a functional unit volume of concrete (*e.g.*, 1 m^3, 1 yd^3), other application- (*e.g.*, pre-cast concrete, pavements) and geographically-specific concrete LCAs have also been published in the literature [12]–[20].

Despite the number of studies that exist in the literature, concrete LCAs are typically deterministic (*vs.* probabilistic) in nature, meaning that environmental impacts (e.g., embodied carbon) are calculated and reported using numerical inputs that do not consider uncertainty. Primarily due to (1) a lack of necessary data and (2) additional statistical analyses that are required to incorporate variability and uncertainty, process-related variability and uncertainty in the environmental impacts are rarely quantified and reported. However, a better understanding of the variability of environmental impacts would best elucidate "hotspots" of uncertainty in the product life cycle and identify where future studies should focus their efforts. In addition, probabilistic analyses provide insight into the amount that environmental impacts can vary and removes the false impression that environmental impacts for a product are static and perfectly quantified. The aforementioned concrete LCAs are examples of deterministic studies that do not account for process- or geographical-related variability. One exception concerns pavement LCAs, which assign distributions to LCA input parameters in order to quantify the uncertainties in carbon emissions [19], [20].

In addition to being predominantly deterministic in nature, concrete LCAs are most often conducted using a cradle-to-gate system boundary [12], [21]–[27]. Such analyses are confined to the manufacturing stage [28]. Concrete has a unique material life cycle, because a non-trivial quantity of CO_2 can be sequestered during the use and end-of-life phases .For instance, Souto-Martinez, *et al*. (2018) reported up to 19% of initial carbon emissions of concrete can be sequestered through *in situ* and end-of-life carbonation, but the theoretical carbon sequestration potential depends on the composition and quantity of the cement binder [29]. Some LCA studies have incorporated concrete carbonation during use and end-of-life, but these studies are also deterministic and do not quantify the variability in embodied carbon calculations [29]–[32].

In order to reduce concrete embodied carbon, supplementary cementitious materials (SCMs), such as fly ash from coal power plants, are often used to replace a portion of the cement quantity in a concrete mixture. Several studies in the literature have quantified the potential reduction in concrete embodied carbon due to fly ash replacement [26], [33], [34], where the majority of studies use a fixed distance assumption or assume that the carbon dioxide emissions allocated to fly ash is zero. However, as utilization rates increase and international imports become more common in many regions of the world, it is likely that fly ash transportation distances will also increase. We hypothesize that the actual environmental benefit of using fly ash to reduce the embodied carbon of concrete is highly dependent on the fly ash transportation mode and distance, thereby necessitating more explicit studies of the effects of fly ash transportation on overall estimates of the embodied carbon of concrete.

The purpose of this study was to develop a probabilistic approach to model the process-related variability inherent to estimates of the embodied carbon of concrete. Probability distributions were created using a variety of life cycle inventory data sources and probabilistic estimates of cement and concrete processing technology. These probability distributions were implemented in a stochastic, process-based, cradle-to-grave LCA that incorporates estimates of *in situ* and post-use CO_2 sequestration *via* concrete carbonation using $1m^3$ as the primary functional unit. The results elucidate the range of emissions that are possible for concrete due to statistical process variability throughout the life cycle. To illustrate the utility of this work, the emissions distributions are applied to five concrete mixture designs of varying compressive strength in order to illustrate how both process-related variability and mixture design variability impact the distribution of concrete CO_2 emissions on a volumetric basis. In addition, the impacts of transportation mode (*e.g.*, trucking, maritime shipping) and distance on estimates of embodied carbon are explicitly studied. It is envisioned that the results of this work will advance the development concrete design methods and strategies that result in surefire reductions in environmental impacts [35]–[37].

2. Life Cycle Assessment (LCA) Methodology

In this analysis, we conduct a process-based LCA, which quantifies the inputs and outputs to nature in order to evaluate environmental impacts over the life cycle of concrete. A process-based LCA involves four stages as outlined by the International Organization for Standards (ISO) 14040; these stages include (1) goal and scope definition, (2) inventory analysis, (3) impact assessment, and (4) interpretation [38].

2.1 Goal and Scope Definition

2.1.1 Goal

The goal of this study was to develop probabilistic distributions that enable designers to quantify the embodied carbon of a given concrete mixture. The intended users are other LCA practitioners or researchers who would like to gain a greater understanding of the variability of embodied carbon in concrete mixtures. The results of this study are intended to be used in comparative assertions or in other concrete LCA literature.

2.1.2 Functional Unit

In this study, we acknowledge that compressive strength is often the most important performance measures in concrete mixture design, and concrete mixtures of different compressive strengths are neither functionally necessarily functionally equivalent nor comparable. Thus, for the case studies analyzed herein, the functional unit is 1 m^3 of concrete for a given compressive strength. However, no other performance requirements are imposed in order to allow users of the model to impose other performance requirements *a posteriori*. In other words, this study enables flexible implementation of many types of performance requirements in downstream uses of the model.

2.1.3 System Boundary

The system boundary includes the following life cycle stages: product stage (A1-A3), use stage (B1), and end-of-life disposal (C4). Figure 1 illustrates this "cradle-to-gate plus carbonation" system boundary. Unlike many prominent concrete LCA studies that use a cradle-to-gate analysis [12], [21]–[26], this analysis includes carbonation impacts from the use phase and end-of-life. This system boundary choice is critical for better understanding the benefit of carbonation in reducing the *net* embodied carbon of concrete mixtures and for making informed choices about when and in what proportions to use fly ash in order to reduce the embodied carbon of concrete. While some construction, use, and end-of-life stage emissions have been omitted, it is assumed that, for a given set of concrete performance requirements (*e.g.,* required compressive strength or service life), that the omitted impacts will be equivalent for all concrete mix designs considered herein.

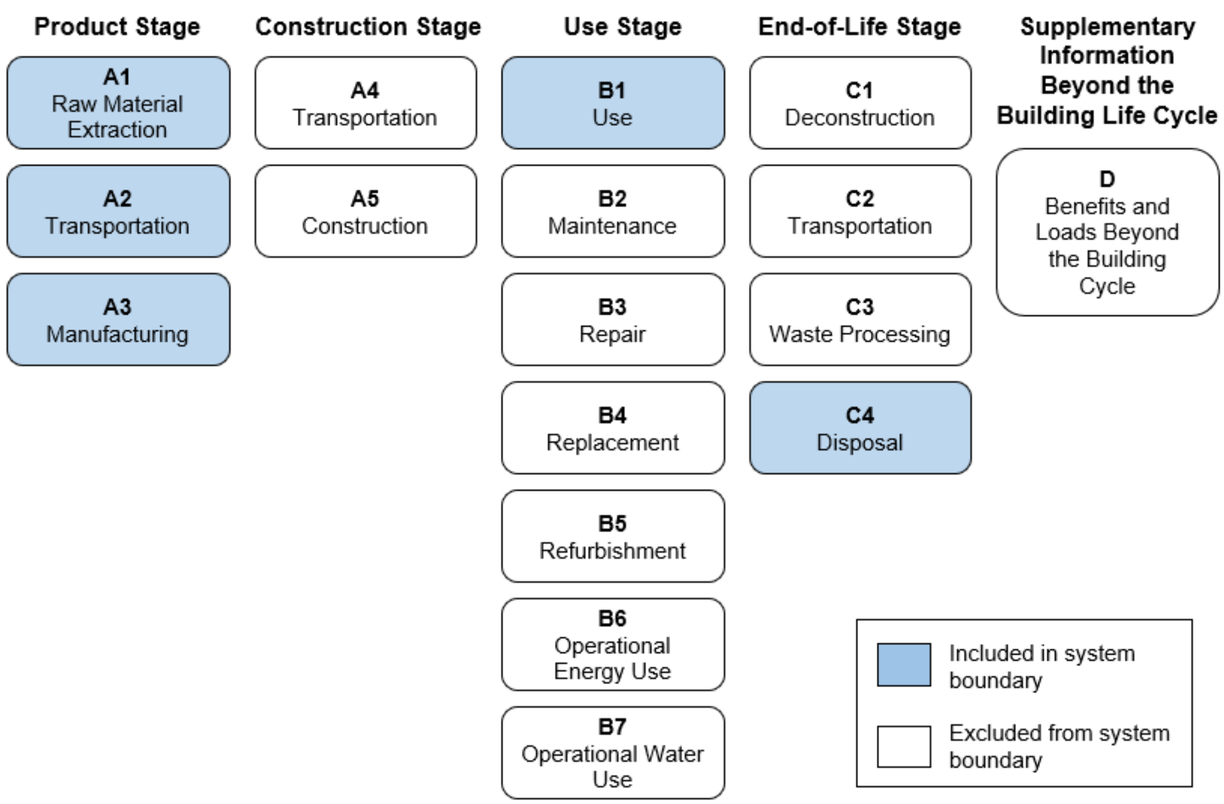

Figure 1. LCA system boundary. Visualization adapted and modified from [39].

2.1.4 Methodological Choices

The environmental impact category considered by this study is global warming potential, which is reported using units of kg CO_2e. Thus, the life cycle emissions associated with global warming, (*e.g.*, carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O)) are accounted for in the life cycle inventory. All greenhouse gas emissions are converted to CO_2-equivalent (*i.e.,* CO_2e) emissions in order to have a single unit of measure of embodied carbon. In addition, in this analysis, SCMs such as fly ash are considered by-products from other industries. Thus, emissions for SCM beneficiation and transportation are included in the analysis, while emissions for their initial generation are excluded. Some studies have considered other allocation analyses for fly ash (e.g., allocation by economic value, allocation by mass) [40]–[42]. However, the current product category rule (PCR) for concrete in the United States, which was developed by the Carbon Leadership Forum for use by the National Ready-Mix Concrete Association (NRMCA), prescribes fly ash as a by-product [43]. In order to remain consistent with other analyses, this study follows the most recent PCR guidelines.

2.1.5 Probabilistic Methods

This analysis differs from a deterministic LCA because it employs density functions (*i.e.,* probability distributions) in order to quantify and represent embodied carbon variability of concrete in the United States. The analysis is conducted in the same manner as a traditional LCA in which the emissions of all constituents are summed and reported in units of carbon dioxide-equivalent. However, in this study, each major input to the life cycle inventory is assumed to be an uncertain parameter rather than a deterministic value. Thus, for each life cycle stage, we performed the following series of steps in order to develop probability distributions representing variability in concrete embodied carbon:

1. Gather relevant data sources related to the life cycle stage
2. Determine and justify an appropriate probability distribution
3. Calculate characteristic parameters for the given distribution based on data sources
4. Stochastically sample from these distributions using Monte Carlo simulation (with n = 10,000)
5. Sum the emissions impacts for each life cycle stage

In cases where there is substantial emissions-related data, density functions are generated as an empirical smoothed histogram. In cases where data are sparse, we assume a mathematical form of the distribution (*e.g.,* normal, uniform) *a priori* and use life cycle inventory data samples to determine the parameters for the distribution. In addition, when there are different processing methods available (*e.g.*, different cement kiln types), we weight the processing emissions based on the proportional representation of that processing method for the United States (US). The data sources and justification for the selected distributions are provided in Section 3.

3. Life Cycle Inventory Data and Assumptions

LCA studies require significant quantities of data in order to account for the life cycle emissions of a product or process. This study utilizes a variety of US-based data sources for the life cycle inventory for concrete. The sections below describe each life cycle stage and the justification for the selected probability distribution. In addition, the life cycle inventory data is summarized in **Table 1**.

3.1 Raw Material Extraction (A1)

3.1.1 Cement

Cement is the primary constituent of concrete. When mixed with water, cement hydrates and hardens to form the paste that binds aggregates together. Cement production is emissions-intensive. Cement production involves raw material extraction, crushing, grinding, kiln firing, and blending. In particular, the kiln-firing step results in the release of large quantities of CO_2 due to two contributors. First, kiln firing is the processing step in which carbon dioxide is driven off during the cement calcination reaction as is shown in Equation 1.

$$CaCO_3 \rightarrow CaO + CO_2(g) \quad \text{(Eq. 1)}$$

where limestone ($CaCO_3$) is heated to form calcium oxide (CaO) and gaseous CO_2. Second, this reaction must occur at approximately 1450 °C, which, typically, cannot be achieved with electricity; thus, the reaction requires large quantities of fuel [44]. A major source of process-related variability in cement emissions is due to the use of different kiln types. For instance, dry kilns that utilize preheater and precalciner technologies are more efficient than wet kilns [45].

Emissions-related data for this life cycle process is acquired using a survey conducted by the Cement Sustainability Initiative that reported the annual fuel and electricity use of 849 cement plants in the United States [46]. Data for the release of CO_2 emissions due to calcination is retrieved from the IPCC guidelines related to greenhouse gas inventories for the calcination reaction and is based on chemical stoichiometry [47]. In addition, emissions factors for fuel and electricity emissions are well-established [48]–[51].

3.1.2 Aggregates

Coarse aggregate and sand are inert mixture ingredients used in concrete and can be up to 85% of the mass of concrete [52]. Natural aggregates typically require very little processing. The key processing steps in aggregate production are (1) acquisition and (2) crushing to the appropriate size. The major source of process-related variability for aggregate-related emissions is due to different types of acquisition methods. For instance, aggregate may be land-won, marine dredged, or acquired from crushed rock; these acquisition methods are associated with different fuel and electricity consumptions.

Data from the USGS is used to determine the fraction of total aggregate that is land-won, marine dredged, or from crushed rock [53]. In addition, emissions-related data for each acquisition method is collected based on previous life cycle inventories in the literature [54]–[56]. Since this study utilizes multiple values per acquisition method, we assume that these values are samples from a normal distribution of emissions.

3.1.3 Water

Concrete requires addition of water for the hydration of cement. Using standard water-to-cement ratios for concrete mix designs and a CO_2 emissions factor for US water production [57], water contributed less than 1% of the total emissions of a cubic meter of concrete and is thus omitted from this analysis.

3.1.4 Fly Ash

Fly ash—a waste product generated at coal power plants—has pozzolanic properties when blended with ordinary portland cement. Consequently, fly ash is often used to replace a portion of the cement used in concrete mixtures. Since fly ash is a byproduct from the combustion of coal, the allocated emissions from fly ash are assumed zero, according to a well-established product category rule for concrete materials [58]. However, due to regulations concerning NOx emissions, some US fly ash sources contain excessive quantities of unburnt, or *loss-on-ignition* (LOI), carbon. Such sources of fly ash require processing to lower the LOI to < 6% by weight [59]; such processing is associated with CO_2 emissions.

Removal of excess carbon-in-ash can be performed *via* multiple different processes. In thermal beneficiation, excess carbon is burned off in a boiler with high combustion performance. The energy from this process is often used to power the beneficiation process but is nonetheless associated with CO_2 emissions. Another beneficiation method, triboelectrostatic separation, exploits the difference in electron affinity between fly ash particles and carbon particles. Under an electric field, carbon particles become positively charged and fly ash particles become negatively charged. Subsequently, the particles are diverted to separate electrodes of opposite charge.

Due to the confidential nature of specific fly ash beneficiation technology and data, the CO_2 emissions associated with this process rely on several assumptions. Among the five leading global vendors of fly ash, three primarily use thermal beneficiation and two primarily use triboelectrostatic separation. Thus, we assume a 3:2 ratio of the thermal and triboelectrostatic separation technologies [60]. For the thermal process, CO_2 emissions are calculated assuming uniform range of possible LOI values of the unprocessed fly ash from 0.2-20.5% [61] . The final LOI is assumed to be 0-2% after beneficiation [62], which meets the current US fly ash LOI standards. In addition, we assume that this carbon is fully converted to CO_2. For the triboelectrostatic method, data are gathered from a study reporting the electricity used to remove excess carbon from fly ash [63] as well as the variable electricity emission factors reported by the EPA [48]. Notably, CO_2 emissions due to fly ash beneficiation resulted in < 1% of the total emissions from a cubic meter of concrete and are thus omitted from this analysis.

3.1.6 Admixtures

Chemical admixtures are used in relatively small quantities to tailor concrete properties for certain applications. In this analysis, we focus primarily on superplasticizing admixtures, which improve the workability and flowability of concrete even at low water-to-cement ratios. Three major types of superplasticizers include sulfonated naphthalene formaldehyde, sulfonated melamine formaldehyde, and polyacrylates.

Superplasticizer production involves proprietary chemical processes. In this study, emissions-related data are acquired from Environmental Product Declarations (EPDs) provided by admixture manufacturing associations and a few life cycle assessment studies [5], [64]–[68]. We use reported emissions as a data point and generate a normal distribution for calculating variability. Although this method generates a highly variable estimate in superplasticizer embodied carbon, the quantity of superplasticizer used in most concrete mixtures is low. Therefore, the contribution to overall uncertainty of the mixture is small.

3.2 Transportation (A2)

Each raw material listed in Section 3.4 must be transported to a ready-mix plant. Transportation involves emissions associated with primary emissions (*e.g.*, burning fuel) and secondary emissions (e.g., fuel production, truck manufacturing). Emission-related data for raw material transportation includes the emission factors for truck and maritime transportation (in kg CO_2e per tonne-mile) [69]. The major source of emissions variability for this input is due to transportation distance variability for each mixture ingredient. The two sections below indicate how transportation distance variability is determined for the mixture ingredients. Note that admixture transportation is ignored due to the very small quantities of superplasticizer that are utilized; admixture and water transportation are assumed to be << 1% of the total emissions from 1 m^3 of concrete.

3.2.1 Cement and Aggregate

In order to determine the distribution of distances that cement and aggregate are transported, this study employs a National Ready Mix Concrete Association report that provides survey data regarding the transportation distances for cement and aggregate, as reported by ready-mix companies [70].

3.2.2 Fly Ash

Quantifying fly ash transportation distances is complex because data representing fly ash transportation distance from the source to the ready-mix plant are currently unavailable. Consequently, in this study, we present a novel method for generating a distribution of transportation distances for fly ash from present-day coal power plants in the US.

In order to generate a distribution of fly ash transportation distances, a geospatial analysis was performed using QGIS v3.4 [71]. Point-source data were collected for coal power plant locations from the US Energy Information Agency [72]. The 9189 power plant locations were reduced to 264 by selecting only those that were coal plants with production capacity larger than 100 MW. This criterion was applied based upon the assumption that power plants with less than 100MW of coal production would not be primary suppliers of fly ash. A 30 arc-second grid was created from the CIESIN gridded dataset for US census data, including population [73]. These data were both re-projected into the USA Contiguous Equidistant Conic projection. Using QGIS' built-in *Proximity (Raster Distance)* tool, the Euclidean distances were calculated between the centroid of each raster grid and the nearest coal plant larger than 100 MW. Euclidean distances were multiplied by a drive distance factor which is used to approximate real transportation distances from straight-line distances [74].

Because the Euclidean distance from coal power plants is not representative of fly ash utilization, population was used as a weighting factor. Using the CIESIN gridded dataset, each grid cell was "weighted" (*w*) by its population (people per square kilometer) and rounded down to the nearest whole number. A histogram of distance values was generated where each grid cell contributes *w* contributions of its distance value to the histogram. A *k*-nearest neighbors smoothing algorithm was used to plot the histogram as a density function (*i.e.,* probability distribution). A key assumption in this analysis is that the closest source of fly ash will be used rather than one further away. Section 5.2 considers how other modes of transportation and distance scenarios can impact the contribution of fly ash to the embodied carbon of a concrete mixture.

3.3 Manufacturing/Production (A3)

Approximately 75% of concrete in the U.S. is produced at ready-mix plants [75]. Although not all US concrete is produced in ready-mix plants, due to data availability, this study uses ready-mix manufacturing data for the life cycle inventory. Thus, this study is applicable only to ready-mix concrete. Here, concrete mixture ingredients are either gravity fed into mixer trucks or are added to a central mixing drum and then transported to a transportation truck, which uses fuel and electricity. The variability in emissions from this batching process is due to individual variabilities among ready-mix plants in technology and efficiencies. This study utilizes a survey from the National Ready Mix Concrete Association on fuel and energy use at U.S. ready mix plants [70].

3.4 Use and End-of-life Carbonation (B1, C4)

During the use phase through end of life, CO_2 from the environment reacts with calcium compounds in Portland cement to form calcium carbonate ($CaCO_3$). This process, which is diffusion-based, is termed concrete carbonation. Due to this process, the use and end-of-life phases effectively reduce the total of embodied carbon of a concrete mixture. The quantity of sequestered CO_2 is a function of the quantity of the calcium oxide (CaO) content of the cementitious material as well as the degree of carbonation (DoC).

The CaO content of cement is a relatively consistent quantity, and thus the maximum CO_2 uptake for a Portland cement can be calculated using the following equation, assuming a 95% clinker content per cement [78]:

$$U_K = (\%CaO) * 0.95 * \left(\frac{clinker}{cement}\right) * \left(\frac{44}{56}\right)\frac{MCO_2}{MCaO} = 0.49\frac{kg\ CO_2}{kg\ cement} \quad \text{Eq. 1}$$

where U_k is the maximum uptake of CO_2 per kilogram of Portland cement and *M* is the molecular weight of a given compound.

However, the CaO content of fly ash is highly variable and can range from 1-40% by weight [78]. Note that Class C fly ash generally contains greater than 20% CaO content, while Class F fly ash has less than 7% CaO content.

The (DoC) in concrete remains a poorly understood and uncertain phenomenon in the literature. DoC is defined as the proportion of CO_2 in the carbonated zone absorbed in a concrete compared to the theoretical limit to the quantity of carbonation. One study by Andrade performed experiments on concrete specimens under several exposure conditions to quantify the variability (in the form of mean and standard deviation) of the DoC for concretes with and without SCMs and reported the variability in the form of mean and standard deviation of DoC [76]. We utilize the statistical information (*i.e.,* mean and standard deviation) from this study for relevant clinker types in order to quantify the variability of concrete carbonation. For instance, for CEM I cement with no SCM replacement, the DoC was found to be 0.611 +/- 0.158; for a CEM II cement with 25% fly ash replacement, the DoC was found to be 0.682 +/- 0.140.

Table 1. Summary of life cycle inventory data sources and distribution parameters

Life Cycle Inventory Data	***Description***	***Distribution Type***	***Distribution Parameters***	***Units***	***References***
A1 - cement	*Cement plant operations fuel and electricity use*	*Empirical (nonparametric)*	*n/a*	*kg CO_2 / tonne cement*	*[46]*
	Calcination reaction emissions	*Deterministic*	*μ = 522*	*kg CO_2 / tonne cement*	*[77]*
A1 – fine aggregate	*Emissions from electricity and fuel use for land-won acquisition*	*Triangular*	*a = 0.25, b = 3.45, c = 1.85*	*kg CO_2 / tonne aggregate*	*[54]–[56]*
	Emissions from electricity and fuel use for marine dredging	*Triangular*	*a = 34.24 b = 41.65 c = 37.95*	*kg CO_2 / tonne aggregate*	*[54]–[56]*

A1 – superplast-icizer	*Superplasticizer manufacturing emissions*	*Normal*	*μ = 1792* *σ = 428*	*kg CO_2 / tonne SP*	*[5], [64]–[67]*
A2 - cement transportation	*Distance*	*Normal*	*μ = 102.5* *σ = 48.7*	*km*	[70]
	Emissions for truck transportation	*Deterministic*	*μ = 0.203*	*kg CO_2 / tonne / km*	[69]
	Emissions for ship transportation	*Deterministic*	*μ = 0.037*	*kg CO_2 / tonne / km*	[69]
A2 – coarse aggregate transportation	*Distance*	*Normal*	*μ = 26.1* *σ = 10.5*	*km*	[70]
A2 – fine aggregate transportation	*Distance*	*Normal*	*μ = 25.9* *σ = 12.0*	*km*	[70]
A2 – fly ash transportation	*Distance*	*Empirical (nonparametric)*	*n/a*	*km*	*Quantified herein*
A3 - manufacturing	*Quantity of diesel*	*Normal*	*μ = 1.968* *σ = 0.328*	*L diesel/m^3 concrete*	[70]
	Quantity of natural gas	*Normal*	*μ = 0.336* *σ = 0.079*	*m^3 natural gas/m^3 concrete*	[70]
	Quantity of electricity	*Normal*	*μ = 5.050* *σ = 0.913*	*kWh/m^3 concrete*	[70]
B1, C4 - carbonation	*Carbonation during use and end-of-life for CEM I*	*Normal*	*μ = 610.8* *σ = 158.0*	*kg CO_2 / tonne CaO*	[76]
	Carbonation during use and end-of-life for CEM II	*Normal*	*μ = 681.7* *σ = 139.6*	*kg CO_2 / tonne CaO*	[76]
	CaO content of fly ash	*Triangular*	*a = 0.01* *b = 0.40*	*Weight %*	[78]

			c = 0.20		
Fuel emissions	*Emissions from generation and use of natural gas*	*Deterministic*	*μ = 2.386*	*kg CO_2e/ unit fuel*	*[49]*
	Emissions from generation and use of diesel	*Deterministic*	*μ = 3.152*	*kg CO_2e/ unit fuel*	*[50]*
Electricity emissions	*Life cycle emissions from electricity*	*Triangular*	*a = 0.228* *b = 0.757* *c = 0.453*	*kg CO_2e/ kWh*	*[48]*

4. Results and Discussion

4.1 Embodied carbon on a per mass basis

The results of the life cycle inventory are reported on a *per mass* basis in **Figure 2**. The variability of embodied carbon is represented using boxplots. Since the range of possible values for all life cycle inputs is large, the plot is categorized into (a) high, (b) medium, and (c) low contributions to life cycle inputs on a per mass basis, which enables visualization of carbon variability regardless of scale. For visualization purposes, the boxplot representing carbonation impacts from the use phase and end-of-life (*i.e.*, negative embodied carbon values) are omitted from these graphs but are discussed in detail in **Figure 3**.

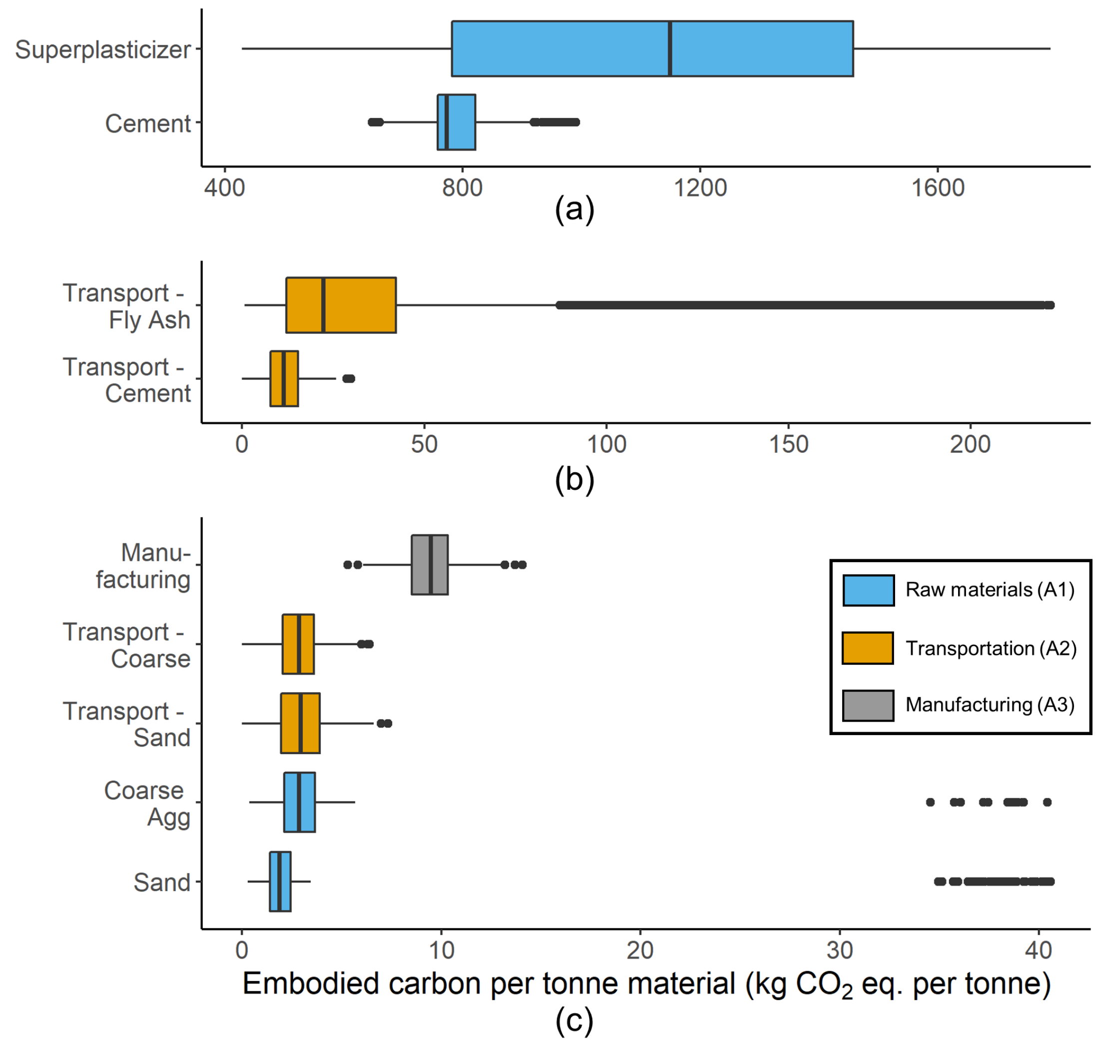

Figure 2. Embodied carbon of life cycle inputs on a per mass (tonne) basis categorized by (a) high, (b) medium, and (c) low impacts per mass.

High contributors to concrete embodied carbon *on a per mass basis* include cement and superplasticizer raw materials with mean values of 793.1 kg CO_2e per tonne and 1121.4 kg CO_2e per tonne, respectively. Medium contributors to concrete embodied carbon on a per mass basis include transportation of fly ash and transportation of cement with mean values of 44.1 kg CO_2e per tonne and 11.3 kg CO_2e per tonne, respectively. Since certain areas of the US must transport fly ash large distances, there is a large skew in the distribution for fly ash transportation emissions. This result will be discussed in greater detail in Section 5.2. The low contributors to embodied carbon are concrete manufacturing (9.4 kg CO_2 per tonne), coarse aggregate transportation (2.8 kg CO_2e per tonne), sand transportation (2.9 kg CO_2e per tonne), coarse aggregate raw material processing (3.4 kg CO_2e per tonne), and sand raw material processing (3.7 kg CO_2e per tonne). **Table 2** reports summary statistics (*i.e.,* median, 25th

quantile, and 75th quantile) for each of these life cycle impact contributors. This table can be used to calculate the embodied carbon of any concrete mixture (*i.e.,* different mixture ingredient quantities), which can be used in other LCA studies and downstream calculations.

Table 2. Summary statistics for life cycle input distributions

Life Cycle Input	*Median Value (kg CO_2e per tonne)*	*25th Quantile (kg CO_2e per tonne)*	*75th Quantile (kg CO_2e per tonne)*
Superplasticizer	*1149.9*	*782.4*	*1457.6*
Cement	*773.7*	*758.0*	*821.6*
Fly ash transportation	*22.3*	*12.1*	*42.2*
Cement transportation	*11.5*	*7.5*	*14.8*
Coarse aggregate transportation	*2.8*	*2.1*	*3.6*
Sand transportation	*2.9*	*2.0*	*3.8*
Coarse aggregate	*2.9*	*2.1*	*3.6*
Sand	*1.9*	*1.4*	*2.4*
Manufacturing	*9.5*	*8.5*	*10.3*
Use and end-of-life carbonation for cement	*297.8*	*242.6*	*350.5*

4.2 Embodied carbon for concrete mixtures of varying strengths

In addition to understanding the embodied carbon for individual life cycle inputs on a per mass basis, the mixture proportions and relative quantities of each constituent will impact the total embodied carbon of a concrete mixture. Thus, in this analysis, ten concrete mixtures of five different compressive strengths are compared. For each target compressive strength, a pair of mixture designs are compared, one which contains no fly ash and one which utilizes 20% fly ash replacement. **Table 3** reports the mixture proportions for each of these concrete mixtures. The mixture designs analyzed in this analysis are from a deterministic LCA report by the NRMCA. Note also that in order to achieve equivalent target compressive strength, the mixtures with fly ash replacement utilize greater total quantity of cementitious materials. This is consistent with the functional unit employed in this study.

Table 3 Constituent quantities for the representative concrete mixtures

Mixture Number	*Strength (MPa)*	*Cement (kg)*	*Fly Ash (kg)*	*Water (kg)*	*Coarse Aggregate (kg)*	*Fines (kg)*	*Water Reducer (kg)*
1a	*21 MPa*	*288*	*0*	*155*	*995*	*807*	*.085*
1b	*21 MPa*	*243*	*61*	*155*	*995*	*759*	*.085*
2a	*28 MPa*	*365*	*0*	*155*	*995*	*744*	*.085*
2b	*28 MPa*	*307*	*77*	*155*	*995*	*683*	*.085*
3a	*34 MPa*	*456*	*0*	*160*	*913*	*750*	*.198*
3b	*34 MPa*	*384*	*96*	*160*	*913*	*675*	*.198*
4a	*41 MPa*	*481*	*0*	*173*	*913*	*772*	*.198*
4b	*41 MPa*	*405*	*101*	*173*	*913*	*692*	*.198*
5a	*55 MPa*	*567*	*0*	*173*	*913*	*701*	*.198*
5b	*55 MPa*	*477*	*119*	*173*	*913*	*608*	*.198*

Figures 3a and **Figure 3b** show the estimated embodied carbon from each life cycle contributor for 1 m^3 of concrete of Mixture 1a and Mixture 1b, which have a target compressive strength of 21 MPa. The relative embodied carbon for each life cycle contribution is different than Figure 2 because concrete mixture ingredients are used in different quantities. This analysis is similarly divided into "high contributors" and "low contributors".

Notably, cement is by far the largest contributor of embodied carbon in both mixtures, as expected, given the emissions associated with heating limestone to high temperatures and the calcination reaction that releases large quantities of carbon dioxide [29]. Furthermore, due to fly ash cement replacement in Mixture 1b (and subsequent lower quantity of cement), the embodied carbon is lower than Mixture 1a. Mixture 1b also has an additional contribution due to fly ash transportation, which is relatively low compared to other impacts, based on our assumption of fly ash transportation from the nearest coal power plant. It is worthwhile to note that superplasticizer has become a relatively low contributor to total embodied carbon of the concrete mixture, due to the low mass of superplasticizer in both mixtures.

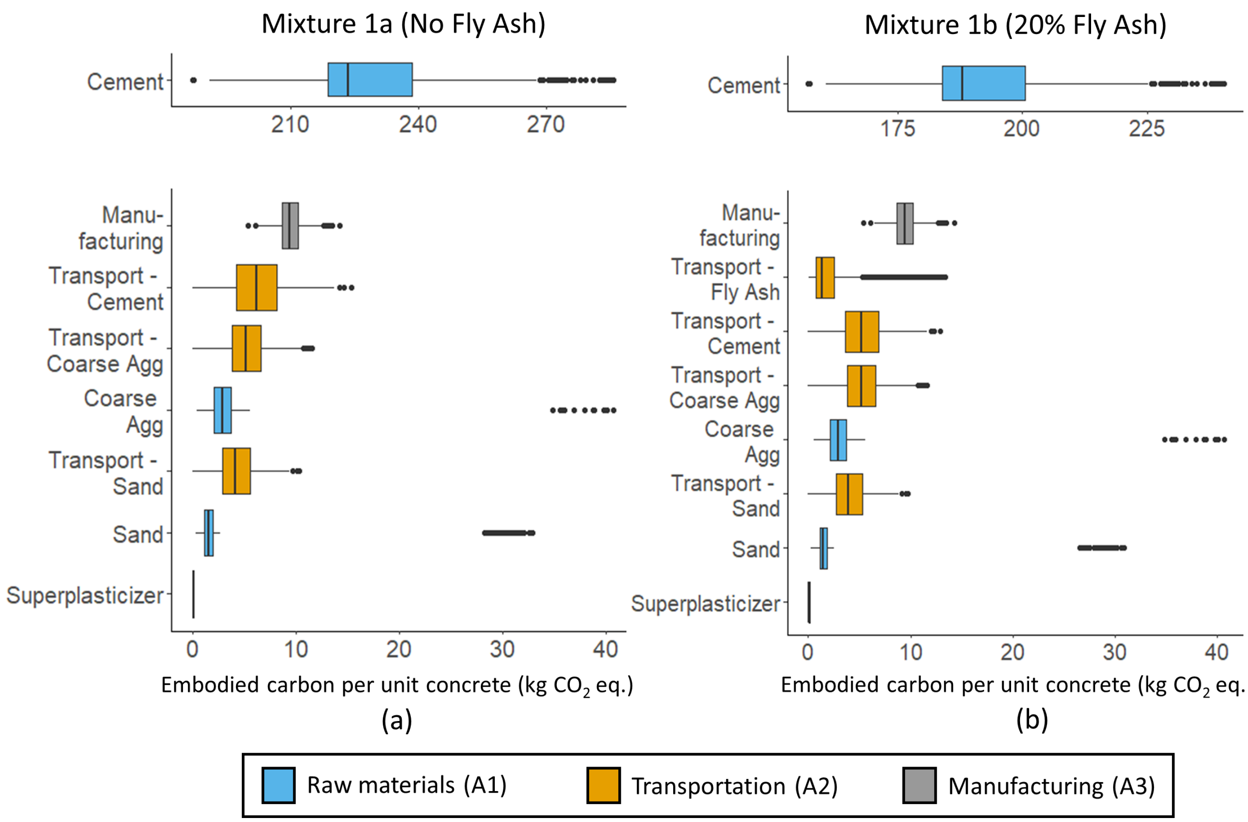

Figure 3. Boxplots illustrating life cycle impacts for Mixture 1a and Mixture 1b.

Analysis of the size of the individual boxplots is also illustrative of sources of variability. In Figures 3a and 3b, the whiskers of the boxplots represent 1.5 times the interquartile range beyond the 25th and 75th percentiles. Comparing the length of these whiskers in both Figures 3a and 3b illustrates that the major source of total variability for both mixtures originates from the variability in cement emissions. For instance, the range of the whiskers for cement in Mix 1a is 70 kg CO_2e, while the next-widest set of whiskers among the other life cycle inputs is 13 kgCO_2e. This analysis illustrates that the best way to reduce uncertainty in embodied carbon is to better understand the emissions associated with cement production (*e.g.*., determine the kiln type and fuels used).

4.3 Total embodied carbon for all mixture designs

In order to compare the total embodied carbon of both mixtures, **Figure 4** reports boxplots representing the net embodied carbon for Mixtures 1a-5b. The net embodied carbon is the sum of the cradle-to-gate and carbonation embodied carbon. One key trend is that embodied carbon values increase as the mixtures' compressive strength values increase, which illustrates the importance of specifying the compressive strength requirement for a given analysis. In addition, for each pair of mixtures of equivalent strength, the mixture with fly ash has a lower *median* embodied carbon value than the complementary mixture (without fly ash). However, the relative range of the distributions illustrates that it is possible for the fly ash mixtures to have higher life cycle embodied carbon than the "no fly ash" mixture of the same strength. Such a phenomenon is likely to occur when fly ash transportation emissions are high.

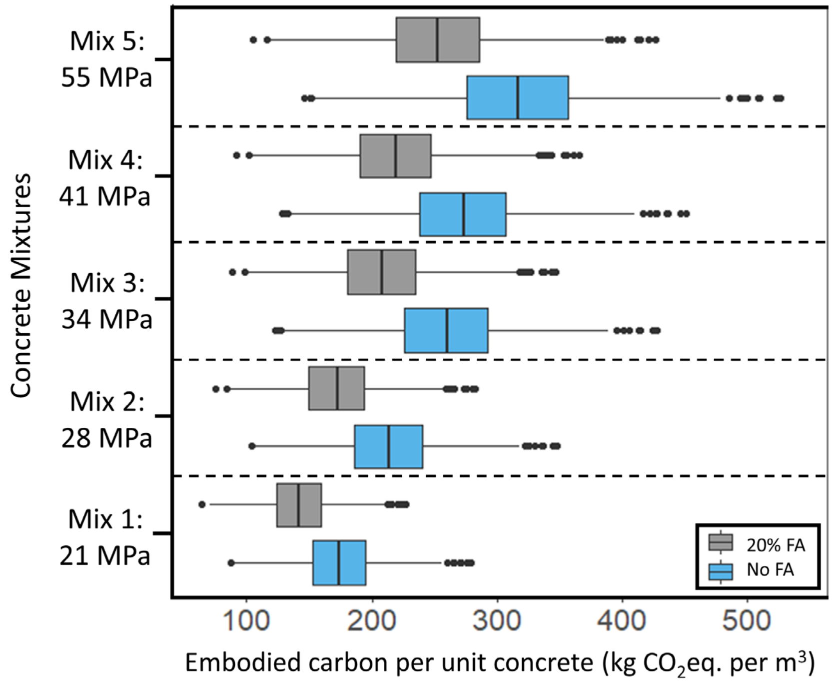

Figure 4. Distributions of net embodied carbon for ten concrete mixtures

In order to further analyze these results, Figure 5 illustrates the breakdown of emissions for Mixtures 1a and 1b in terms of the cradle-to-gate, carbonation, and net total embodied carbon, where the net total embodied carbon is the sum of the cradle-to-gate and carbonation embodied carbon. Mixture 1a has a slightly more negative value of carbonation-related embodied carbon because cement has a greater propensity to sequester CO_2 than fly ash. However, fly ash (especially fly ash with high proportions of calcium oxide) has been shown to sequester carbon dioxide over the concrete life cycle. In addition, Mixture 1b contains a higher quantity of total cementitious materials content than Mixture 1a in order to ensure equivalent compressive strength. For these reasons, Mixture 1b sequesters nearly as much carbon dioxide as Mixture 1a.

While Mixture 1a has the propensity to sequester slightly more carbon dioxide, its net overall life cycle emissions are higher due to the initial emissions associated with cement manufacture and the quantity of cement in the mixture. This finding is consistent with the findings of Souto-Martinez, *et al.* [30], who concluded that, concrete elements that sequester the most CO_2 do not always result in the lowest net CO_2e emissions. The net total embodied carbon for Mixture 1a is slightly greater than Mixture 1b, with mean values of 174.8 and 144.8 kg CO_2e per m^3 of concrete, respectively. Accounting for carbonation is shown to significantly reduce the net embodied carbon of both concrete mixtures. This finding emphasizes the importance of quantifying concrete carbonation when making design decisions related to reducing concrete embodied carbon.

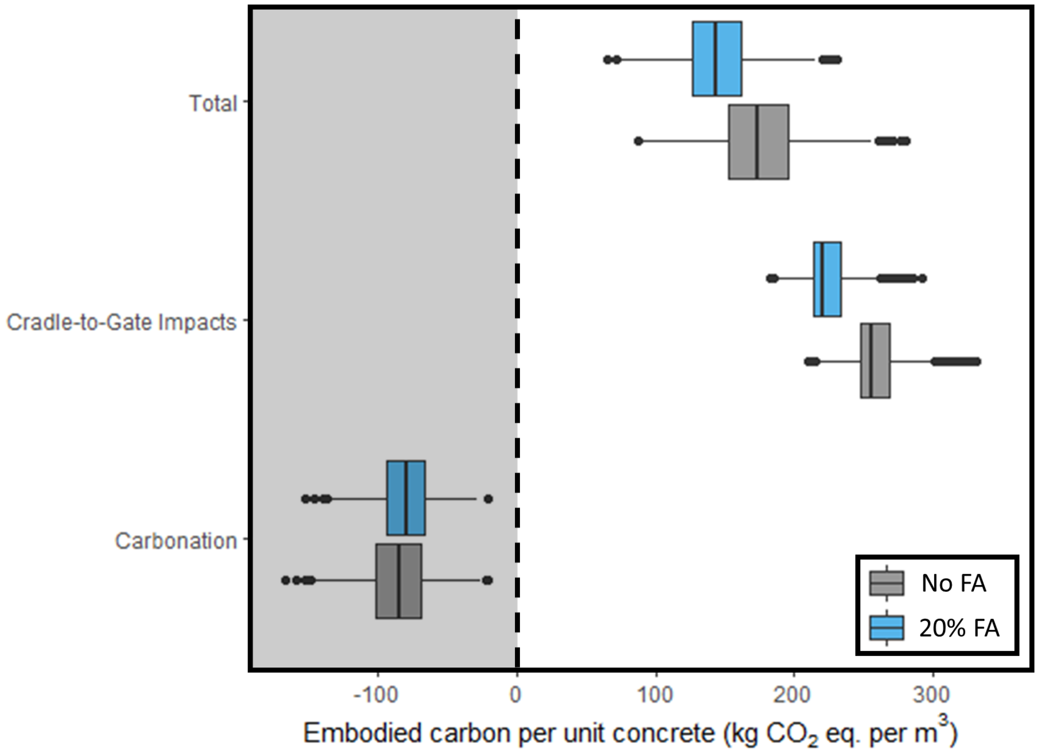

Figure 5. Total embodied carbon of concrete Mixture 1a and Mixture 1b.

The cradle-to-gate results from this study are compared to the results from other cradle-to-gate embodied carbon studies for 21 MPa (3000 psi) concrete of similar mix designs. For instance, the 2016 NRMCA industry-wide EPD for ready-mixed concrete reports a global warming potential of 291 kgCO_2e per m^3 for a 21 MPa mix design with 20% fly ash replacement [79]. Comparing this value to the distribution in Figure 5, we see that the EPD-reported embodied carbon values lie on the higher end of the distribution for Mixture 1b, which is likely due to the use of a relatively old LCI data source for cement from 2010. Similarly, this study reports a cradle-to-gate embodied carbon value of 337 kgCO_2e per m^3 for a 21 MPa mix design with no fly ash replacement [70]. We attribute the comparatively high-embodied-carbon values from these studies to the fact that, over the past decade, carbon dioxide emissions from cement kilns have decreased in the US, due to the adoption of more efficient kiln technology. Thus, older studies tend to report cement emissions that are higher than the cement emissions distribution reported in this study [46].

Lastly, we analyze the same ten mixture designs in terms of embodied carbon, normalized by compressive strength. Figure 6 illustrates that the embodied carbon per strength ratio is not constant. In fact, mixtures with high compressive strength exhibit a lower normalized embodied carbon than their low-strength counterparts. This data may be useful to during building design and enable design requirements to be fulfilled with the lowest possible quantity of embodied carbon.

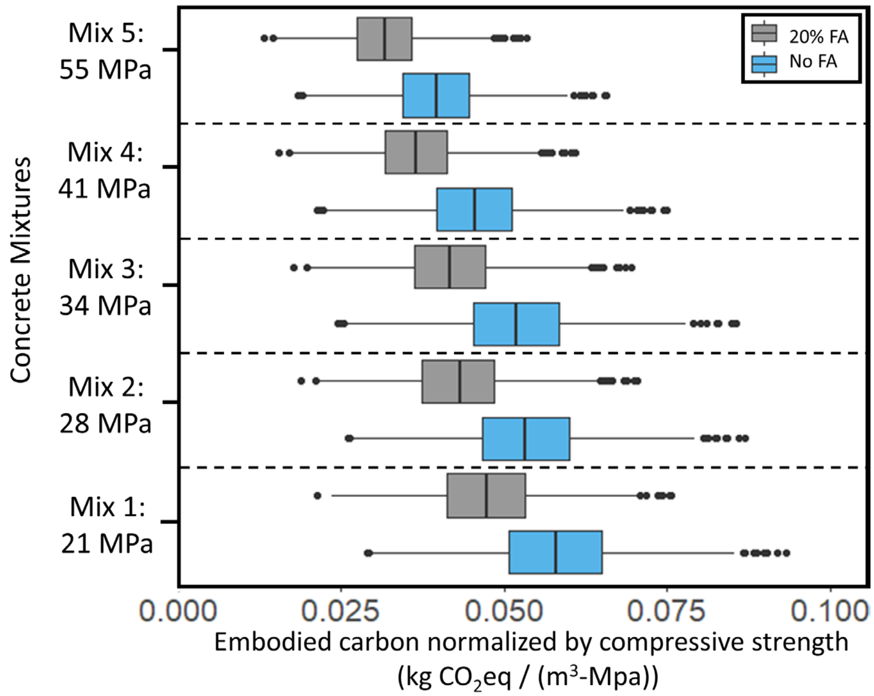

Figure 6. Distributions of embodied carbon, normalized by compressive strength

5. Uncertainty in Fly Ash Transportation Emissions

In this section, results of the novel method for calculating fly ash transportation emissions are analyzed in greater depth. In addition, we investigate the importance of accurate representation of fly ash transportation mode and distance by calculating breakeven distances in which embodied carbon benefits are realized using fly ash—namely the distance in which the benefits are eclipsed by the impacts due to fly ash transportation.

5.1 Novel method for calculating transportation emissions for the nearest fly ash source

As was introduced in Section 3.2.2, the critical assumption for this analysis is that fly ash is transported from the nearest coal power plant to the project location. In other words, the distribution generated from this calculation employs the 'ideal' location from which to source fly ash. **Figure 7** is a raster plot representing the transportation distances to the nearest fly ash source for the contiguous US. Red regions indicate long transportation distances, while blue regions represent locations that have short fly ash transportation distances.

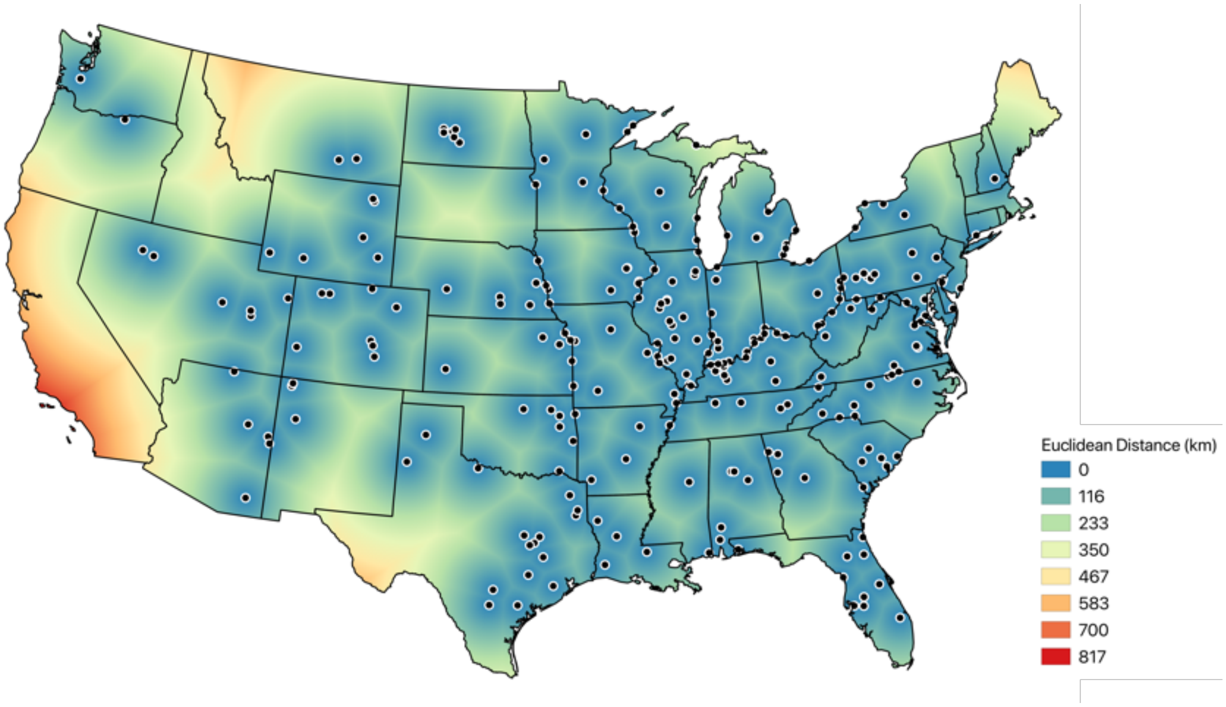

Figure 7. Raster plot representing the distance, in kilometers, to the nearest source of fly ash in the US.

Next, by using the population weighting process, we generate a distribution of emissions due to the transportation of fly ash, which is plotted as a density function in **Figure 8**. In addition, this density function illustrates emissions associated with fly ash transportation for several major US cities. Note the outlier regions on the west coast, which have high population weights and a lack of local fly ash requiring higher fly ash transportation distances. The mean value of this density function is 21.3 kg CO_2e per tonne fly ash. An important result from this analysis is that when the assumption of the *most-local-fly-ash* is true, there is always be an embodied carbon benefit to using fly ash in the US.

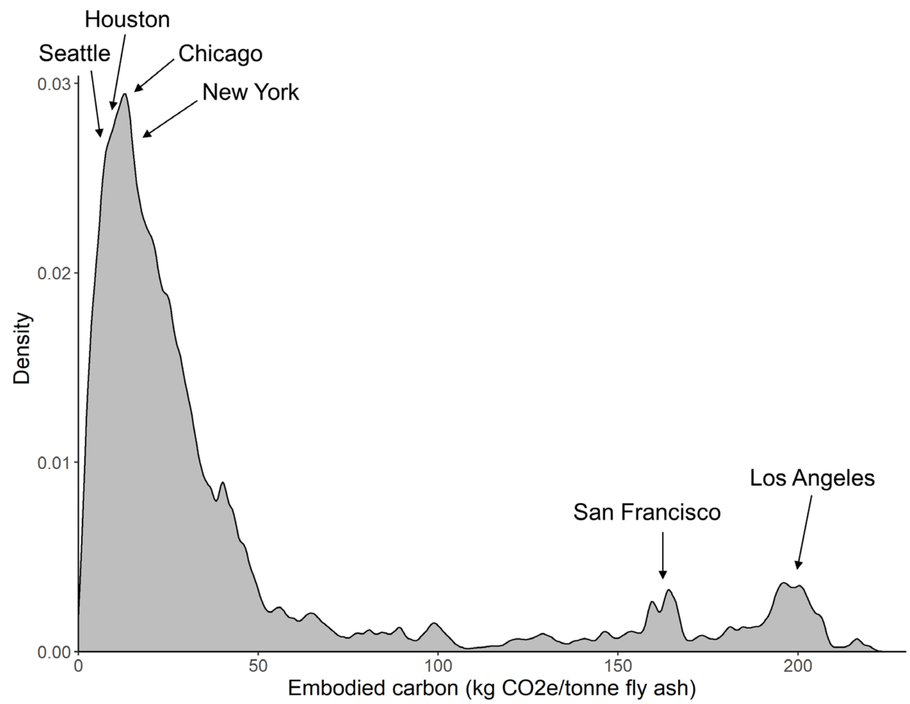

Figure 8. Density function of embodied carbon due to domestic fly ash transportation.

5.2 Comparison of possible fly ash transportation assumptions

In past years, fly ash utilization rates in the US have been low (less than 50% until 2016 [80]), making the *most-local-fly-ash* assumption reasonable. In recent years, however, fly ash utilization has increased, and the price of domestic fly ash has followed suit. Consequently, longer distance transportation and imports of international fly ash have started to become more common. Thus, as fly ash utilization continues to increase, it is likely that the *most-local-fly-ash* assumption will not be accurate moving into the future.

In order to compare the impact of the fly ash transportation, we use various transportation scenarios to compare the *most-local-fly-ash* assumption from the previous section in terms of their embodied carbon impact. As domestic fly ash sources are depleted, the distance to sources of fly ash will increase. Thus, an alternate scenario to consider is the *breakeven trucking* distance. This scenario represents the maximum distance by which it is still beneficial in terms of total embodied carbon to transport fly ash *via* semi-truck. Another alternate scenario is to determine the embodied carbon impact from importing fly ash from international fly ash suppliers. Since China, India, and Turkey are the three largest fly ash exporters [81], we select an example scenario in which fly ash is transported *via* cargo ship from Shenzhen, China to New York, NY. Additional trucking is temporarily ignored for this scenario for the sake of generalization. Furthermore, we temporarily use average embodied carbon values for each impact category in order to compare possible fly ash transportation assumptions.

Figure 9 illustrates the difference in total embodied carbon for each of these scenarios due to transportation differences. In addition, each scenario is compared to the baseline mixture that contains no fly ash in order to determine the relative embodied carbon savings. The dotted regions represent the reduction in life cycle embodied carbon due to carbonation, which is slightly greater for mixtures without fly ash. Therefore, the net embodied carbon is represented by the solid outline. Under the *most-local-fly-ash* assumption, there is a significant benefit to fly ash replacement in terms of embodied carbon. For the *breakeven-trucking* assumption, there is any reduction in embodied carbon is eliminated when fly ash is transported more than 2655 km by truck. Finally, for the *international shipping* assumption, the benefit of using fly ash is eliminated due to the long-distance maritime shipping (and additional trucking will further increase the embodied carbon of this mixture). In fact, the mixture with 20% fly ash has an embodied carbon value 4% higher than the mixture without fly ash.

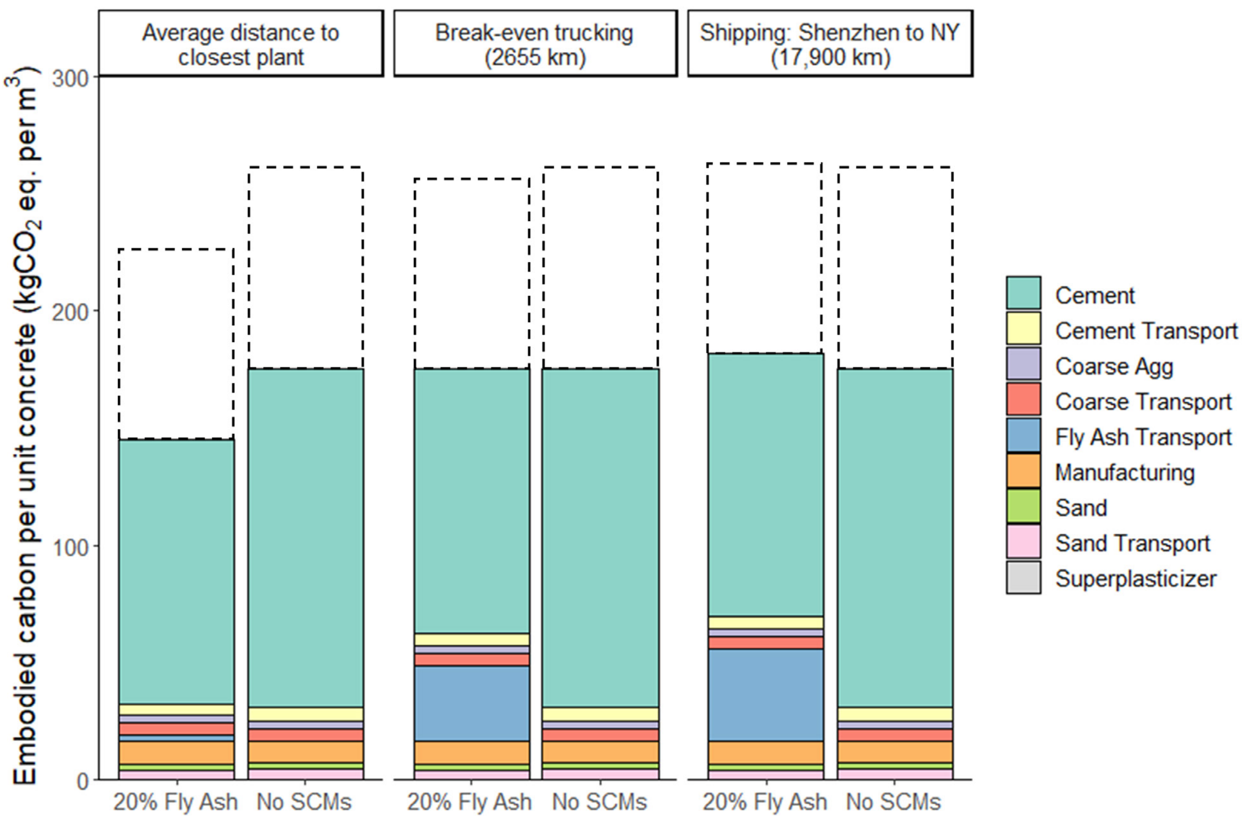

Figure 9. Comparison of embodied carbon for different transportation scenarios and fly ash replacements, using average embodied carbon values for each impact category. The solid outline represents total embodied carbon inclusive of carbonation.

This analysis illustrates the criticality of using the appropriate assumption for fly ash transportation to capture holistic greenhouse gas emissions. In other words, determining the correct fly ash transportation assumption (*i.e.*, mode and distance) is key in assessing the benefit of using fly ash in order to reduce the embodied carbon of concrete mixtures.

5.2.1 International fly ash shipments

Lastly, as international imports of fly ash become more common, it is important to consider how different maritime shipping distances will impact the benefit of fly ash utilization. Utilization of SCMs, like fly ash, in simplified LCAs are often characterized as being able to substantially reduce the embodied carbon of concrete. Such conclusions are due to the fact that many studies assume a relatively low, deterministic value for fly ash transportation distance [33], [34], [82].

However, this analysis has shown that the benefit of using fly ash to reduce concrete embodied carbon is highly dependent on the method of transportation and distance. Figure 10 can be used as a design tool to determine the quantity of embodied carbon savings that have been "spent" by fly ash transport, which is shown as a percent of possible CO_2 savings if there is no transportation. Thus, the 100% dashed line is the "breakeven" line at which all embodied carbon savings from cement replacement have been eliminated because of fly ash transportation. In other words, for transportation scenarios above this dashed line, there is a higher value of embodied carbon for that mixture than for one that uses no fly ash. To use the graph, the gray contours represent maritime shipping distances in kilometers for international shipments of fly ash; the contour line can be followed along its increasing slope for the additional trucking that may occur from transporting fly ash to the appropriate location. At this point on the graph, the percentage of embodied carbon savings that have been "spent" on fly ash transportation can be obtained.

Thus, the major rules-of-thumb for building designers are that fly ash maritime shipping distances greater than 15,110 kilometers will result in higher embodied carbon than a mixture with no fly ash. In addition, shorter shipping distances with additional trucking can negate the embodied carbon benefits of fly ash.

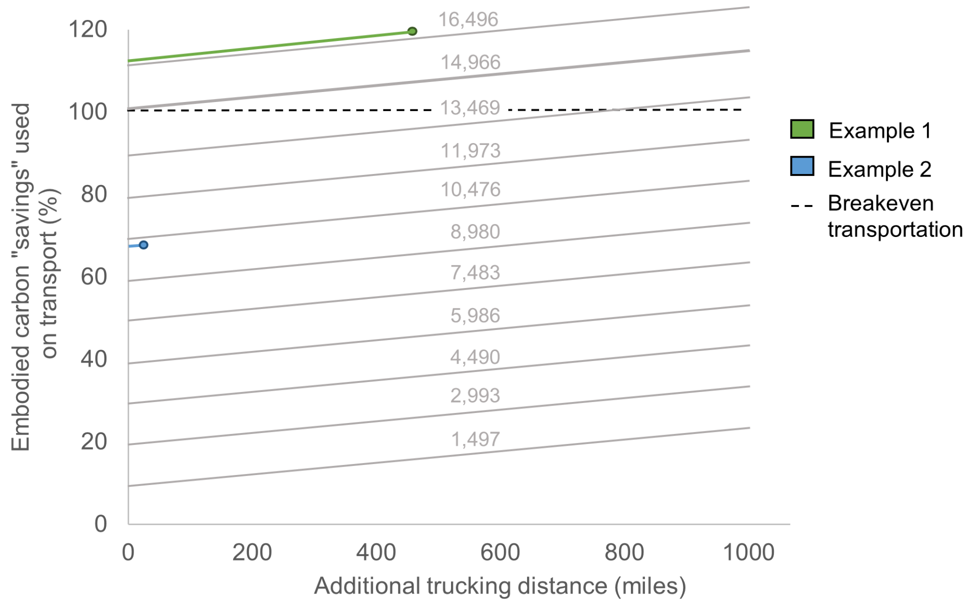

Figure 10. Impact of fly ash maritime shipping and trucking combinations. Example 1 represents maritime shipping from Mumbai, India to Norfolk, Virginia with an additional 442 km of trucking. Example 2 represents maritime shipping from Shenzhen, China, to Oakland, California, with an additional 20 km of trucking.

Two example scenarios are provided from realistic fly ash transportation scenarios from real international coal power plants. **Table 4** shows the scenario information for both examples and the examples are plotted in **Figure 10**. Thus, we can use the graph to determine that, for Example 1, 118% of the embodied carbon savings have been spent on fly ash transportation. For Example 2, 69% of the embodied carbon savings have been spent on fly ash transportation. From these example scenarios we can see that international fly ash imports may significantly reduce the embodied carbon benefit of fly ash. It is important to note that this analysis has not taken into full account the durability benefits gained through the use of fly ash in concrete, which can ultimately change the service life of concrete.

Table 4. International fly ash shipping examples

	Transportation Example 1	***Transportation Example 2***
Coal plant name	*Dahanu Thermal Power Station*	*Shenzhen Mawan Electric Power Limited*
Distributing port name	*Mumbai Port Trust*	*Shenzhen Port Authority*
Receiving port name	*Port of Virginia*	*Port of Oakland*
Receiving city	*Washington D.C.*	*San Francisco*

Total maritime shipping (km)	*16,660*	*10,280*
Total trucking (km)	*442*	*20*

6. Conclusions

The goal of this work was to analyze the variability in embodied carbon of concrete mixtures in the U.S. by attributing probability distributions to each life cycle impact using a variety of life cycle inventory data sources. We find that, on a per mass basis, cement and superplasticizer raw materials have the highest values of embodied carbon; however, on a per volume of concrete basis, cement contributes the vast majority of impacts to estimates of the embodied carbon of concrete.

One key output of this study are the distributions generated herein, which can be used to determine the distribution of total embodied carbon for any concrete mixture design, which before now has not been possible. As a method for demonstrating the utility of the provided distributions for quantifying total embodied carbon, we compare the distributions for ten concrete mixtures of five different compressive strength values ranging from 21 MPa to 54 MPa. For each strength value, two mixtures designs are analyzed - one without fly ash and one with 20% fly ash replacement. We show that, with the assumption that fly ash comes from the nearest source and with the inclusion of concrete carbonation in the LCA system boundary, each mixture with fly ash exhibits a reduced total embodied carbon compared to the complementary mixture without fly ash. Subsequently, in this study, we analyze the transportation assumptions in detail, finding that fly ash transportation is critical in determining when fly ash replacement is beneficial for reducing concrete embodied carbon. We find *via* deterministic calculation that the *breakeven* trucking distance is 2665 km for domestic fly ash, and the break-even maritime shipping distance is 15,100 km for international fly ash. The fly ash transportation design tool developed herein can aid designers in reducing the embodied carbon of concrete by utilizing of fly ash as a supplementary cementitious material, given that the results presented herein illustrate that, depending on transport distance, surefire reductions are not necessarily guaranteed.

This analysis also illustrates two meta-conclusions concerning the way concrete LCAs should be conducted. First, from the probabilistic analysis, it was found that the best way to reduce uncertainty in embodied carbon of concrete is to better understand the emissions associated with cement production (*i.e.*, determine the kiln type and fuels used). In other words, to reduce uncertainty in concrete embodied carbon, the specific cement production processes to make the type of cement used in the mixture must be known. Second, it was found that incorporating carbonation calculations into the LCA system boundary is critical. When concrete carbonation is considered, the calculated reductions in embodied carbon due to fly ash replacement are not as significant compared to a cradle-to-gate embodied carbon comparison. This finding highlights the importance of including carbonation analyses in concrete LCA.

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