

Benchmarking GHG Emissions from California Concrete and Readily Implementable Mitigation Methods

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for Sustainable Transportation

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16. Abstract The demand for concrete, which is conventionally composed of granular rocks (aggregates), water, and Portland cement (as well as other additives depending on desired performance) continues to grow. The manufacturing of Portland cement leads to notable greenhouse gas (GHG) emissions, which has driven interest in alternative concrete mixture designs, cement production processes, and other emissions mitigation strategies. To demonstrate the efficacy of such mitigation strategies, environmental impact assessments are commonly performed. However, examination of the probability that a reduction in GHG emissions will occur given known limitations on data quality and variability in data remains poorly studied. Additionally, the common practice of focusing primarily on GHG emissions can lead to selection of emissions mitigation methods with unintended consequences, such as increases in other environmental impacts. This work models 12 potential concrete mixtures capable of achieving the same concrete strength and three potential GHG emissions mitigation strategies: changing kiln fuel mix, changing electricity mix, and using a carbon capture and storage (CCS) system. Focusing on GHG and air pollutant emissions, both deterministic comparisons of mean emissions as well as the probability that the alternative mixtures and mitigation strategies can reduce emissions is examined. This work shows that, even when mitigation strategies are employed, GHG emissions are correlated to the cement content of the mixture. Additionally, as modeled, CCS leads to mean reduction in GHG emissions of over 80% for all mixtures, but also led to increases in other emissions (i.e., NO _x , SO _x , VOC, CO, PM ₁₀ , and PM _{2.5}). The probability of a reduction in emissions were greatest for GHGs due to the tighter distribution in emissions modeled. Probabilities for reducing other impacts, such as PM ₁₀ and PM _{2.5} emissions, could be improved with better data quality. This work demonstrates how concurrent environmental impact assessment across several impact categories with consideration for uncertainty and variability can be a robust tool for evaluating various mixture designs and environmental impact mitigation strategies.			
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A National Center for Sustainable Transportation Research Report

December 2021

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Benchmarking GHG Emissions from California Concrete Production and Readily Implementable Mitigation Methods

EXECUTIVE SUMMARY

The demand for concrete, which is conventionally composed of granular rocks (aggregates), water, and Portland cement (as well as other additives depending on desired performance) continues to grow. The manufacturing of Portland cement leads to notable greenhouse gas (GHG) emissions, which has driven interest in alternative concrete mixture designs, cement production processes, and other GHG emissions mitigation strategies to decrease associated impacts. To demonstrate the efficacy of such strategies, environmental impact assessments are commonly performed. However, examination of the probability that reduction in GHG emissions will occur given known limitations on data quality and variability in data remains poorly studied. Additionally, focusing solely on GHG emissions can lead to selection of emissions mitigation strategies with unintended consequences, such as increases in other environmental impacts. The goal of this work is to bridge these gaps by addressing uncertainty and environmental impacts beyond GHG emissions to provide a means for benchmarking and improving concrete environmental impacts.

This work models 12 potential concrete mixtures capable of achieving the same concrete strength and three potential mitigation strategies: changing kiln fuel mix, changing electricity mix, and using a carbon capture and storage (CCS) system. Focusing on GHG and air pollutant emissions, both deterministic comparisons of mean emissions values as well as the probability that the alternative mixtures and mitigation strategies can reduce emissions is examined. Deterministic comparisons were made using an environmental impact assessment methodology. This methodology was expanded using distributions of potential emissions to determine the mean potential impacts and the probabilities of achieving those emissions. Three key sources of uncertainty were considered: (1) data variability (e.g., inherent variation in emissions); (2) data uncertainty (e.g., uncertainty stemming from datasets available); and (3) basic uncertainty (e.g., uncertainty attributed to modeling certain types of emissions).

This work shows that, even when mitigation strategies are employed, GHG emissions are correlated to the cement content of the mixture. Additionally, as modeled, CCS leads to mean reduction in GHG emissions of over 80% for all mixtures, but also led to increases in other emissions (i.e., NO_x, SO_x, VOC, CO, PM₁₀, and PM_{2.5}). The probability of a reduction in emissions was greatest for GHGs due to the tighter distribution in emissions modeled. Probabilities for reducing other impacts, such as PM₁₀ and PM_{2.5} emissions, could be improved with better data quality, which would reduce uncertainty and the distribution of modeled emissions. This work demonstrates how concurrent environmental impact assessment across several impact categories with consideration for uncertainty and variability can be a robust tool to aid decision makers in both selecting materials and identifying system-scale process changes to employ to reduce the environmental impacts from cement and concrete production.

1. Introduction

1.1. Infrastructure material consumption and environmental impacts

The demand for construction materials is increasing in both industrializing and industrialized countries [1]. The increase in built-up regions leads to both environmental impacts associated with producing the built systems as well as those associated with land uptake for buildable areas, which can be at the expense of biodiversity and agricultural use [1]. Regardless of level of industrialization, regions dealing with growing urban populations, such as California, have higher cement demand. This consumption of materials to meet growing demand is resulting in substantial anthropogenic environmental burdens [2], [3] from material production and use as well as impacts hidden in supply chains [4]. These environmental burdens highlight the importance of developing robust tools to quantify and mitigate the environmental impacts associated with consumption of infrastructure materials, such as concrete.

Because physical infrastructure is necessary for human society, infrastructure materials represent the largest use of materials by weight globally, which in turn requires vast energy inputs and produces large waste flows [5]. Figure 1 shows trends in wood, steel, and cement production from 1961–2010. As can be seen from the diagram, while there is a slight increase in wood production, there is a greater increase in steel production and an even greater increase in cement production over the over fifty-year span. Portland cement is the most commonly used hydraulic binder in the production of concrete, and as such, it represents only a fraction by weight of the concrete produced. The increasing trend in cement/concrete production is a key driver in its impact on the environment.

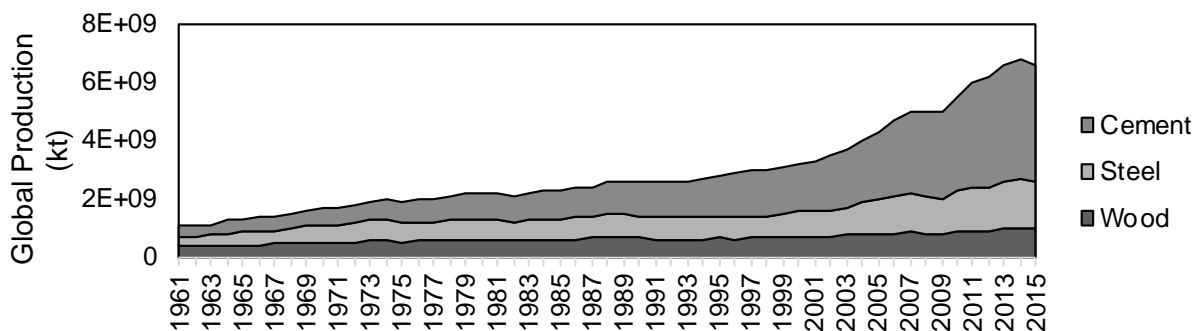


Figure 1. Global production of steel, cement, and wood (in the form of wood-based panels and sawn wood (results based on data from [6]–[9]))

The Intergovernmental Panel on Climate Change (IPCC) estimates that CO₂ emissions will need to reach net-zero 2050 to limit global warming to 1.5 °C [10]. Approximately 64% of global carbon emissions are associated with energy or process emissions [11]. CO₂ emissions resulting from energy and industrial processes can be attributed almost equally to three sectors: industry, buildings, and transport [12]. Of these, buildings and the construction industry have been among the largest consumers of material by weight for almost a century [13]. Of the emissions associated with industry, over 55% of emissions are associated with material

production, with the production of steel and cement constituting almost half of the emissions associated with industry [11]. With the expected doubling of demands for materials by 2050 [14], it is unlikely that process efficiency improvements in material production can occur rapidly enough or be great enough to meet emission reduction needs.

1.2. Cement and concrete consumption and environmental impacts

The demand for hydraulic cement and cement-based materials, such as concrete, has escalated sharply within the past several decades. The production of hydraulic cement now exceeds 4 billion metric tons annually [15].

Large levels of production, fuels needed to meet energy requirements, and limestone decarbonation contribute to sizeable burdens associated with cement and cement-based materials manufacture. While the raw materials for the production of conventional hydraulic cements – mainly limestone and clay – are available in most regions [16], there are high environmental impacts from manufacturing cement. Conventional Portland cement contains finely ground clinker, a kilned and quenched material, inter-ground with gypsum, which aids in setting control. The two main factors driving GHG emissions in the production of conventional cement are associated with its clinker manufacture, which demands: (i) high thermal energy demand to reach the ~1450°C kiln temperature necessary to make reactive products; (ii) raw material-derived CO₂ emissions produced during the calcination process (from limestone decarbonation: $\text{CaCO}_3 + \text{energy} \rightarrow \text{CaO} + \text{CO}_2$).

Cement is used in the production of other materials, most commonly concrete, mortar, and plasters. The environmental impacts of producing these cement-based materials has been of growing concern as they are leading to 8-9% of anthropogenic greenhouse gas (GHG) emissions, 2-3% of energy demand, and 2% of global water withdrawals every year [17]–[19]. Additionally, the air pollutant emissions associated with cement and cement-based materials are substantial: over 7% of anthropogenic nitrogen oxide (NO_x) emissions, over 4% of anthropogenic sulfur oxide (SO_x) emissions, and approximately 5% of anthropogenic particulate matter emissions smaller than 10 microns (PM₁₀) [20]. These high environmental impacts are of concern as industrializing countries require concrete to build up their infrastructure and industrialized countries continue to demand concrete for infrastructure maintenance and repair.

Challenges in reducing energy-derived emissions and raw material-derived emissions have sparked several research efforts into mitigation strategies. The use of mineral additives and alternative cements is a critical step in reducing CO₂ emissions from Portland cement [21]. Mineral additives can be used to reduce the demand for cement by partially replacing it and contributing to hydration products and/or improving particle dispersion, such that less cement is necessary to achieve similar properties. This replacement can occur at a cement manufacturing plant, but as is more common in the United States, the blending in of these mineral admixtures can also occur during concrete batching. The properties of mineral admixtures vary from being pozzolanic (i.e., a material that is not cementitious on its own, but reacts with cement to produce desirable properties in concrete) to cementitious (i.e., possesses some cementitious properties) to inert mineral fillers [22]. While the application of such

additives has been common practice in the manufacture of concrete to achieve certain desired properties, such as reduced heat of hydration, the efficient use of these additives to provide similar performance while reducing GHG emissions from concrete production is a current focus of research [23].

1.3. Environmental impacts of cement and cement-based materials

1.3.1. Greenhouse gas emissions

While over 50% of the emissions from cement clinker manufacture are associated with the calcination process, fuel selection for these kilns plays a significant role in emissions. The most used fuels in cement kilns are coal, oil, gas, biomass, and wastes [24]. Each of these has different emissions associated with their combustion ranging from approximately 0.04 kg (for some derived gases) to 0.26 kg (for blast furnace gas) of CO₂ emissions per MJ of energy [25]. While the use of exclusively biomass – for which a negligible CO₂ emissions are often modeled to reflect the effects of photosynthesis – may not be feasible, there is the potential to select fuels for kilns to mitigate CO₂ emissions; the International Energy Agency (IEA) targeted a 12% reduction in CO₂ emissions through fuel switching in their 2018 cement roadmap [26].

1.3.2. Aggregates

The exorbitant demand for resources and potential implications on resource scarcity from the production of concrete have been a growing topic of interest. This interest in large part stems from the high levels of aggregates needed in the production of concrete – for every kg of cement used in a m³ of concrete, 4 to 10 kg of aggregates are used [27]. While aggregates are commonly considered to be a widely available resource, there have been instances of resource scarcity noted for these materials [28]. The over extraction of sand and unregulated aggregate quarrying can lead to over-exploitation of resources, cause ecosystem damage thus affecting biodiversity, and have potential cascading affects that impact human well-being including affecting natural land barriers and warring by competing acquisition parties [29]. This issue is compounded by the fact that in many regions, aggregate use is not monitored as well as cement, mineral admixtures, or chemical admixtures because aggregates are less commonly a traded commodity. While burdens on biodiversity and human well-being can vary by region, more easily quantifiable factors such as particulate formation and energy demand for the excavation and processing of these resources is often not accounted for in environmental impact comparisons.

1.3.3. Water

Issues surrounding the availability of water, including when and where it is needed, are of global concern [30]. In many regions around the world, water withdrawal exceeds the naturally renewable water supply [31]. Much of anthropogenic water withdrawal is used in agriculture; however, approximately 20% is extracted for industrial purposes [32]. In cases where water use is potable, industrial demand could be placing additional stress on a scarce resource. While terminology on the environmental impacts associated with water use can vary, herein, we refer to water demand to discuss either water consumption or water withdrawals. Water

consumption refers to the removal of water from a reservoir and using the water such that it does not directly return to that same reservoir. Water withdrawal refers to the sum of water removed from a reservoir, whether it is returned to that same reservoir or not.

Water is a primary constituent in concrete, as it facilitates the hydration reactions with hydraulic cement to form the rock-like cement-based materials. The demand for water as a constituent on a per mass basis can be as high as cement consumption [17]. Yet, this bound water in the cement-based materials product is just a small fraction of the water consumed in the production of cement-based materials. Water used for processes and for energy resources constitute approximately 87% of the water consumed for concrete and mortar manufacture, and they are responsible for even higher fractions of water withdrawals [18]. The energy-related water consumption is a function of energy and resource requirements; both electricity or thermal energy inputs have associated water consumption [33].

While the cement binder is the primary focus of GHG emissions mitigation strategies, the complexity of water demand as it relates to the supply chain makes pathways to lowering water demand more difficult. For example, in cement production, differences in kiln type informs both utilization of water in the pre-blending process and energy efficiency [34], where both energy type and energy quantity will drive additional water demands [35]. While kiln efficiency and fuel resources affect embodied energy and CO₂ emissions as well, mitigation strategies to reduce water demand do not always align with those to reduce these other impacts (e.g., a high-water demand fuel resource may also be a low CO₂ emitting one). Additionally, water consumption for dust suppression results in demand at quarrying sites and electricity requirements contribute notably to water demand [18]. As such, in drought-prone regions, like much of California, careful attention must be paid to supply chain management to accurately track and reduce water demand.

1.3.4. Particulate matter emissions

The emissions of particulate matter (PM), especially of small particle sizes, are of concern due to the effects of inhalation on human health, such as respiratory infection, pulmonary disease, and lung cancer, among others [36]. In 2013, over 80% of the world's population was living in areas with levels of PM that exceed World Health Organization guidelines, with the highest increase in PM emissions in World Majority countries [37]. Human intake fraction of air pollutant emissions, such as PM, is necessary to determine human health impacts. For PM emissions, driving factors that influence intake fraction include the emissions release height (e.g., at ground level, from an emissions stack) and “archetypal” environment (e.g., remote, rural, or urban) [38].

For cement and concrete, there are several sources of PM and dust along the supply chain. These include emissions from quarrying and crushing, transportation, material grinding, material storage, clinker production, and batching [39]–[41]. In the production of cement, process stages including quarrying, transportation, raw meal preparation, and grinding account for approximately 2 to 2.6 kg of PM emissions per metric ton of cement produced [34]. A much greater quantity of PM emissions in cement production is cement kiln dust, which leads to

approximately 39 kg of PM per metric ton of cement produced [34]. These particles are particularly of concern if inhaled because of their composition, i.e., silica content and heavy metal compounds (to be discussed in greater detail in the next section), which have notable human health implications [41]. However, this cement kiln dust can be reused in the production of more clinker if it has appropriate alkali content, thus reducing emissions [39]. Because of the ability to use controls such as filters and this reuse of cement kiln dust, the majority of PM emissions from cement production, aggregate acquisition, and concrete batching are fugitive emissions, such as associated with material transfer, milling operations, and transportation [39], [40], [42]. In addition to these fugitive emissions, there are additional PM emissions from the different energy sources utilized: both fuels for generation of electricity and for thermal energy are known to release PM emissions [36], [43], [44].

Quantities of emissions and contributions to local air quality concentrations can be larger or smaller based on local regulations, geography, topology, and wind movement. Considering the effects of policy decisions, the impacts for single country studies indicate the potential magnitude of cement production related PM emissions. The cement industry in China resulted in more than a quarter of the country's PM smaller than 2.5 microns ($PM_{2.5}$) and PM smaller than 10 microns (PM_{10}) in 2005 [45]. In the United States, cement has been reported as one of the ten largest sources of criteria air pollutants for the industrial sector [46].

1.3.5. Heavy metal emissions

Emissions of heavy metals, not unlike those of PM, are of concern due to local impacts on human health. High levels of metals can result in kidney damage, neurological damage, various cancers, and DNA damage, among others [47], [48]. While the harmful effects of increased levels of metal exposure on human health have been known for a long time, exposure continues and is increasing in several areas; although, exposure in the most economically developed parts of the world has seen a decline over the past 100 years [47]. Humans can be exposed to heavy metals in a variety of ways, but atmospheric emissions and inhalation are considered to be of the greatest concern due to factors including the potential for widespread dispersion [47].

In the production of cement, heavy metal emissions can arise from the raw materials and from the fuels used. The quantity of heavy metal emissions from cement can be highly dependent on production methods [48]. The utilization of alternative fuels, such as tires or industrial waste streams, and the use of raw materials that contain heavy metals, such as natural resources with trace metals, both can contribute to heavy metal emissions in cement manufacture [34], [49]. Cement production has been noted as one of the main sources of heavy metal emissions in Europe [50] and has been reported as leading to the emissions of 17 types of metals [39]. Notably, cement production is considered to be a main source of As, Cd, Cr, Cu, and Hg emissions in some regions [51]. Considering that As, Cd, and Hg are among the most threatening metals to human health [47], monitoring and control of heavy metal emissions from cement plants should be implemented where it is not already in use.

While consideration for cement production on heavy metal emissions does not appear to be under debate, the quantity of emissions does [48]. A report by the United Nations Environment Programme showed that cement production was responsible for approximately 9% of anthropogenic Hg emissions [49]; however, those values do not match industry's experience in emissions or reporting [52]. While it is clear that the dust from cement production can contain these heavy metals [48], several metal species are known to become chemically incorporated into the clinker crystal matrix or absorbed in cement kiln dust [34], [52]. Further, appropriate control devices can be used to mitigate heavy metal emissions [34], [52].

1.4. Summary of review and overview of work conducted

As the production of concrete and other cement-based materials increases to meet the demand for infrastructure, greater efforts have been made to reduce the environmental impacts associated with these construction materials. The benefits of these new technologies and strategies are often presented with some quantification of the environmental impacts from the materials. However, the anticipated likelihood of improvements is often unclear due to factor such as variations in data quality and unaddressed uncertainty across studies. Additionally, often only the targeted environmental impact is evaluated. Selection and evaluation of a single impact could lead to unintended consequences. For example, evaluating only cement and the associated GHG emissions could lead to increases PM emissions from increased amounts of another constituents. It could also lead to unintended increases in other environmental impact categories that were not assessed, for example water consumption. Further, the uncertainty associated with quantitative impact assessments could lead to decision without considering the probability of actually obtaining the desired outcome.

In this work, the environmental impacts of concrete mixtures are evaluated as are three potential mitigation strategies: changing kiln fuel mix, changing electricity mix, and using a carbon capture and storage (CCS) system. To compare results across mixtures and mitigation strategies, the probability of reducing GHG and air pollutant emissions is quantified. To determine these probabilities, environmental impact assessments are performed considering variability (i.e., inherent variation in flows that could affect impacts studied through the supply chain) and uncertainty (i.e., a function of data quality for the system modeled); here, uncertainty is discussed both in terms of data uncertainty, which directly reflects data utilized, and basic uncertainty, which reflects uncertainty attributable to the type of flow studied. Findings will support informed decision making for mitigating environmental impacts from cement and concrete production.

2. Methods and Materials

2.1. Deterministic environmental impact assessment

In order to quantify the environmental impacts of concrete mixtures, environmental impacts were examined using a deterministic method and a probabilistic method. The implementation of the probabilistic method used herein, which captures data variability, data uncertainty, and basic uncertainty, is outlined in [53]. These parameters reflect known drivers in emissions distributions (e.g., [54]) and commonly accepted sources of data uncertainty [55], [56]. These parameters were used to determine distributions in environmental impacts through Monte Carlo simulations accounting for each variable and uncertain parameter. The distributions were used to assess differences in environmental impacts based both on mean comparisons (deterministic determination of reduction) and the frequency with which reduced emissions would be achieved (probability of reduction). Input and output flows were used to form a deterministic model based on the same literature sources, and it included additional consideration for water consumption and withdrawal. A summary of the deterministic flows, the sources for the quantities modeled, and assumptions made are presented in Appendix A. In this work, the inventories for concrete production are used to focus on example mixtures produced in California with the discussion focused on GHG, NO_x, SO_x, particulate matter smaller than 2.5 microns (PM_{2.5}), PM₁₀, volatile organic compounds (VOCs), lead (Pb), and carbon monoxide (CO) emissions. The GHG emissions (from CO₂, CH₄, and N₂O) are quantified in terms of CO₂-eq using the Intergovernmental Panel on Climate Change (IPCC) 100-year global warming potential (GWP) weighting scheme [57]. The scope of the model focuses on cradle-to-gate stages, considering Portland cement production, mineral admixture production, aggregate acquisition, chemical admixture production, concrete batching, material transportation, thermal energy (for kiln processes), and electricity production. Key modeling assumptions are summarized below (based on the explanation in [53]).

The model employed a Portland cement production model that considered the kiln efficiency based on [58] and allows for selection of kiln types with the subsequent electricity required based on [34]. Emissions from limestone decarbonation were determined using stoichiometry, assuming a 65% lime content in clinker and 5% gypsum in the cement. The thermal energy required for kilns were modeled based on [20]. Air emissions are based on [39], [58], [59]. To simplify modeling efforts in this work while exemplifying benefits from mineral admixtures, only fly ash is considered as a mineral admixture with no additional energy requirements as reported by [60]. Aggregates, both coarse and fine, were modeled with the energy demand from and air emissions from [43]. The model deterministic model developed in unison with this work allows for evacuation of mixtures containing several different SCM, mineral admixtures, and chemical admixtures. However, only fly ash was evaluated in this work. Concrete batching energy is based on [61] and emissions from batching [40]. Transport emissions from truck, rail, and ship using [62]–[64]. Electricity emissions were based on the California electricity grid mix [65], and GHG emissions and air pollutants are based on the electricity mix are from [20]. The model uses the described inventory, the flows from the concrete constituents and production processes are summed across materials and processes. The GHG and air pollutant emissions data are then used to quantify the impacts for the production of a given concrete mixture.

Building from the same scope as the deterministic considerations, distributions for environmental impacts were determined by considering variability in raw material inputs, in energy production, and uncertainty associated with emissions. The model used here, formally presented in [57], employs distributions of inputs and variability in emissions outputs across energy production, material acquisition, transportation, concrete consistent production (e.g., cement, fly ash, aggregates), and concrete batching (as shown in Figure 2). When such distributions were not directly reported, data from literature was used to create discrete (when a single data point was available), linear (when two data points were available), triangular (when three data points were available), or lognormal (when four or more data points were available) distributions [57]. This work assumes no variability or uncertainty from variations in constituent masses.

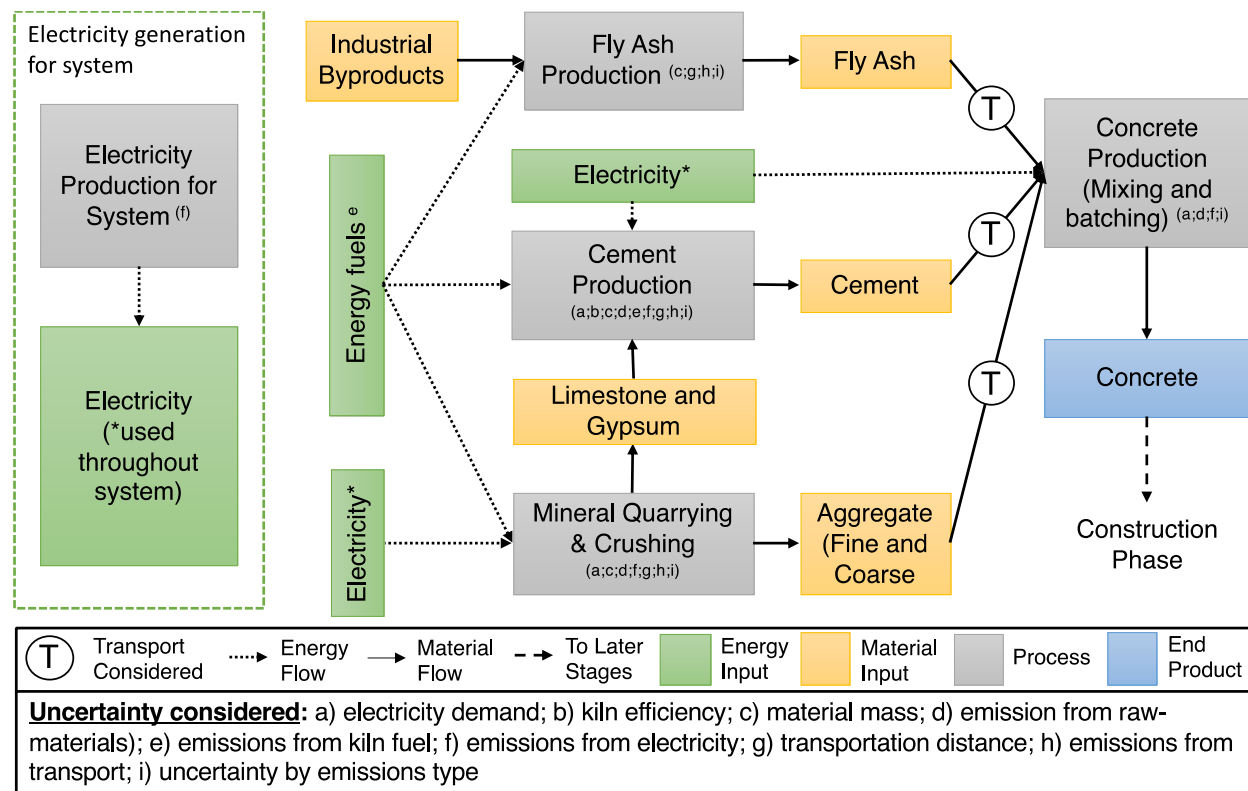


Figure 2. Diagram of system and processes modeled, including notation of the uncertainty considered by the model (based on Miller, 2021 [57]).

The system modeled for concrete production is shown in Figure 2, with notation indicating the sources of variability and/or uncertainty considered in the model at each phase. Assumptions made in the model for the various production processes, (e.g., kiln fuel mix, kiln efficiency, transportation distances, variability in emissions) are based on values from literature and are summarized in Table 1. Using the model, three production-related mitigation strategies were considered as examples: (1) changes to kiln fuel mix (assessed by using natural gas instead of higher emitting fossil fuels); (2) changes to electricity grid mix (assessed by replacing fossil fuel-derived energy with wind energy); and (3) CCS (assessed by modeling amine scrubbing with an

efficiency of 90% of CO₂ from the cement kiln flue gas and with an energy requirement of 2 GJ per tonne CO₂ captured) [57]. While there are several means for CCS technologies to be implemented, this work considers recapture based on Hills *et al.* [66] and does not model utilization of CO₂ in the cement-based materials. CO₂ utilization was considered outside the scope of work, and it is noted that such utilization could lead to varying environmental benefits [67]. It is also noted that non-hydraulic cements, such as those that can solidify by reacting with CO₂, have been considered as a means to reduce CO₂ emissions from the cement and concrete industries [68]; however, these too are outside the scope of this work.

Table 1. Key data, assumptions, and sources for modeling uncertainty in concrete mixture production from Miller, 2021 [57]

Associated Input	Explanation of value, assumption, or calculation	Source
Cement*		
Electricity demand	based on demand for various production processes across the U.S.	[34]
Clinker and gypsum amounts	95% clinker and 5% gypsum	[69]
Lime content in clinker	65% lime-based clinker (CaO)	approximation
Production State	California / in state, as California produces enough cement to meet in-state demand	approximation based on [70]
Transport Distance	150 km, assumed in state	[63], [64]
Kiln Types **	~15% dry kilns, ~85% pre-calcining kilns	[34]
Clinker production emissions	Various values for each category	[39], [58], [59]
Kiln Efficiency (fuel sources) **	Fuel types and amounts used in kilns in California	[71]
Kiln Efficiency (lower heating values of fuels) **	Values for fuels and waste combustions used as thermal energy sources in kilns	[72], [73]
Kiln Efficiency (clinker production) **	Amount of clinker produced in California	[71]
Variability of kiln efficiency **	+/- 5%, based on global data	[58]
Variation for GHG and air pollutant emissions	Distribution of emissions from combustion of kiln fuels	[25], [43], [72], [74]
Coal Fly Ash*		
Energy demand for production	0 kWh	[60]

Associated Input	Explanation of value, assumption, or calculation	Source
Transportation Distance	1000 km, by rail; assumed from closest coal-based electricity plants	[63], [64]
Aggregates*		
PM10 emissions	equivalent to source	[43]
PM2.5 emissions	no data available, assumed to be the same amount as PM10 emissions in reference	assumption, mass from [43]
Transportation	75 km, assumed in state	[63], [64]
Emissions from processing	equivalent to source	[75]
Water		
Energy demand	assumed to be negatable	assumption
Concrete Mixing and Batching*		
Electricity demand	assumed values from LBNL report	[61]
* California electricity (used for processes and constituents above)		
Electricity Grid Mix	Assumed California 2016 average grid mix	[65]
Variability in GHG emission	Weighted average from U.S. generation data	[44]
Variability in air pollutants emissions	Weighted average from U.S. generation data	[44]
Variability in Pb emissions		[76]
Energy Demand Variability	variability of demand for cement production from 1990-2016	[58]
<p>** The kiln efficiency (by kiln type) was determined from the energy consumption of kilns, based on fuel type demand and lower heating values, and divided by the clinker production. A triangular distribution was produced using the assumed variability of +/- 5%, as listed above</p> <p>NREL, National Renewable Energy Laboratory; USEPA, United States Environmental Protection Agency; GNR, (the Cement Sustainability Initiative's) Getting the Numbers Right; CARB, California Air Resource Board; GREET, Greenhouse gases, Regulated Emissions, and Energy use in Technologies model; CEC, California Energy Commission</p>		

In the model, the data uncertainty and basic uncertainty were quantified using a pedigree matrix following the works by Frischknecht *et al.* [55] and Weidema and Wesnæs [56]. These authors propose modeling data uncertainty considering several factors, including a “basic” uncertainty associated with the emissions type. Monte Carlo simulations (n = 100,000) were

used to create distributions of environmental impacts for each stage of production to produce one cubic meter of concrete. Herein, emissions distributions are discussed in terms of the full distribution, the distribution associated with only considering data variability, the distribution associated with basic uncertainty, and the distribution associated with data uncertainty. This separation in distribution sources is used to inform findings.

2.2. Concrete mixtures modeled

To evaluate the probability of reducing emissions from concrete production, representative concrete mixtures that meet the California Department of Transportation (Caltrans) 2018 Structural Concrete Specifications [77] were evaluated (Table 2) with the mixture proportions as determined by Miller, 2021 [57]. The representative cases selected were the high (CA-HC) and low (CA-LC) cementitious material content (15% fly ash, 85% Portland cement). An alternative set of mixtures with 300 kg of cementitious material per cubic meter of concrete (300C-) with 0-30% fly ash were also studied. Initial mixtures were from Miller, 2021 [57]; to assess the effect of varying fly ash content, intermediary mixtures were extrapolated from those original mixtures.

Table 2. List of mixture names and the corresponding mixture proportions (provided in kg of constituent per cubic meter of concrete).

Mixture	Portland cement (kg/m ³)	Fly ash (kg/m ³)	Water (kg/m ³)	Fine aggregate (kg/m ³)	Coarse aggregate (kg/m ³)
CA-HC*	403	71	448	932	405
CA-MH	387	68	431	957	416
CA-MD	372	66	413	982	427
CA-ML	356	63	396	1007	438
CA-LC*	340	60	378	1032	449
300C-0FA	300	0	300	1158	503
300C-5FA	285	15	294	1162	505
300C-10FA	270	30	289	1165	506
300C-15FA*	255	45	283	1169	508
300C-20FA	240	60	277	1173	510
300C-25FA	225	75	272	1176	511
300-30FA*	210	90	266	1180	513

*Indicates mixture based on Caltrans Stand. Spec. [77] used by Miller, 2021 [57] to estimate similar strength concrete mixtures; all others were linearly interpolated or linearly extrapolated from these values.

The intermediary mixtures were determined through linear interpolation between the mixtures provided in Miller, 2021 [57]. Between CA-HC and CA-LC mixtures, three intermediaries were determined: CA-MD, CA-MH, CA-ML. These mixtures were selected such that the cementitious contents were evenly spaced between the HC condition and the LC mixtures. Linear interpolation was also used to determine 300C- mixtures with 25% fly ash and 20% fly ash between the 30FA and 15FA cases. Additionally, cases for fly ash content down to 0% were linearly extrapolated at 5% increments from the defined 15% and 30% fly ash mixtures. All mixtures are shown in Table 2.

3. Results

To present results, the CA-HC mixture with no manufacturing improvements is modeled as a baseline. Using this mixture as a baseline facilitates comparisons of the influence of using different constituents (represented by the other 11 mixtures) and the three emissions mitigation strategies (the manufacturing improvements). Findings are discussed below. Additionally, the mean reductions and the three mitigation strategies considered, along with the associated probabilities of reduction, are presented in a combined figure in Appendix B.

3.1. Changing concrete constituents alone (no manufacturing improvements)

To evaluate the effect of changing concrete constituents, without any manufacturing improvements on mean reductions in GHG emissions and air pollutant emissions, as well as the probability of lower emissions, the 11 other concrete mixtures are compared to CA-HC (see Figure 3). The mixtures with lower Portland cement content exhibit lower GHG emissions. This is due to the majority of GHG emissions in concrete coming from the cement production process [57], [78]. Likewise, the mean emissions of NO_x, SO_x, CO, VOCs, and Pb all lower as the cement content of the mixtures decreases. PM_{2.5} and PM₁₀ emissions remain nearly constant for the CA-LC, CA-ML, CA-MD, and CA-MH cases. However, there are moderately lower PM₁₀ emissions for the 300C- mixtures with higher fly ash content.

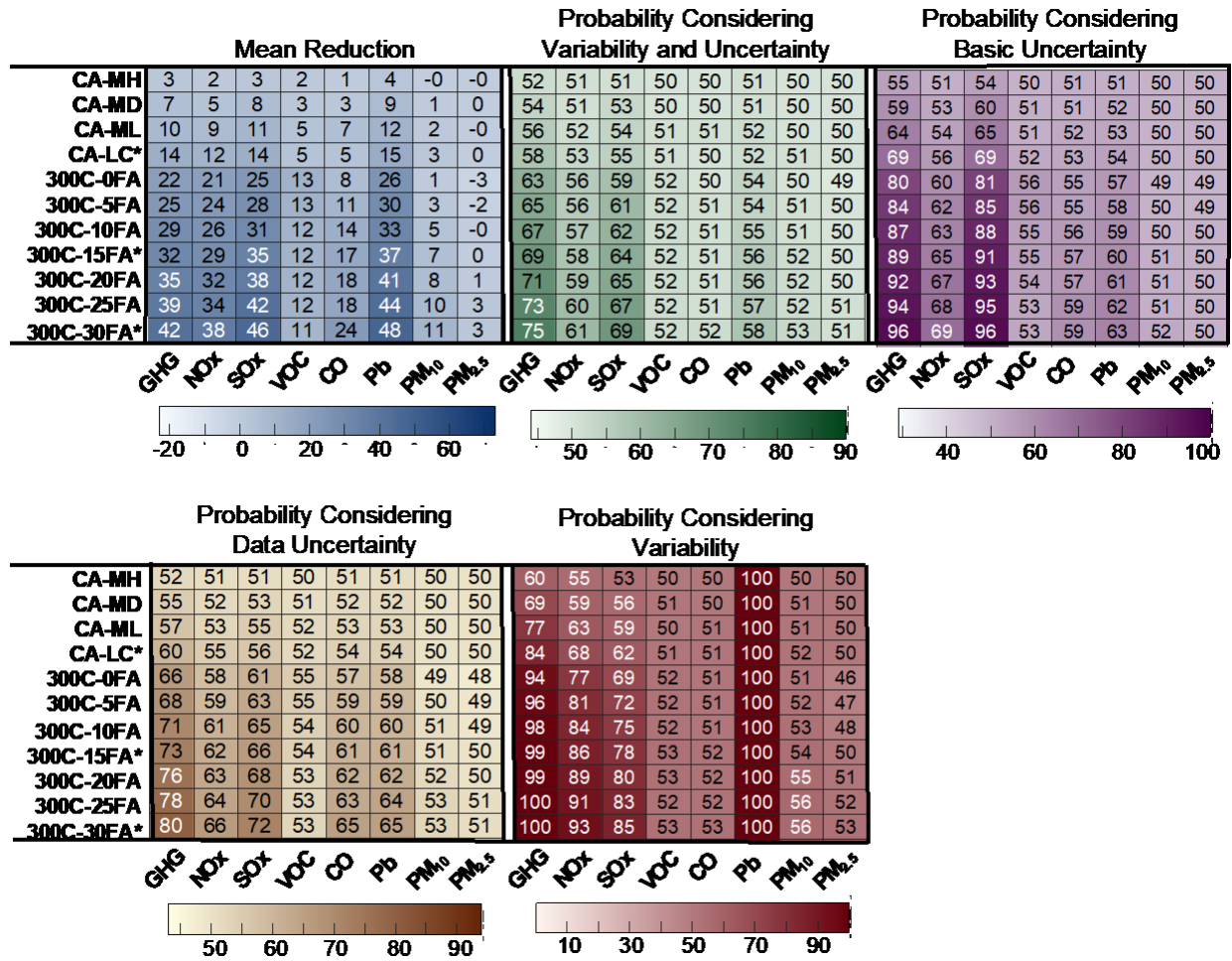


Figure 3. The mean reductions for mixtures without mitigation strategy compared to the CA-HC case and the probabilities of the mean reduction considering variability and uncertainty, only basic uncertainty, only data uncertainty, and only variability.

Aggregate mass remains nearly constant across mixtures with the same cementitious content; however, lower cementitious content mixtures were modeled with greater aggregate content to maintain the same volume. Shifts in aggregate demand between mixtures leads to certain changes in emissions. For example, compared to the CA-HC, the 300C- mixtures require greater coarse aggregate (~24% higher) and fine aggregate (~25% higher) contents, which would be associated with greater production and transportation emissions to supply aggregates. This change would lead to an alteration in the source of PM emissions (i.e., a lower ratio coming from thermal fuels relative to quarrying), while leading to similar total emissions.

The probability of achieving lower emissions with different concrete mixture proportions, shown in Figure 3, indicates that GHG emissions can be reduced with a relatively high probabilities through use of less Portland cement. Miller (2021) notes that the reason for this is two-fold: (1) as Portland cement content changes, there is a high magnitude in of GHG emissions reduction relative to the other emissions, leading to a higher probability of a

reduction occurring; (2) there are tighter distributions for GHG emissions relative to other emissions (Miller notes that this is due to better data quality) [57].

3.2. Kiln fuel switching

The mean reduction in emissions achieved between concrete mixtures when higher GHG emitting fossil fuels are replaced with natural gas to meet thermal energy demands in the cement kiln is shown in Figure 4. Changing the kiln fuel mix only led to slightly greater reductions in emissions relative to the comparisons drawn with no fuel improvements. The exception to this trend is for CO and SO_x emissions. For CO, the mean emissions increased approximately 4% for CA-HC and approximately 2% for CA-ML. For SO_x emissions, mean reductions were 19-33% greater than when no kiln fuel switching occurred. This change was largest for the CA-HC, CA-MH, CA-MD, and CA-ML, and CA-LC mixtures. As discussed above, this is due to the higher cement contents compared to the 300C- mixtures.

The change in kiln fuel increased the probability emissions reduction would occur when considering basic uncertainties and variability associated with GHG and SO_x emissions, especially amongst the CA-HC, CA-LC, and related mixtures. However, little change occurred in the data uncertainty and, thus, the combined probability of reduction considering both variability and uncertainty only increased by 2-8% for those impact categories.

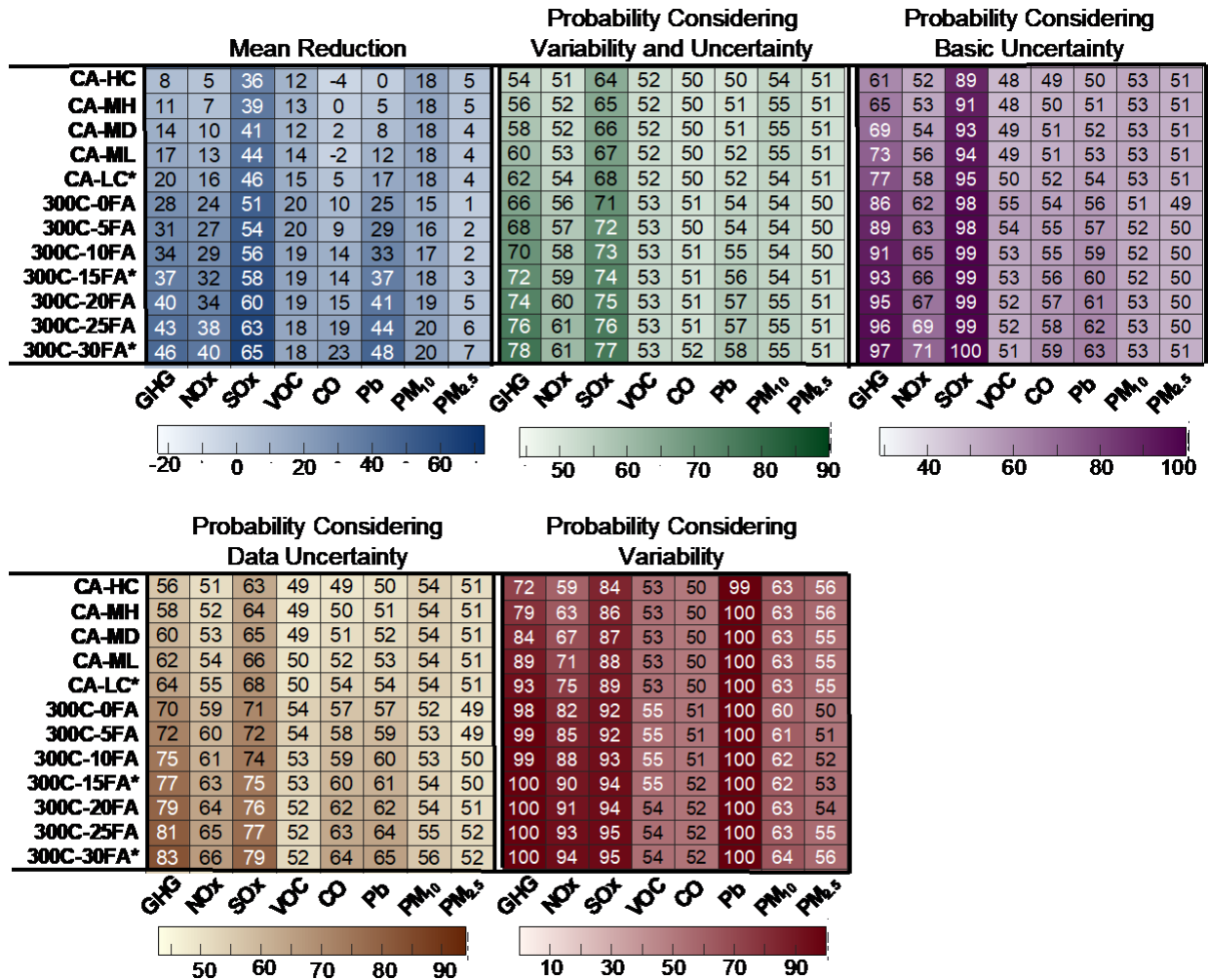


Figure 4. The mean reductions for mixtures with all kiln fuel fossil fuels changed to natural gas compared to the CA-HC case with no mitigation strategy and the probabilities of the mean reduction considering variability and uncertainty, only basic uncertainty, only data uncertainty, and only variability.

3.3. Electricity mix switching

To assess the potential benefits of using lower GHG emitting electricity sources, the electricity mix throughout the concrete supply chain was modeled as though fossil fuel energy sources were replaced by wind energy (Figure 5). The average electricity mix in California was used in initial modeling, and because it has a relatively high percentage of renewable and carbon-neutral energy, the emissions for this mitigation strategies changed very little compared to the cases without mitigation strategies [34]. However, in an area with a higher emissions grid, the benefits from this strategy would be expected to be greater.

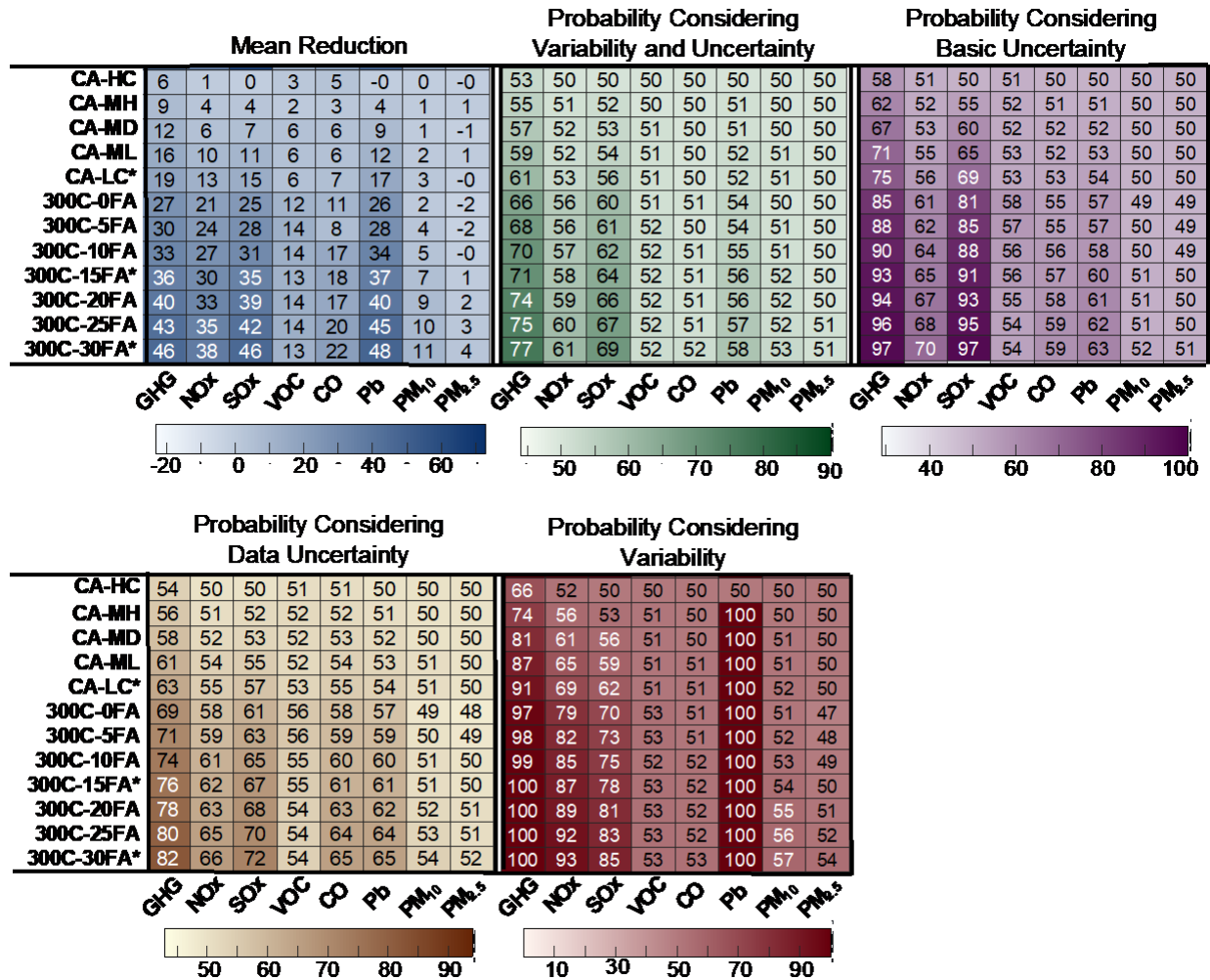


Figure 5. The mean reductions for mixtures with all fossil-fuel electricity changed to wind-based electricity compared to the CA-HC case with no mitigation strategy and the probabilities of the mean reduction considering variability and uncertainty, only basic uncertainty, only data uncertainty, and only variability.

3.4. Carbon Capture and Storage (CCS)

The potential of using CCS technologies during cement production to reduce emissions attributed to concrete mixtures was modeled. The mean reduction of emissions and associated probabilities of emissions mitigation are shown in Figure 6. While the kiln fuel and electricity mitigation strategies led to small changes in the mean reductions, larger reductions for GHG emissions were found from the CCS system modeled. This shift is due to the CCS being modeled as capturing 90% of kiln CO₂ emissions. The difference in GHG mean reductions compared to the cases with no mitigation ranged from 31-59% larger reductions. However, due to the increase in energy required for the CCS process, some of which was modeled as from fossil fuel resources, the SO_x, VOC, and CO emissions increased for the many of mixtures.

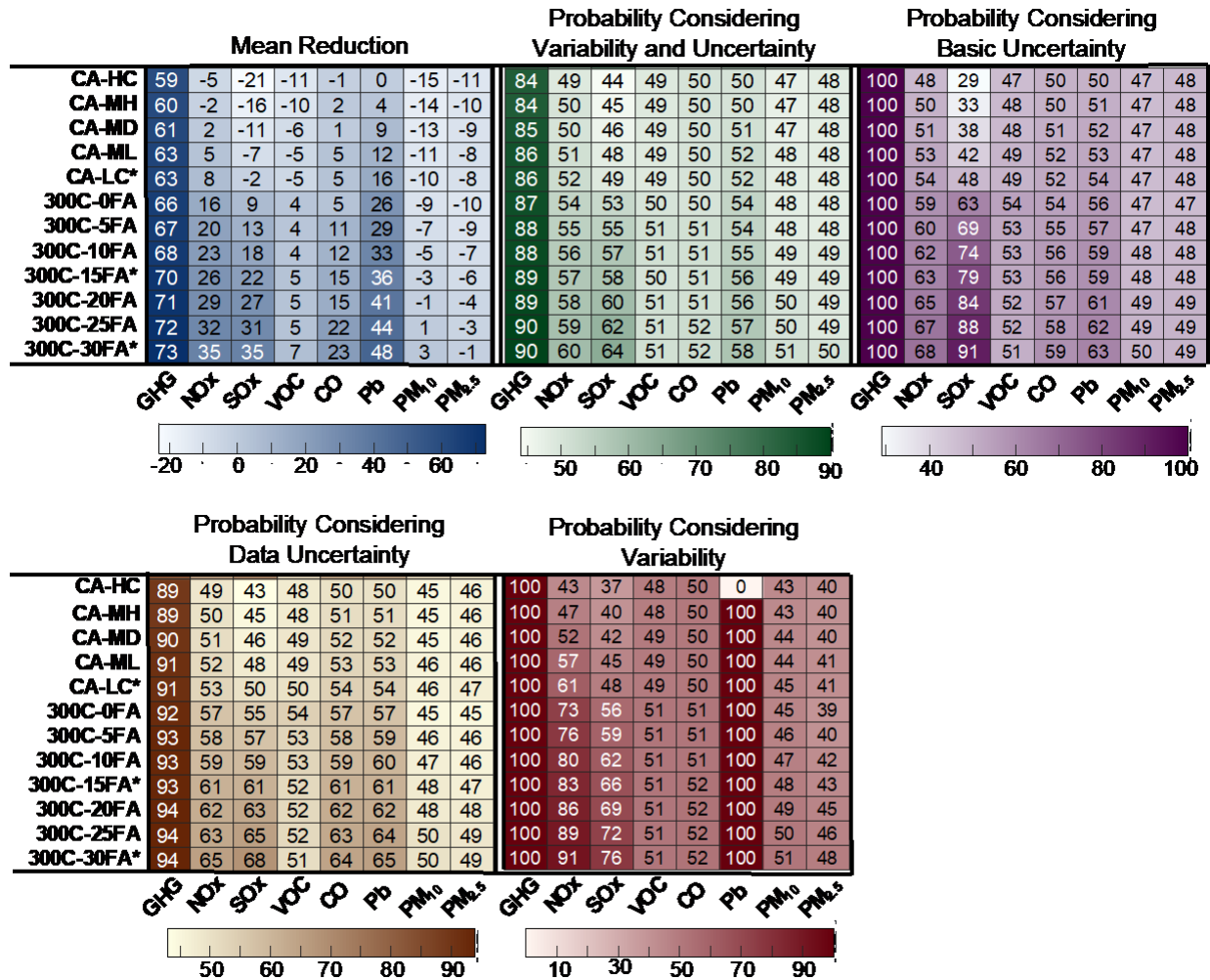


Figure 6. The mean reductions for mixtures with CCS of the kiln emissions and fuels compared to the CA-HC case with no mitigation strategy and the probabilities of the mean reduction considering variability and uncertainty, only basic uncertainty, only data uncertainty, and only variability.

For PM₁₀ and PM_{2.5} emissions, the use of CCS process led to increased emissions for all mixtures when compared to the condition without mitigation strategies. With the exception of 300-25FA and 300C-30FA mixtures, the modeled PM₁₀ and PM_{2.5} impacts were greater than the CA-HC case without mitigation strategies. The mixtures with higher cement content generally had higher SO_x, VOCs, CO, PM₁₀, and PM_{2.5} emissions. These increases were a result of the energy demands for CCS, which were modeled as being met by the kiln fuel mix. As such, tradeoffs or concurrently improving multiple aspects of production should be considered when selecting appropriate emissions mitigation strategies for concrete production.

As with the change in mean reduction for GHG emissions due to CCS, the probability of a reduction also increased for the mean reduction of the GHG impacts. The probabilities for PM₁₀ and PM_{2.5} emissions decreased by approximately 1-3% overall. For VOC and CO emissions, the

probability slightly decreased, by approximately 2-4%. The change in probability for VOC and CO was more pronounced for the CA- mixture, likely due to the higher cement contents relative to the 300C- mixtures.

4. Conclusion

The emissions from cement and concrete production are a challenge with many potential mitigation strategies; however, the probability of reduction is often not presented. In this work, 12 concrete mixtures were compared, as were the effects of three GHG emissions mitigation strategies on GHG, NO_x, SO_x, VOC, CO, Pb, PM₁₀, and PM_{2.5} emissions. Key findings of this work are:

- Even with the mitigation strategies, GHG emissions remain coupled to the Portland cement content of the mixture, with GHG emissions decreasing as the cement content decrease. However, this trend is less pronounced with use of CCS at cement kilns.
- The probability of GHG emissions reduction was greater than 50% in all cases examined, and for CCS, emissions mitigation exceeded 80% due to both the magnitude of GHG emissions reductions and the tight distribution of GHG emissions.
- Data uncertainty and variability varied for different emissions, with some, such as CO, Pb, PM₁₀, and PM_{2.5}, having relatively high uncertainties. Improved data quality, especially in the case of Pb would lead to tighter emissions distributions, which in turn, could have improved understanding of drivers to reduce emissions of these air pollutants with high probability [57].
- While use of CCS was shown to reduce GHG emissions with a high probability, increases in other emissions (caused by greater energy demand from the CCS process) should be considered as when implementing this technology.

As noted above, improved data quality for the production processes and air pollutant emissions would improve the selection of mitigation strategies and drive higher probabilities of emissions reduction. Further, the evaluation of other environmental impacts in addition to GHG emissions, such as air pollutant emissions, allow policy makers to evaluate and weigh unintended consequences associated with mitigation strategies. Such considerations are particularly important when considering the magnitude of concrete consumed. Assessments that include consideration of unintended consequences and uncertainty in environmental impacts modeled can support decision-making and should be considered when evaluating new technologies and environmental impact mitigation strategies.

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Data Summary

Products of Research

Sources of greenhouse gas (GHG) emissions, air pollutant emissions, and water demands during the concrete supply were compiled into two comparison methods: (1) a deterministic tool to output impacts from concrete production; and (2) a set of distributions that can be used for a statistical representation of impacts from concrete production. The deterministic tool has been coded in Microsoft Excel to ease its use.

Data Format and Content

Data are presented in Excel documents:

Supplementary Data 1: GHG emissions, air pollutant emissions, and water demands calculation method

Data Access and Sharing

The deterministic emissions will be published as an open access concrete impact calculator.

To access emissions distributions:

Miller, Sabbie. (2021). The role of data variability and uncertainty in the probability of mitigating environmental impacts from cement and concrete. *Environmental Research Letters*. <https://doi.org/10.1088/1748-9326/abe677>

Reuse and Redistribution

Data is currently available upon request from the PI, Sabbie Miller. Users are free to re-use the data with due citation of the work. If the data are modified or re-distributed, this must be stated explicitly and must be done in a manner that does not compromise the integrity of the data.

Appendix A

Table 3. Constituents, processes, and data sources/assumptions for deterministic environmental impact model

Constituent	Process	Flow data source & notes
Portland cement	quarry operations, raw meal preparation, finish grinding - wet kiln	[34], [43]
	quarry operations, raw meal preparation, finish grinding - long dry kiln	[34], [43]
	quarry operations, raw meal preparation, finish grinding - preheater kiln	[34], [43]
	quarry operations, raw meal preparation, finish grinding - precalciner/preheater kiln	[34], [43]
	kilning - wet kiln	[58]; air emissions calculated as cement manufacturing emissions sans energy derived emissions from [39], [59], [79]; water from [18]
	kilning - long dry kiln	[58]; air emissions calculated as cement manufacturing emissions sans energy derived emissions from [39], [59], [79]; water from [18]
	kilning - preheater kiln	[58]; air emissions calculated as cement manufacturing emissions sans energy derived emissions from [39], [59], [79]; water from [18]
	kilning - precalciner/preheater kiln	[58]; air emissions calculated as cement manufacturing emissions sans energy derived emissions from [39], [59], [79]; water from [18]
	calcination emissions	stoichiometry, assuming 65% lime content in clinker & 5% gypsum in cement
Gypsum	quarry operations, preparation	assumed same as limestone filler; air emissions from [43]; water from [18]

Constituent	Process	Flow data source & notes
Limestone Filler	quarry operations, preparation	[80]; conversions from [72]; air emissions from [43]; water from [18]
Interground limestone	quarry operations, preparation, grinding electricity	Modeled as limestone filler model + grinding electricity; grinding electricity approximated at the lower end of clinker electricity demand (30% of the 110 kwh/t reported by [81], which is on the lower end of energy reported by [82]) - lower end selected because limestone is softer than clinker, even though studies have shown that intergrinding, especially in a laboratory setting could lead to higher processing times to achieve the desired gradation
Natural pozzolans	quarry operations, preparation	assumed same as limestone filler; air emissions from [43]; water from [18]
Fly ash	N/A	[75]; water from [18]
Granulated blast furnace slag	quenching and granulation, dewatering and drying, iron removal, crushing, and grinding	[83]
Calcined clay	grinding, packing, operation, other processes kilning	[21]; air emissions based on raw materials used in cement; water from [18]
Fine Aggregates	quarry operations, preparation	[75]; water from [18]
Coarse Aggregates	quarry operations, preparation	[75]; water from [18]
Plasticizers and Superplasticizers	Raw material supply, transport prior to production gate, and manufacturing	[84]
Air Entrainers	Raw material supply, transport prior to production gate, and manufacturing	[85]

Constituent	Process	Flow data source & notes
Hardening Accelerators	Raw material supply, transport prior to production gate, and manufacturing	[86]
Set Accelerators	Raw material supply, transport prior to production gate, and manufacturing	[87]
Water Resisting Admixtures	Raw material supply, transport prior to production gate, and manufacturing	[88]
Retarders	Raw material supply, transport prior to production gate, and manufacturing	[89]
Additional Processes	Process	
Batching	Batching (per cubic meter), For water (per kg batching water)	[61]; [40]
	Aggregate transfer (per kg aggregate)	[40]; uncontrolled emissions
	Sand transfer (per kg sand)	[40]; uncontrolled emissions
	Cement unloading (per kg cement)	[40]; accounts for emissions controls through use of fraction from AP 42
	SCM unloading (per kg SCM)	[40]; controlled emissions
	Hopper loading (per kg material)	[40]; uncontrolled emissions
	Mixer loading (per kg material)	[40]; accounts for emissions controls through use of fraction from AP 42
Transportation	Transportation, truck	[62]; air emissions based on median from distributions fit to data from [63] & [64] (single point use if only one datum, uniform distribution if two data, triangular distribution if three data, lognormal distribution for four or more data); water from [18]

Constituent	Process	Flow data source & notes
	Transportation, rail	[90]; air emissions based on median from distributions fit to data from [63] & [64] (single point use if only one datum, uniform distribution if two data, triangular distribution if three data, lognormal distribution for four or more data); water from [18]
	Transportation, ship	[90]; air emissions based on median from distributions fit to data from [63] & [64] (single point use if only one datum, uniform distribution if two data, triangular distribution if three data, lognormal distribution for four or more data); water from [18]

Appendix B

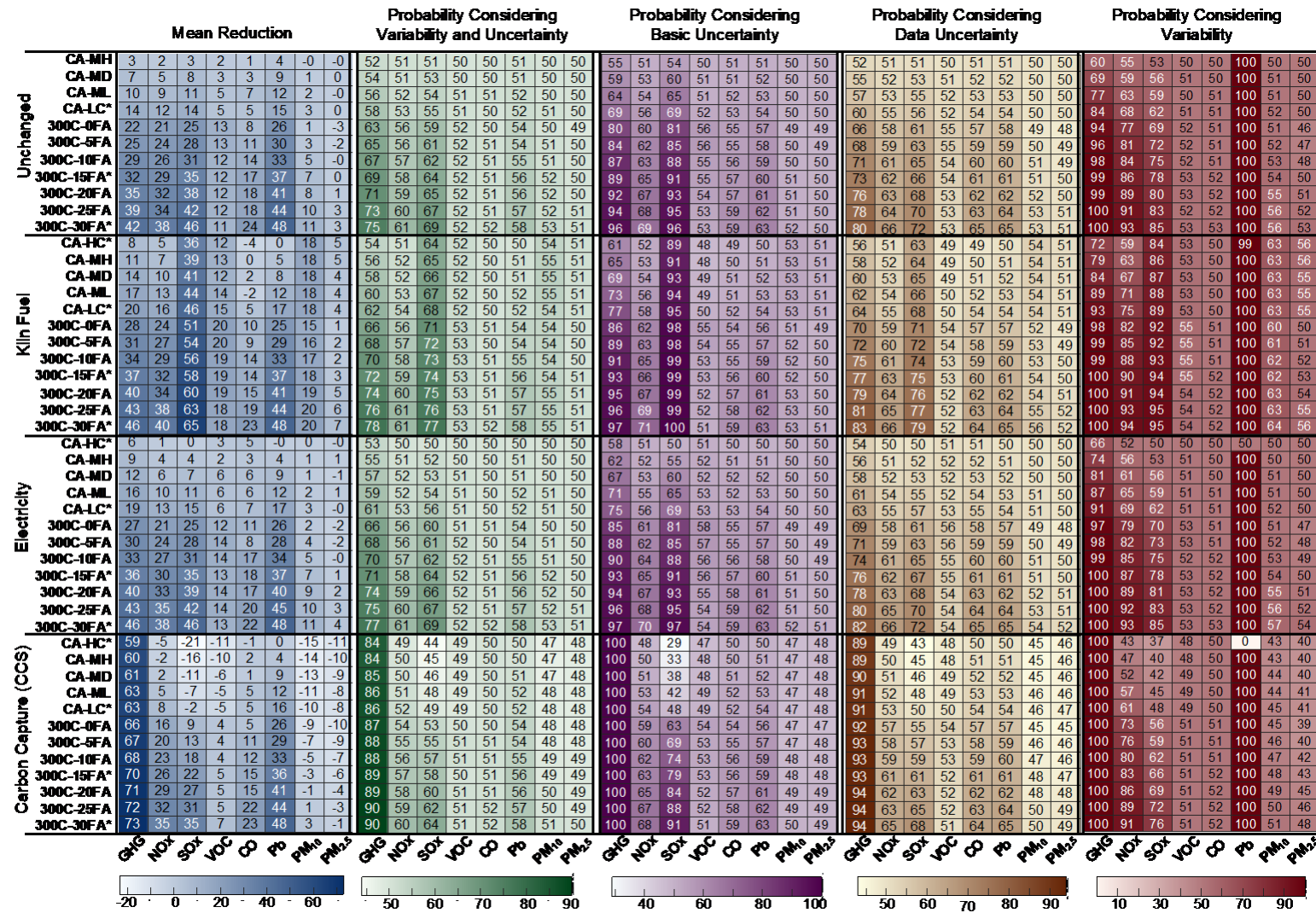


Figure 7. Mean reductions for the mixtures and mitigation strategies considered herein compared to CA-HC, and the probability of reduction considering all variability and uncertainty, only basic uncertainty, only data uncertainty, and only variability.