

Green concrete containing diatomaceous earth and limestone: Workability, mechanical properties, and life-cycle assessment[☆]

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ABSTRACT

To arrest the growth of global environmental issues, curbing the greenhouse gas emissions and primary energy consumption in concrete production is among the efficacious strategies. Cement production being the most energy- and emission-intensive activity in concrete manufacture, the partial replacement of cement clinker with supplementary cementitious materials (SCMs), which require less intensive processing, is the effective approach. Due to the unbalanced source distribution of commonly-used SCMs, e.g. fly ash (FA) and ground granulated blast furnace slag (GGBFS), this study analyzed the substitution of Portland cement (PC) in mortar and concrete mixes with up to 40% of the highly-reactive pozzolanic diatomaceous earth (DE) with abundant deposit in western United States and many other countries, aiming to provide an alternative SCM with local availability in many regions of limited FA and GGBFS supply. The influence of DE on the density, compressive strength, workability, soundness, and environmental impacts of the production of a mix was studied at different substitution levels in binary, ternary, and quaternary systems. DE-containing mixes showed improved strength development and early properties suitable for general applications. The approach of life-cycle assessment quantified the influence of DE on environmental impacts from material production, processing, and transportation in mortar or concrete manufacture. Substitution of PC with DE at 30 wt% resulted in over ~30% lower global warming potential and energy use as well as appreciably reduction in air pollutant emissions. The use of DE as SCMs in concrete is promising, worth the consideration regarding availability and cost.

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1. Introduction

The global production of Portland cement (PC) was ~4.1 Gt in 2017 (Myers et al., 2017); considering typical material constituents in concrete (Miller et al., 2016), this amount of PC is incorporated into over 26 Gt of concrete, which also consumes ~19 Gt of aggregates and 2–3 Gt of fresh water (Miller et al., 2018). The enormous consumption of raw materials and associated massive production result in significant environmental impacts on local-to-global scales.

Among all processes in concrete production, PC manufacturing has the dominant environmental impact (e.g., CO₂ emissions) and energy consumption (Monteiro et al., 2017). The emission averages of cement manufacturing amount to ~0.8 t CO₂/t cement due to its

high kiln temperature (i.e., ~1450 °C) and decomposition of limestone in the raw materials (Gartner, 2004). Partial replacement of PC clinker by SCMs is a strategy to reduce the environmental impact of the construction industry (Juenger and Siddique, 2015). In United States, PC comprises clinker and gypsum while the supplementary cementitious materials (SCMs) are added during concrete mixing; whereas in Europe SCMs are ground with clinker during cement manufacturing. SCMs are subdivided into human-made and naturally-occurring pozzolans ((alumino)silicates that react with Ca(OH)₂ generated by cement hydration to form additional cementitious materials). The former group includes industrial byproducts (e.g., ground granulated blast-furnace slag (GGBFS), fly ash (FA), and silica fume (SF)) and artificial-origin SCMs (e.g., calcined-clay and rice husk ash (RHA)) while the latter includes volcanic ashes and diatomaceous earth (DE) (Snellings et al., 2012). These SCMs have cementitious and/or pozzolanic properties. Among all SCMs, GGBFS and FA are most widely used. Cementitious materials containing PC-FA or PC-GGBFS blends with typical substitution levels of 10–30% exhibit comparable mechanical

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properties to concretes with a high content of PC at late ages (after 28 days) (Lothenbach et al., 2011).

Industrial byproduct SCMs are often modeled as generating low impacts from processing compared to PC clinker manufacturing (Gursel et al., 2016). Thus, PC-SCMs blends with high substitution level are favorable to reduce life-cycle environmental impacts (Celik et al., 2015). However, there is a lack of examination whether the studied SCM (e.g., RHA and FA) is among the most appropriate and economical choice of SCMs for the specific geographical region. Furthermore, without seeking more appropriate and efficient uses, the net benefit of utilizing these SCMs may not be maximized due to the environmental impact of long-distance transportation as a result of their nonuniform distribution and low local availability in many areas. Although “green” concretes with high volume (high substitution level) of FA, LS, or volcanic ash have exhibited relatively low Global Warming Potential (GWP) per compressive strength (Celik et al., 2015) (Gursel et al., 2016), strength gain of concretes with high-volume of SCMs can be impractically slow (strength at least 30% lower than plain cement concrete at 28 days) (Celik et al., 2014a) (Celik et al., 2014b). The GWP intensities, which is obtained by normalizing the total GWP emissions to the strength, at 28 and 90 days of these “green” concretes are only 10–20% lower than plain PC concrete (i.e., with the absence of SCMs). The anticipated net benefits are lowered in column designs since larger cross-section has to be selected to offset the negative influence of these cement blends on strength development by compromising self-weight of a structure (Miller et al., 2015).

According to United States Geological Survey (USGS), California and/or Texas are the largest producers and consumers of PC and concrete in the US in the latest years; whereas, California produces limited or negligible amounts of FA, GGBFS, SF, calcined-clay, and RHA (Gocha, 2015). In scenarios of concretes produced in California, these SCMs have to be modeled as imports from distances of over 1000 km (Fan and Miller, 2018). The SCM with abundant availability in California is diatomaceous earth (also termed diatomite), a biogenic pozzolan. Vast reserves of DE are also reported in other western states (e.g., Oregon, Washington, and Nevada), and many other countries (e.g., China, Japan, Turkey, Czechia, and Thailand). DE is highly porous with voids of up to 80–90% (Lemons, 1997), and the silica content of DE is typically 86–94 wt% (Tsai et al., 2004). Thus, DE has broad applications as filters, adsorbents, fillers, and insulators (Xu and Li, 2014). The silica content of low-grade DE can be as low as 50 wt% due to intermixed calcite (Fragoulis et al., 2005), clay minerals (Degirmenci and Yilmaz, 2009), heavy metals, and organic matters. Therefore, such low-grade DE has limited use as filters or adsorbents but can be used in the production of cement and/or as concrete materials (i.e., lightweight aggregates) (Xu and Li, 2013) (Crangle, 2018). The average cost of DE used as lightweight aggregate and binder in the US is \$10/tonne, ~9% of the average price of PC. Whereas, the price of SF is up to \$1000/tonne (Crangle, 2018). DE used in agroindustry, brewery, and distillery industries is disposed of by landfill; however recycled DE can also be blended in concrete (Letelier et al., 2016). Unlike many SCMs (e.g., FA and SF), which are typically grey or black, DE can be blended with white Portland cement (wPC) since DE is naturally dull-white. Thus, partial substitution of PC with DE, as a natural pozzolan, in concrete production can be both environmental-friendly and economical.

The high porosity of DE (Lemons, 1997) results in high water demands. Thus, the substitution level of untreated DE is often limited to ~10% without compromising strength or using superplasticizers (Starnatakis et al., 2003) (Yilmaz and Ediz, 2008) (Degirmenci and Yilmaz, 2009). Mechanical properties of cementitious materials mixed with more than 20% DE have not been reported (Yilmaz, 2008). GWP of manufacturing superplasticizers is

relatively intense: the GWP of manufacturing 1 kg of superplasticizers is merely 11% lower than PC clinker manufacturing with 1 kg (Gursel et al., 2016); nevertheless, DE level in PC-DE blends can be significantly boosted with a low dose of superplasticizer (up to 1.5 wt% of cementitious materials). DE is a highly-reactive pozzolanic material (Degirmenci and Yilmaz, 2009), and the use of DE for PC substitution may resemble that of SF. Thus, “green” concrete with high-volume of DE can exhibit high-strength at both early- and late-ages, which may promote the use of DE-containing “green” concretes in the construction industry.

Considering 1) the addition of limestone powder (LS) benefits mechanical properties and durability of PC concrete (Lothenbach et al., 2008; Zajac et al., 2014), 2) DE often intermixes with calcite (CaCO_3), and 3) limestone, one of the most common rocks, is very abundant in the western US, the use of PC-DE-LS ternary blends can be more practical and environmental-friendly in these regions. This study aimed to determine mechanical properties, early-age properties (setting times, workability, water demand, and soundness), and environmental impacts of “green” white PC mortar and concrete mixed with DE, limestone powder, and superplasticizer. GWP (expressed in CO_2 -equivalent), energy use, and major air pollutants were assessed for different mortar and concrete mixes. The GreenConcrete Life-Cycle Assessment (LCA) tool was used (Gursel et al., 2016). For each process involved in the manufacture of related raw materials and the concrete, impacts directly due to the production phase as well as incurred in other phases of the supply chain were evaluated comprehensively, by means of discussing geographical influences on transportation and energy use and technological choice for each phase.

2. Experimental materials and methods

2.1. Materials

The study used ASTM Type I white Portland cement (wPC) with 57.3% C_3S , 30.1% C_2S , 4.6% C_3A , 1.6% ye'elimite, and 3.1% calcium sulfates. Note that ye'elimite occurs due to the high sulfate content in the raw materials during clinker manufacturing (Taylor, 1997). Freshwater diatomaceous earth (DE, California) containing ~83% amorphous silica, ~9% montmorillonite, and ~6% moisture was used as received. Fine commercial limestone powder (LS, 99% purity) from California and gypsum powder (99%, California) were used. The bulk oxide composition, specific surface area, and density of wPC, DE, and LS are given in Table 1. The specific gravity of #30–100 grade sand used in mortars was 2.65. A polycarboxylate ether (PCE)-based superplasticizer (BASF) with solid content of 40 wt% was used to maintain the required workability (slump or flow of ~100 mm) of pastes and mortars. For concrete mixtures, Pea gravel with a maximum size of 9.5 mm was used as coarse aggregate, and Vulcan sand with fineness modulus of 2.9 was used as fine aggregate.

2.2. Mixture proportions

Pastes were prepared for tests of setting time, mini-slump, consistency, soundness, and density. Mortars were prepared for compression test and life cycle assessment. Concrete specimens were cast for compression test and life cycle assessment.

The mixture proportions of cementitious materials (Table 2) are labelled 30DE-5LS-3C\$, for instance, for 30 wt% DE, 5 wt% LS, and 3 wt% gypsum replacement. Pastes were prepared with a water-to-cementitious material ratio (w/cm) of 0.48. Here, cementitious materials refer to PC, DE, LS, and gypsum. Mortars were prepared with w/cm = 0.48 and a sand-to-cementitious material ratio of 2.75. Mortars are labelled M-30DE-5LS-3C\$, for instance, for 30 wt%

Table 1
Chemical composition (wt.%) and physical characteristics of the white Portland cement, diatomaceous earth, and limestone powder.

	wPC	DE	LS
SiO ₂	20.55	85.63	1.09
Al ₂ O ₃	4.42	3.96	0.22
Fe ₂ O ₃	0.26	1.04	0.15
CaO	66.28	0.57	55.09
MgO	0.86	0.46	0.50
SO ₃	4.10	<0.01	<0.01
Na ₂ O	0.04	<0.01	0.02
K ₂ O	0.20	0.14	0.01
TiO ₂	0.07	0.26	0.02
P ₂ O ₅	0.04	0.09	0.04
MnO	0.01	0.02	0.01
SrO	0.04	0.01	0.01
Cr ₂ O ₃	<0.01	<0.01	<0.01
ZnO	<0.01	<0.01	<0.01
LOI	1.64	7.74	42.83
Free CaO	0.2	–	–
Specific surface area [m ² /kg]	1183	27848	2805
Density [kg/m ³]	3140	2000	2740
D10 [μm]	1.45	1.24	0.92
D50 [μm]	10.94	6.77	2.98
D90 [μm]	36.85	18.41	8.16

DE, 5 wt% LS, and 3 wt% gypsum replacement, and wPC reference mortars are labelled as M-wPC. The workability of pastes was adjusted using PCE superplasticizer when necessary. PCE was added into mortar mixes until the flow reached 105–117 mm. Since SCMs with high surface area significantly modify the reactivity of the aluminates phases (Antoni et al., 2012) (Rossen et al., 2015), gypsum content was adjusted to modify the early behavior of the blends.

The concrete mixture proportions (w/cm = 0.48) are listed in Table 3. The total aggregate-to-cementitious materials ratio was 3.9:1. The ratio of PC replacement (CR) to total cementitious materials was 5–35 wt% by weight. The DE content varied between 10 wt% and 30 wt% while LS content was held constant at 5 wt% in ternary blends. PCE was added until the diameter of slump flow reached 550–670 mm. Concrete specimens are labelled C-30DE-5LS, for instance, for 30 wt% DE and 5 wt% LS replacement, and wPC concrete references are labelled as C-wPC.

Table 2
Formulation for the PC-DE-LS blends.

	wPC [wt.%]	DE [wt.%]	LS [wt.%]	Gypsum [wt.%]	wPC replacement [wt.%]
wPC	100	–	–	–	–
5LS	95	–	5	–	5
10LS	90	–	10	–	10
5DE	95	5	–	–	5
10DE	90	10	–	–	10
15DE	85	15	–	–	15
20DE	80	20	–	–	20
25DE	75	25	–	–	25
30DE	70	30	–	–	30
10DE-10LS	80	10	10	–	20
15DE-5LS	80	15	5	–	20
20DE-5LS	75	20	5	–	25
20DE-10LS	70	20	10	–	30
30DE-5LS	65	30	5	–	35
40DE-5LS	55	40	5	–	45
20DE-5LS-1.5C\$	73.5	20	5	1.5	26.5
20DE-5LS-3.0C\$	72	20	5	3	28
30DE-5LS-1.5C\$	63.5	30	5	1.5	36.5
30DE-5LS-3.0C\$	62	30	5	3	38

Table 3
Concrete mix proportions.

	w/cm = 0.48						kg/m ³ of concrete			
	Proportions (by wt.%)						PCE (%)	CM	C	CR
	wPC	DE	LS	Aggregates						
			Fine	Coarse						
C-wPC	100	–	–	208	181	0.58	433	433	0	
C-5LS	95	–	5	208	181	0.58	432	410	22	
C-10DE	90	10	–	208	181	0.91	429	386	43	
C-20DE	80	20	–	208	181	1.12	424	339	68	
C-30DE	70	30	–	208	181	1.42	420	294	126	
C-20DE-5LS	75	20	5	208	181	1.11	423	317	106	
C-30DE-5LS	65	30	5	208	181	1.41	419	272	147	

2.3. Methods

2.3.1. Setting time, consistency and soundness test

All pastes were mixed using a Hobart N50 mixer at 136 RPM for 2min and 281 RPM for 2min. The water of normal consistency of pastes (without PCE) was determined according to ASTM C187 (ASTM-C187, 2016). Water of normal consistency is defined as the quantity of water that permits a Vicat plunger to penetrate a paste by 9–11 mm during 30s of settling after completion of mixing. Setting time (initial and final) of pastes (w/cm = 0.48 and with normal consistency) was determined by Vicat needle penetration tests according to ASTM C191 (ASTM-C191, 2018). Initial setting is defined as a penetration of Vicat needle by 25 mm during 30s of settling after completion of mixing. Final setting is defined as the first time at which the needle does not mark the paste with a complete circular impression. The setting times were measured to the nearest 1 min. Soundness of the pastes with normal consistency and with w/cm = 0.48 (with PCE) was determined using Le-Chatelier method in EN 196-3 (EN-196-3, 2016). The pastes were filled in steel molds with glass plates placed on the top and bottom. The whole assembly was immersed in water at 24 °C for 24 h, then the distance between the two indicators was measured. Subsequently, the assembly was immersed in water and heated to 100 °C for 1 h. The distance between the indicators was measured again. The difference between the two distances was recorded as soundness expansion.

2.3.2. Mini-slump

The mini-slump test setup is a mini-cone with downscaled Abrams cone geometry (Tan et al., 2017): 3.5 mm top diameter, 6 mm bottom diameter, and 6 mm height. The test was performed by placing a cone on a flat sheet of moistened borosilicate glass (400 mm × 400 mm). Freshly mixed paste in section 2.3.1 was immediately poured into the moistened cone after completion of mixing, followed by compaction using a moistened steel rod. After 10 min of mixing, the cone was lifted vertically in 3 s, and the diameter of the spread was obtained at 30 s.

2.3.3. Density of the cement Paste samples

For density measurements, the freshly mixed pastes ($w/cm = 0.48$) were sealed into polyethylene containers at 24 °C and were demolded after 24 h. The density of the pastes was determined based on Archimedes Principle using a precision scale and a density kit by submerging the specimens into water.

2.3.4. Compressive strength

Mortars were mixed using Hobart N50 mixer. The freshly mixed mortars were cast into cubic molds (50 mm × 50 mm × 50 mm) and cured at 24 °C and 100% relative humidity (RH) for 1, 7, 28, and 90 days. For each group, at least three cubes were compressed at a loading rate of 0.5 MPa/s according to ASTM C109 (ASTM-C109, 2016). 75 mm × 150 mm concrete cylinders were cast into plastic molds and demolded after 24 h. The cylinders were cured at 24 °C and 100% RH. For each group, at least three cylinders were compressed at a loading rate of 0.5 MPa/s according to ASTM C39 (ASTM-C39, 2018). Before compression test, the surface of all specimens was air-dried, and the concrete cylinders were sulfur-capped.

3. Life cycle assessment (LCA)

3.1. Methodology of LCA

According to ISO 14040, the approach of life cycle assessment stands for collecting, interpreting, and evaluating the inputs, outputs, and potential environmental impacts through the life cycle of a product system. Thereupon, a standard framework includes: definition of goal and scope, life cycle inventory (LCI) analysis in relation to the scope, life cycle impact assessment (LCIA) in relation to LCI, and interpretation of results for the three sectors. Common applications of LCA approach are product development, process improvement, and public policy making, etc. (ISO-14040, 2006)

In relation to LCA application to concrete industry, scope is defined for the product system, the system boundary, and the functional unit (e.g., 1 m³ of ready-mixed concrete for comparison convenience). Separate from other production systems by the system boundary, the product system (i.e., concrete production) comprises functionally-defined unit processes (e.g., pyroprocessing in cement production) connected by material and energy. LCI involves data gathering and measuring of inputs (materials, electricity, fuel, etc.) and outputs (GWP and air pollutant emissions, etc.) within the scope. LCIA provides an evaluation of the significance of potential environmental impacts through association with results from LCI (ISO-14040, 2006).

LCA studies on different types of concrete have been conducted. It is shown that the production of recycled aggregate concrete generates higher GWP and demands higher energy use than that of natural river aggregate concrete mainly due to energy demand in crushing (Marinkovic et al., 2010). It has also been shown that among the four most commonly produced cement types defined in the European standard EN 197-1, CEM I and III, with the highest and the lowest clinker content respectively, also have the highest and

the lowest GWP, the ozone layer depletion potential, and the acidification potential (Moretti and Caro, 2017).

3.2. GreenConcrete LCA tool

The GreenConcrete LCA Tool is utilized in this study. The tool is embedded with definition of the product system and system boundary which is determined by the cradle-to-gate approach (Gursel et al., 2016), from material extraction and production or processing, to transportation to concrete plant, and to concrete mixing and batch, instead of cradle-to-grave, from raw material extraction to disposal (see Fig. S1 in Appendix). The Tool is also embedded with definition of unit processes and a database of unit process-specific factors of energy consumption, GWP, and CO, NO_x, SO₂, PM10, Volatile organic compound (VOC), and lead emission. Based on this definition of the scope, the user inputs the functional unit, by specifying the concrete volume and mix design, and assumptions for each unit process (e.g., mode and distance for transportation process, technological options for cement production process, and the fuel mix characteristic of the region or the plant). Upon the completion of LCI inputs, the tool performs the quantification of the LCI results, which is outputted for the user's own interpretation (Gursel, 2014).

3.3. LCA of production of mortar and concrete mixes with DE

3.3.1. Scope of LCA

The environmental impacts of the production of mortar and concrete containing DE and LS were evaluated using the GreenConcrete LCA Tool. The process flow and system boundary of the LCA study is based on the assumptions of the scope in the tool, demonstrated in Fig. 1. Thorough examination has shown that LCA studies in many literature are focused on the evaluation of greenhouse gas and criteria air pollutant emissions while GWP in CO₂ equivalent (CO₂-eq) is the most widely used factor (Gursel, 2014). Therefore, GWP, criteria air pollutant emissions as well as energy use are considered in this study. GWP and energy use were then normalized with respect to 28-day strengths (*f_c*) of the mortar and concrete mixes. Air pollutant information was also modeled. Note that the emissions of CO, NO_x, SO₂, and PM10 were modeled for both material production or processing and transportation; however, the modelling of VOC and lead emissions did not include contribution from transportation, due to the lack of VOC and lead data in the database of the tool.

3.3.2. Assumptions of life-cycle inventory (LCI) inputs

14 mortar mixes and 7 concrete mixes were compared to evaluate their environmental impacts. LCA manufacturing and location-based assumptions and calculations involved in the LCI of the raw materials of each mix are described below.

White Portland cement was delivered from a cement plant in Long Beach, CA to Berkeley, CA (concrete plant), the distance of which is ~637 km by Class 8B truck. For Portland cement production, California electricity fuel mix is used in the calculations. Due to the stricter requirement of color for wPC compared to ordinary Portland cement (OPC), mostly petroleum coke is used as the fuel for pyroprocessing of wPC while the U.S. average fuel mix for pyroprocessing of OPC uses ~20% petroleum coke and more than 60% coal ((Gursel, 2014), see Table S1 in Appendix) which is not suitable for fueling the production of wPC because the non-white impurities from coal can contaminate the product. Moreover, the production of wPC is more energy-intensive than that of OPC: thermal energy of 4950 MJ is required in the production for 1 tonne of wPC (ICR, 2003), ~25% more than for 1 tonne of OPC produced with preheater/precalciner kiln. Because of the limited content of

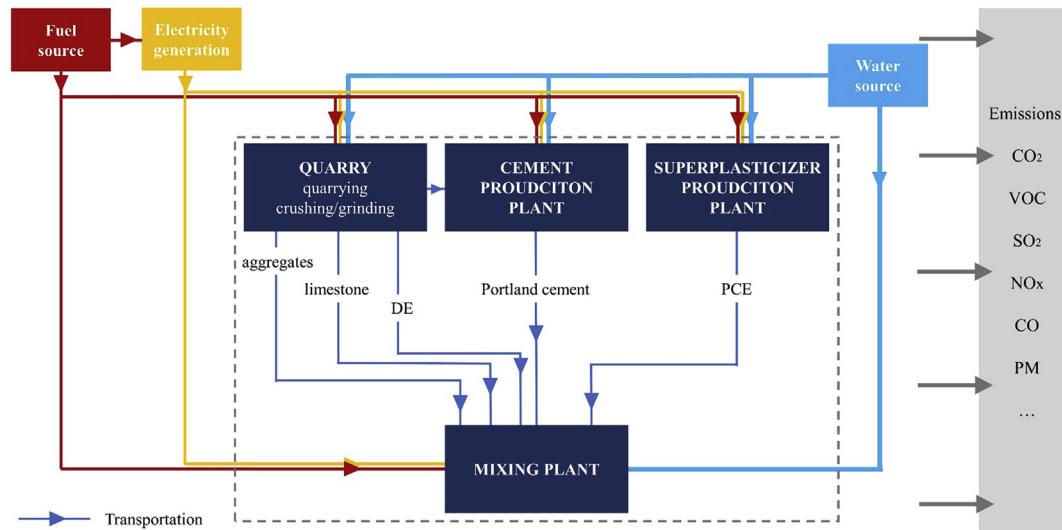


Fig. 1. Process flow of the production system. Dashed box indicates the system boundary.

Fe in wPC raw materials, the higher energy use in wPC production results from the higher kiln temperature or the addition of CaF₂ to maintain the same temperature as an OPC kiln. Since wPC can generally be produced with the typical kiln for OPC, it is assumed for this LCA study that the air pollutant emissions due to wPC are comparable to OPC, using common technology options in Table 4, despite higher energy use. Electrostatic precipitator (ESP) is assumed for particulate control in cement manufacturing due to its higher efficiency than fabric filter.

Limestone powder (LS) was transported from Lucerne Valley, CA, for ~698 km to Berkeley by Class 8B truck. The gypsum used in the adjustments was produced and delivered from Stockton, CA to Berkeley for a distance of ~116 km using Class 8B truck. DE was delivered from Shasta County, CA, by Class 8B truck for ~391 km. The energy requirement for quarrying and processing of this DE deposit is unknown; thus, it is assumed to be similar to the average energy required for natural pozzolan (NP). DE is considerably more porous than typical NP (i.e., volcanic ash) (Deniz, 2011), thus the processing of DE only requires gentle milling/grinding; besides, no calcination is required for DE from this deposit. Thus, the energy requirement for DE processing is overestimated when we use the average energy demand for natural pozzolan processing.

PCE superplasticizer with 40% solid content was used. The product is assumed to be transported by rail first from Cleveland, OH to Chicago, IL via CSX railroad system and then from Chicago, IL to Emeryville, CA via BNSF (Burlington Northern-Santa Fe) railroad system; from there, it is assumed to be delivered to Berkeley with Class 2B truck. The estimated transportation distance is 4463 km by rail and an additional 5 km by Class 2B truck.

The fine aggregate is assumed to be transported by Class 8B truck from Pleasanton, CA to Berkeley with a distance of ~53 km. The coarse aggregate is natural gravel, delivered from British Columbia, Canada for 1800 km by barge to a port at San Francisco and transported from the port to Berkeley for 40 km by Class 8B truck.

Table 4 summarizes LCA assumptions for material choices, transportation modes, technological alternatives, and geographical factors, which are essential parts of the input to the GreenConcrete LCA Tool. Table 5 shows the electricity grid mix by energy source of the different regions, which are determined as locations of the material suppliers in the LCA assumptions.

4. Results

4.1. Setting time, consistency and soundness of pastes

The w/cm ratio desired for normal consistency increased with increasing DE level (Table 6 and Fig. 2a). The high water demand of wPC-DE blends is due to the highly porous structure of DE (high specific surface area). Water demands for 20DE and 30DE were 63% and 101%, respectively, higher than that for the wPC control group. Water demands for wPC-LS blends were comparable to wPC control due to their similar surface area. 20DE-5LS and 30DE-5LS ternary blends also showed comparable water demands to 20DE and 30DE, respectively. The addition of gypsum into 30DE-5LS blends slightly reduced the water demands. Thus, superplasticizers are required at high DE level.

The substitution of wPC with LS had no significant influence on the setting of the blends. The initial and final setting of wPC-DE binary and wPC-DE-LS ternary systems were retarded at high DE level. The initial and final setting time of 30DE increased by 30% and 52%, respectively, compared to wPC control. The addition of gypsum elevated the initial and final setting times of 30DE-5LS. The delayed times of initial and final setting were due to the dilution of the hydration system (i.e., higher w/cm). The expansion of all blends in the soundness test was not greater than 1 mm. Thus, all blends conformed with the standard.

4.2. Effects of PCE on the early-age properties of pastes

To cast pastes (w/cm = 0.48) with a slump of 110 ± 15 mm, the PCE solid content was adjusted by up to 0.9% of cementitious materials weight. PCE dosage increased as the DE level in the blends increased (Fig. 2b and Table 7). The substitution of wPC with up to 10% LS showed no distinct influence on the slump without the addition of PCE. Both LS and DE accelerated the initial and final set. The result that fine LS accelerated the setting and hydration of wPC is consistent with (Lothenbach et al., 2008). For replacement with up to 15 wt% DE, the final setting time was extended by 15 min, whereas the initial setting time was shortened by 16 min. This fact was due to a competition between retardation of wPC hydration by PCE (Winnefeld et al., 2007) and acceleration of C₃A hydration by the high surface area of amorphous silica (Rossen et al., 2015).

Table 4
Assumptions used in LCA calculations.

User input data	Type of material	
Type of Portland cement	wPC Type I	
Type of SCMs	Limestone, gypsum, Diatomaceous earth	
Type of admixture	PCE	
Electricity grid mix	Location	
Concrete plant	Berkeley, California, U.S.	
Cement supplier	Long beach, California, U.S.	
Fine aggregate supplier	Pleasanton, California, U.S.	
Gypsum supplier	Stockton, California, U.S.	
Limestone supplier	Lucerne Valley, California, U.S.	
DE supplier	Shasta County, California, U.S.	
PCE supplier	Cleveland, Ohio, U.S.	
Coarse aggregate supplier	British Columbia, Canada	
Transportation details	Mode	Distance (km)
Cement raw materials to cement plant	Class 8B truck (0.76 MJ/ton-km)	1
Gypsum to cement plant	Class 8B truck	277
Cement to concrete plant	Class 8B truck	637
Gypsum to concrete plant	Class 8B truck	116
DE to concrete plant	Class 8B truck	391
Limestone to concrete plant	Class 8B truck	698
PCE to concrete plant	Rail (0.24 MJ/ton-km) and Class 2B truck (3.86 MJ/ton-km)	4463 km by rail and 5 km by truck
Fine aggregate to concrete plant	Class 8B truck	53
Coarse aggregate to concrete plant	Barge (0.25 MJ/ton-km) and Class 8B truck	1800 km by barge and 40 km by truck
Technology options	Type of technology selected	Distance (m)
Cement raw materials prehomogenization	Dry, raw storing, preblending	
Cement raw materials grinding	Dry, raw grinding, vertical roller mill	
Cement raw materials blending/homogenization	Dry raw meal homogenization, blending, and storage	
Clinker pyroprocessing	U.S. average kiln ^a	
Clinker cooling	Rotary (tube) cooler	
Cement finish milling/grinding/blending	Roller press	
Cement PM control technology	Electrostatic precipitators	
Conveying within the cement plant	Screw pump	20 m between process stations
Concrete batching plant loading/mixing	Mixer loading (central mix)	
Concrete batching plant PM control	Fabric filter	

^a Note, energy use of pyroprocessing for wPC is ~720 MJ/tonne higher than grey cement using U.S. average kiln. GWP emissions of pyroprocessing for wPC is accordingly higher.

Table 5
Electricity grid mix percentage by the source of energy.

User-Input Data	California (%)	Ohio (%)	British Columbia (%)
Coal	0.15	66.82	–
Natural gas	43.4	17.72	6
Fuel oil	0.02	0	–
Petcoke	0.2	1.01	–
Nuclear	8.69	13.35	–
Hydropower	21	0.31	86
Biomass	2.82	0.78	6
Geothermal	5.69	–	–
Solar	11.79	–	–
Wind	6.24	–	2

^a Percentages may not add up to 100% due to rounding of the numbers during calculations.

When the DE level was above 20%, it accelerated the initial and final set. This acceleration mechanism may resemble the role of silica fume in PC-SF hydration system (Rossen et al., 2015), in which SF significantly accelerates the early hydration of both C₃A and C₃S. With an additional 3% gypsum, the initial and final set of 30DE-5LS-3.0C\$ were delayed by ~40 min, compared to those of 30DE-5LS. All these mixes at w/cm = 0.48 exhibit practical times of initial and final set.

The addition of PCE systematically delayed the early and final setting of 20DE and 30DE blends (w/cm = 0.48, Table 8 and Fig. 2c). A nonlinear correlation between PCE addition and slump of 20DE and 30DE blends was observed. The slump of both blends increased by over 200 mm by adding extra 0.1% PCE.

Neither PCE nor gypsum influenced the soundness of the blends at w/cm = 0.48. The soundness of all blends conformed with the standard.

4.3. Density of Hardened Paste

The density of the wPC-DE pastes (w/cm = 0.48) systematically decreased as DE level increased (Fig. 3) because DE is lighter than wPC. The density of 30DE paste was 10% lower than that of wPC control. The density of wPC-LS increased as LS content increased, although LS is slightly lighter than wPC. The result is caused by the observed small amount of free water on the surface of the LS-pastes after demolding, so the effective water-to-cement ratio decreased (i.e., dilution effect (De Weerd et al., 2011)) with the LS substitution. Comparing to the 20% DE substituted sample, a higher density was not observed for 20%DE-5%LS substituted sample since the DE modified the consistency of the pastes. The addition of 1.5–3 wt% gypsum showed negligible influence on the density of DE-LS substituted pastes.

4.4. Mortar compressive strength

At LS levels of 5% and 10%, the LS-containing mortar showed higher 1-day compressive strengths but lower values at later ages (Fig. 4 and Table 9). The former was due to the decreased porosity of mortars containing LS while the latter was caused by less binding material due to wPC dilution. The results for the 1-day compressive strength of M-10DE, M-20DE, and M-30DE were comparable to the M-wPC control while the strength gain of these PC-DE blended mortars was considerably larger than M-wPC control at 7–90 days. For example, the compressive strengths of M-20DE at 28 and 90 days were 138% and 145%, respectively, relative to M-wPC. Higher strengths at 28 and 90 days were achieved with mortars with a wPC substitution level of up to 45% (40% DE and 5% LS). The strength of M-40DE-5LS at 28 and 90 days were 120% and 132%, respectively, relative to M-wPC. The 30% DE-5% LS mortar mixture exhibited the highest strengths at 28 and 90 days as 71 and 82 MPa, respectively. The significant increase in strength of DE-containing mortars was due to the pozzolanic reaction of high surface DE.

As mentioned before, DE with high surface area modifies the early hydration of PC. Thus, M-20DE-5LS and M-30DE-5LS blends were further substituted with 1.5% and 3% gypsum to adjust the reactivity of the aluminates phases (Myers et al., 2017). With additional 1.5% sulfate, the strength of M-20DE-5LS-1.5C\$ at 1–90 days increased by 3%–10% relative to M-20DE-5LS (i.e., without gypsum adjustment). Compared to M-20DE-5LS, the strengths of M-20DE-5LS-3.0C\$ at 1–90 days decreased by up to 16% due to the over-dose of gypsum. M-30DE-5LS-3.0C\$ and M-30DE-5LS exhibited comparable strengths.

Table 6
Water-to-cementitious materials ratio for normal consistency, setting time, and soundness of the blends.

	w/cm for normal consistency	Initial setting (min)	Final setting (min)	Final setting – Initial setting (min)	Soundness expansion (mm)
wPC	0.281	56	73	17	1
5LS	0.283	55	71	16	<1
10LS	0.286	54	70	16	1
5DE	0.321	50	73	23	<1
10DE	0.376	55	75	20	<1
15DE	0.413	58	85	27	1
20DE	0.46	57	89	32	1
25DE	0.513	71	103	32	<1
30DE	0.565	73	111	38	1
20DE-5LS	0.462	57	90	27	<1
30DE-5LS	0.566	72	110	36	1
30DE-5LS-1.5C\$	0.563	79	118	39	<1
30DE-5LS-3.0C\$	0.558	84	121	37	1

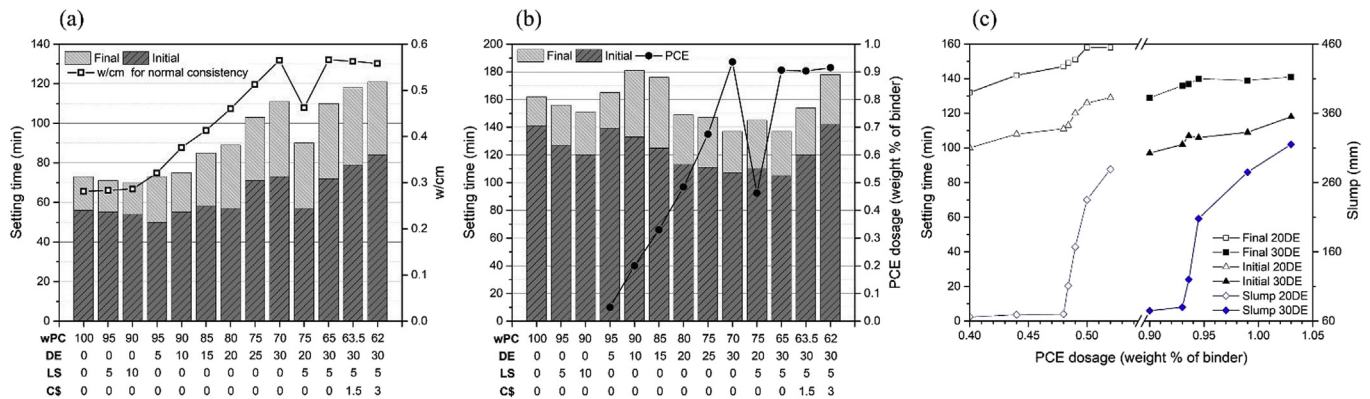


Fig. 2. (a) Final and initial setting time of cement pastes and w/cm ratio for normal consistency; (b) final and initial setting time of cement pastes with w/cm of 0.48 and PCE dosages; (c) setting time and slump versus PCE dosage for 20DE and 30DE cement pastes (w/cm = 0.48).

Table 7
PCE dosage, setting time, slump and soundness of mixtures with w/cm = 0.48.

	PCE dose of DE (%)	PCE dose of binder (%)	Initial setting (min)	Final setting (min)	Final setting – Initial setting (min)	Slump (mm)	Soundness expansion (mm)
wPC	0	0	141	162	21	101	1
5LS	0	0	127	156	29	101	<1
10LS	0	0	120	151	31	99	<1
5DE	1	0.05	139	165	26	108	<1
10DE	2	0.2	133	181	48	104	<1
15DE	2.2	0.33	125	176	41	112	1
20DE	2.42	0.484	113	149	36	111	1
25DE	2.7	0.675	111	147	36	110	1
30DE	3.12	0.936	107	137	30	120	<1
20DE-5LS	2.31	0.462	110	145	33	120	<1
30DE-5LS	3.02	0.906	105	137	32	119	<1
30DE-5LS-1.5C\$	3.01	0.903	120	154	34	121	1
30DE-5LS-3.0C\$	3.05	0.915	142	178	36	119	1

4.5. Concrete compressive strength

Compressive strengths of all DE containing concretes were ~3–7 MPa higher than C-wPC control at 7 days (Table 10). The wPC-DE-LS ternary-blended concrete exhibited slight lower strengths compared to mixtures with the same DE levels due to their lower contents of binding phase at an early age. 28-day compressive strengths (f_{c28}) of all DE-containing concrete mixtures were 5–15 MPa higher than C-wPC. Due to the pozzolanic reaction, strength gain from 7 to 28 days significantly increased as DE level in concrete mixtures increased. The concrete with 35% replacement

(30% DE and 5% LS) reached ~50 MPa at 28 days, 41% higher than C-wPC. Concrete containing 5%LS exhibited slight higher strengths relative to C-wPC at 7 and 28 days, which is consistent with (Felekoglu et al., 2006).

4.6. Life cycle assessment

4.6.1. Results of the scenario

The CO₂-equivalent (CO₂-eq) is used to express the GWP of multiple compounds of greenhouse gas, which include major compounds (CO₂, CH₄, and N₂O) and minor compounds (e.g., SF₆,

Table 8
Setting time, slump and soundness of mixtures (w/cm = 0.48) with different PCE doses.

	PCE dosage of DE (%)	PCE dosage of binder (%)	Initial setting (min)	Final setting (min)	Final setting – Initial setting (min)	Slump (mm)	Soundness (mm)
20DE	0	0	72	109	37	62	1
	2	0.4	100	132	32	66	<1
	2.2	0.44	108	142	34	69	<1
	2.4	0.48	111	147	36	70	<1
	2.42	0.484	113	149	36	111	1
	2.45	0.49	120	151	31	167	<1
	2.5	0.5	126	158	32	235	1
	2.6	0.52	129	158	29	279	1
30DE	0	0	–	–	–	61	–
	3	0.9	97	129	32	75	1
	3.1	0.93	102	136	31	80	<1
	3.12	0.936	107	137	30	120	<1
	3.15	0.945	106	140	34	208	<1
	3.3	0.99	109	139	30	275	<1
	3.43	1.03	118	141	23	315	<1

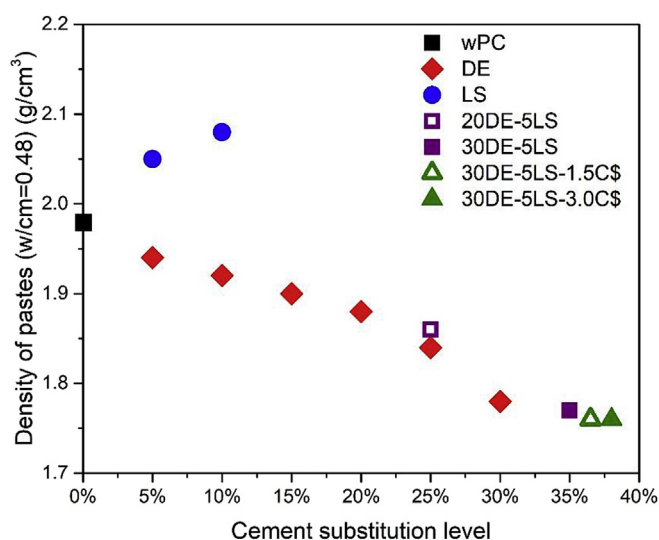


Fig. 3. Density of pastes (w/cm = 0.48) as a function of cement substitution level by mass.

hydrofluorocarbons, and perfluorocarbons)(Capper et al., 2008). The 100% wPC mortar mixture (M-wPC) produced the highest total GWP of 578 kg CO₂-eq/m³ (Fig. 5a) while M-40DE-5LS-3C\$ mortar (52% wPC content) results in the lowest total GWP of 316 kg CO₂-eq/m³. For mortar mixes, the total GWP decreases as an increasing amount of wPC is substituted with SCMs with 27.3% reduction in M-20DE-5LS-3C\$, 36.6% reduction in M-30DE-5LS-3C\$, and 45.4% reduction in M-40DE-5LS-3C\$, compared to M-wPC reference. Among all mortar production activities, cement production has the dominant contribution to the total GWP with the highest at 90.2% in M-wPC control and the lowest at 81.3% in M-40DE-5LS-3C\$. Energy consumption of the mortar mixes (Fig. 5b) at higher substitution level is reduced in a similar trend as for GWP, compared to the M-wPC reference.

The GWP emissions (Fig. 5c) and the energy consumption (Fig. 5d) of the production of the concrete mixes are reduced by lowering the cement content of the binder, in a pattern which resembles that of the mortar mixes. From 100% wPC concrete (C-wPC) to the C-30DE-5LS mix, total GWP decreases by 37% from 423 to 267 kg CO₂-eq/m³ of concrete; and the total energy consumption decreases 27% from 3061 to 2240 MJ/m³. Compared to the mortar mixes, the production of a unit amount of concrete mixes with same DE content (e.g., C-30DE compared to M-30DE), the concrete

mix results in lower GWP emissions and energy consumption because, despite the increase from long-distance transportation of the coarse aggregate, an overall lower binder content and thus less wPC content is the actual decisive factor.

Fig. 6a shows the f_c of mortars and the GWP intensity per unit volume of mortar normalized to its strength. By taking into consideration both the mechanical properties and the environmental impact of the mortars, the GWP intensity is a good measure of the impact of a mix design. Lower GWP intensity and higher compressive strength are achieved simultaneously when more wPC is substituted with SCMs. For example, the M-10DE mortar mix with f_c of 52.5 MPa produces 9.9 kg CO₂-eq.m⁻³/MPa while the M-30DE-5LS-3C\$ mortar mix with compressive strength of 72.1 MPa produces 5.1 kg CO₂-eq.m⁻³/MPa, rising the strength by 37% and reducing GWP intensity by 48%. Therefore, with appropriate modification of mix proportions and selection of SCMs, the GWP intensity of mortars can be controlled with no concern of compromising the strength. Likewise, the energy intensity (Fig. 6b), which is obtained by normalizing the total energy consumption of a mortar mix to its 28-day strength, follows the trend of the GWP intensity of mortar mixes. At DE level of 20–40%, the energy intensity of the mortars decreases by 35–53% compared to the M-wPC mortar reference. Low energy intensity reflects that substitution with DE in mortars is more economical regarding energy use in gaining strength than using plain cement.

The same methodology was used to estimate the GWP intensity (Fig. 6c) and energy intensity (Fig. 6d) of the concrete mixtures; the results of concrete mixes show similar trends with respect to the mortar mixes. The lowest GWP intensity and energy intensity of concrete production are both achieved by the C-30DE-5LS, which has the highest f_c, the lowest GWP, and lowest energy use. On the opposite side, the highest intensities are from the wPC reference concrete mix (C-wPC) with the lowest compressive strength, the highest GWP, and the highest energy use. This contrast reflects that increasing the DE level of the concrete in the studied range is beneficial due to its contribution to the strength gain and the reduction in GWP emissions and energy consumption of concrete production.

Apart from cement production, the contributors to GWP emissions in mortar production, from the greatest to the least, are transportation to the mixing plant (up to 53.8 kg CO₂-eq/m³, amounting to 17.6% of total GWP), production of superplasticizer (up to 10.1 kg CO₂-eq/m³), production of fine aggregate (2.4 kg CO₂-eq/m³), mortar mixing and batching (1.0 kg CO₂-eq/m³), and total SCMs (i.e., DE, LS, and C\$ combined) processing (up to 0.7 CO₂-eq/m³). Transportation to mixing plant, the second largest source of GWP after cement production, decreases with decreasing wPC level

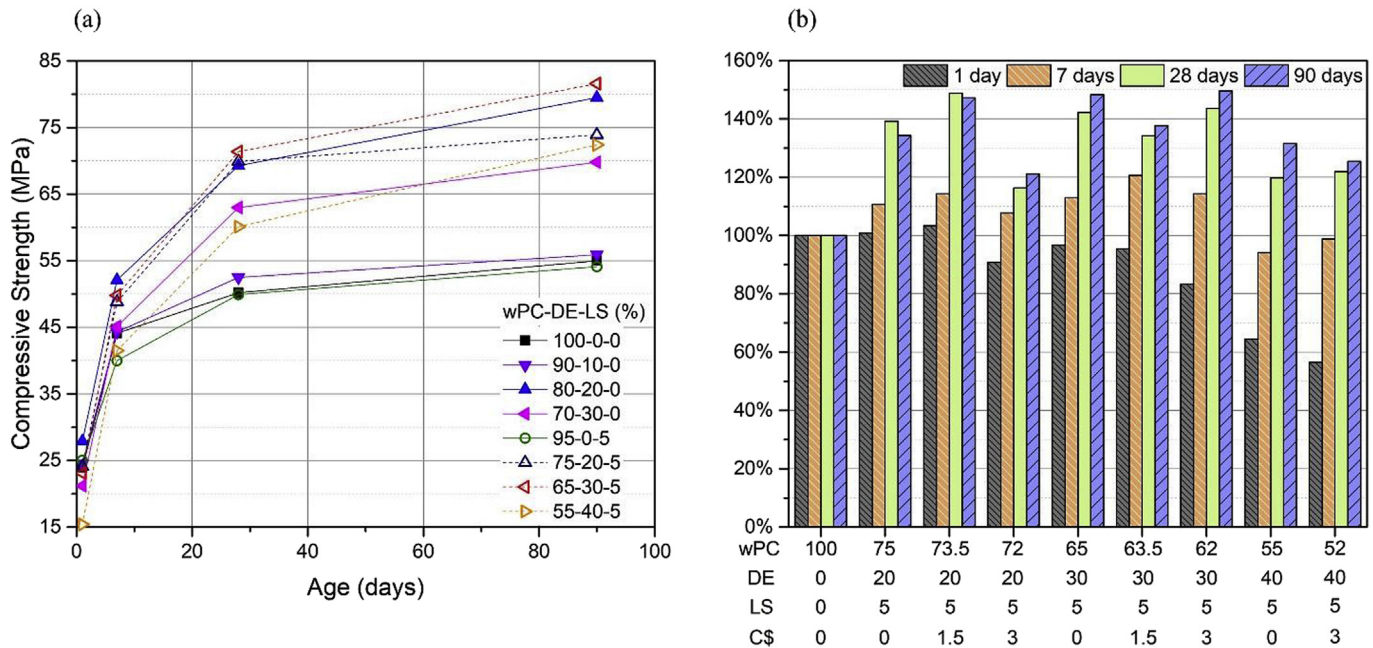


Fig. 4. (a) Evolution of the compressive strength of mortar mixes over time; (b) compressive strength of mixes normalized to wPC control.

Table 9
Compressive strength of mortars.

	Compressive strength (MPa)			
	1 days	7 days	28 days	90 days
M-wPC	23.9	44.1	50.2	55.0
M-5LS	25	40	49.9	54.1
M-10LS	24.6	41.8	46.5	50.2
M-10DE	24.5	44.3	52.5	55.9
M-20DE	27.9	52.1	69.3	79.5
M-30DE	21.2	45.1	63.0	69.8
M-10DE-10LS	22.7	41.0	49.3	56.0
M-15DE-5LS	28.9	54.9	69.8	77.2
M-20DE-5LS	24.1	48.8	69.9	73.9
M-20DE-10LS	20.3	36.8	53.6	59.1
M-30DE-5LS	23.1	49.8	71.4	81.6
M-40DE-5LS	15.4	41.5	60.1	72.4
M-20DE-5LS-1.5C\$	24.7	50.4	74.7	81.0
M-20DE-5LS-3.0C\$	21.7	47.5	58.4	66.6
M-30DE-5LS-1.5C\$	22.8	53.2	67.4	75.7
M-30DE-5LS-3.0C\$	19.9	50.4	72.1	82.3

Table 10
The compressive strength of concretes.

	Compressive strength (MPa)	
	7 days	28 days
C-wPC	30.1	35.5
C-5LS	31.4	36.5
C-10DE	34.9	40.9
C-20DE	35.9	46.7
C-30DE	36.9	48.9
C-20DE-5LS	33.9	46.9
C-30DE-5LS	34.6	50.2

since the cement plant is farther than the DE mine to the mixing plant. The cumulative GWP of mortar production excluding binder-related activities (i.e., SCMs processing and cement production) fluctuates between 54.5 and 59.5 kg CO₂-eq/m³ but weighs differently in the total mortar-production GWP, varying between 12.1% for 5LS to 22.7% for 40DE-5LS-3C\$ due to the decreasing GWP contributions from the binders at increasing substitution levels

with SCMs.

Transportation activities have the second highest energy demand after cement production (Fig. 7). As the substitution level increases, the mortar mixes show a similar pattern in total energy consumption compared to GWP emissions, despite a more noticeable component of the highly energy-intensive production of PCE. The production of PCE reaches as high as 12% of total mortar-production energy use in the M-40DE-5LS-3C\$ mix while in the same mix cement production and transportation are 71% and 14% of total energy use, respectively. Nevertheless, for mixes containing up to 30% DE, the comparatively low dose of PCE keeps the environmental impact of superplasticizer relatively inconsequential to the entire production system.

Fig. 7c and d shows similar results for the concrete mixes. After cement production, transportation is the next largest contributor to GWP emissions during concrete production due to the long delivery distance and high demand for wPC and coarse aggregates, followed by the production of superplasticizer, the production of aggregates, concrete mixing and batching, and SCMs processing. Notably, the proportion of transportation in total GWP and energy use is higher for the concrete than the mortar mixes due to a higher volume fraction of aggregates and the significantly longer transportation distance of the coarse aggregate compared to the fine aggregate. The order of contributions of the major concrete production processes to energy consumption is the same as that to GWP emissions while the proportioning slightly varies due to the high energy intensity in the production of superplasticizers, which still does not exceed 3.2% of the total energy demand.

The SCMs processing (Fig. 8) has minimal contribution to total GWP, ranging up to 0.7 kg CO₂-eq/m³ of mortar, which amounts to 0.3% of GWP from the entire mortar production process at the maximum substitution level of 48% in M-40DE-5LS-3C\$. The processing of DE generates at least 20% lower GWP than LS at the same substitution level (10%) during mortar production. GreenConcrete Tool and previous LCA studies show that more widely used SCMs, FA, GGBFS, and RHA, have ~5, 16, and 9 times as much GWP emissions as LS per unit mass (Celik et al., 2015) (Gursel et al., 2016). The advantage in GWP reduction of DE processing is due

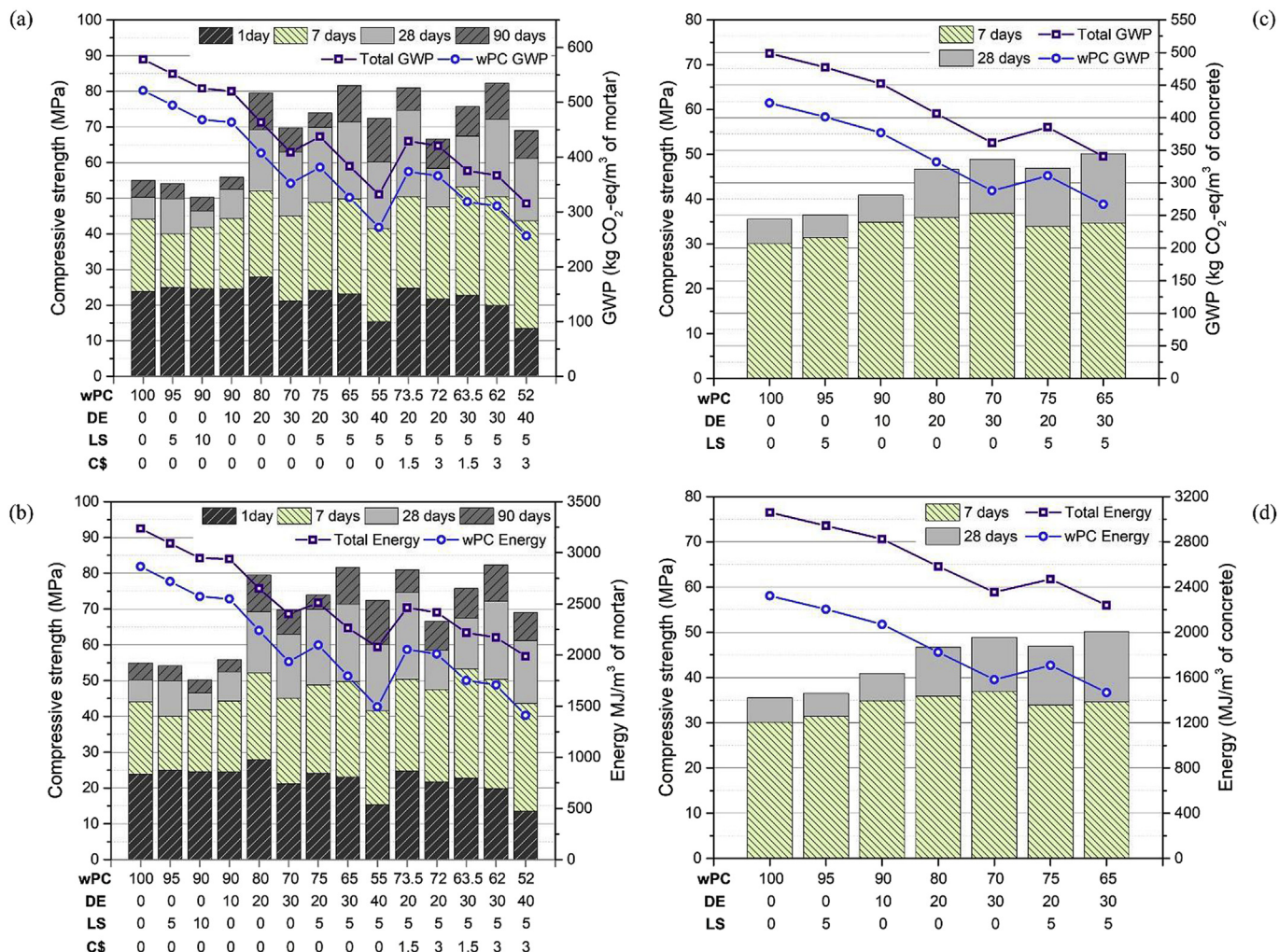


Fig. 5. Compressive strengths of mortar mixes over time and (a) GWP emissions/(b) energy consumption due to total production and cement production; compressive strengths of concrete mixes and (c) GWP emissions/(d) energy consumption.

to the low fuel demand for milling and drying while FA processing includes drying, and RHA and GGBFS processing involves grinding.

The energy use in the processing of SCMs shows a similar pattern to the GWP emissions due to the same activities. Per unit mass, the processing of DE is less energy-intensive than that of LS. With a maximum substitution level at 48% in the M-40DE-5LS-3C\$ mix, the maximum energy consumption in SCMs processing reaches 13.9 MJ/m³ of mortar while the same quantity of wPC induces a much higher energy demand equal to 1301.3 MJ/m³ of mortar, being 93 times more energy-consuming.

The SCMs have a negligible contribution to the total GWP emissions in the concrete production processes. At the maximum substitution level in C-30DE-5LS, the processing of LS and DE accounts for 0.43 kg CO₂-eq/m³ of concrete while, in the same mix, wPC, which forms the rest 65% of the binder, accounts for 267 kg CO₂-eq/m³ of concrete. Similarly, the energy use in the processing of LS and DE is negligible compared with the total energy consumption due to concrete production. The energy for SCMs processing in C-30DE-5LS is 8.6 MJ/m³ of concrete while producing the same quantity of wPC consumes as much as 790 MJ/m³ of concrete.

With decreasing cement levels in the binder, the emissions of each air pollutant species (Table 11) decreases for both mortar (Fig. 9a) and concrete (Fig. 9b) mixes. The wPC mortar has the highest emissions of each air pollutant species while the maximum

reduction achieved by mortars with maximum 48% SCMs level is 24% in CO emissions, 37% in NO_x emissions, 44% in SO₂ emissions, and 33% in PM10 emissions. Likewise, in concrete mixes, from the C-wPC mix to the C-30DE-5LS mix, the reduction is 17% in CO emissions, 21% in NO_x emissions, 30% in SO₂ emissions, and 19% in PM10 emissions.

CO is a primary air pollutant, mostly originated from incomplete combustion of carbonaceous fuels, common in automobile exhaust. The largest source of CO emissions among all processes in mortar production (Fig. 10a) is transportation (~55–65%), due to the high diesel fuel use for long-distance transportation by heavy truck, followed by cement production (~25–40%), aggregate production (~3.5–4.5%), and PCE production (up to ~3%). The SCMs processing slightly contributes to the total CO emissions. Similar for the concrete mixes (Fig. 10b), transportation has the highest contribution (~60%) to total CO emissions, followed by cement production (~30%). The production of coarse aggregate has slightly greater CO emissions compared to fine aggregate. SCMs processing has a negligible contribution to CO emissions in concrete production.

The cement production is the largest source of NO_x emissions during mortar production (Fig. 11a) as the pyroprocessing generates thermal NO_x in addition to fuel NO_x due to high kiln temperature promoting the oxidation of atmospheric nitrogen gas, whereas transportation is the largest source of NO_x during concrete

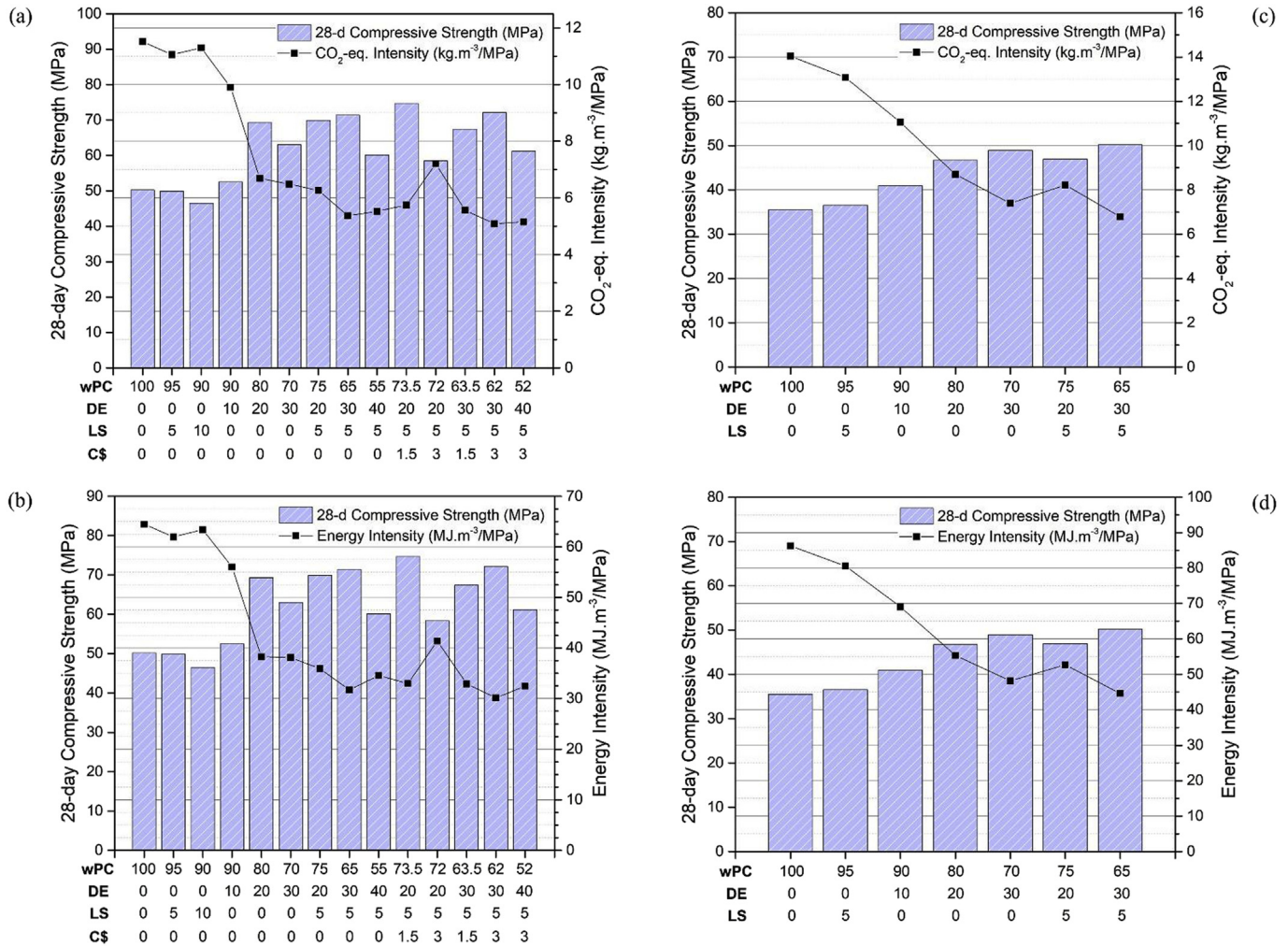


Fig. 6. 28-day compressive strengths of mortar mixes, and (a) GWP intensity/(b) energy intensity; 28-day strengths of concrete mixes and (c) GWP intensity/(d) energy intensity.

production (Fig. 11b). The discrepancy is due to large scale of diesel fuel combustion in the long-distance delivery of the coarse aggregate to the mixing plant, during which fuel NO_x is emitted from the incomplete combustion of diesel. As a consequence, despite the higher cement paste content in the mortar compared to the concrete mix at the same substitution level, the total NO_x emissions during concrete production are higher than that during mortar production. Therefore, increasing the local availability of the ingredients is highly feasible to reduce NO_x emission while switching to cleaner transportation is also viable. The processing of SCMs, the production of aggregates, and mixing and batching contribute negligibly to the total NO_x emissions in both mortar and concrete production.

The primary pollutant SO₂ is most significantly emitted from stationary sources instead of mobile sources due to stricter regulation on sulfur content for diesel and gasoline fuels. In the mortar samples (Fig. 12a), the SO₂ emissions mostly (85–95%) happen during cement manufacturing from the volatile sulfur in combustion fuel and cement ingredients during pyroprocessing, followed by transportation (3–7%) and superplasticizer production which is the highest in M-40DE-5LS-3CS at 7.5% of total SO₂ emission. Similarly, in concrete manufacturing (Fig. 12b), the SO₂ emissions of cement production dominate (75–85%), followed by transportation (around 15%) and the production of superplasticizer (~1–3%). SO₂ emissions due to the SCMs processing and the production of

aggregate are insignificant in both mortar and concrete production.

Particulate matter is both a primary pollutant emitted from mechanical or chemical processes and a secondary pollutant formed from smaller particles or compounds in air. The PM10 emission associated with mortar production is ~150–220 g/m³ of mortar (Fig. 13a), higher with higher wPC content. Major sources of PM10 are transportation to mixing plant and cement production due to fuel use while mortar mixing and batching also have a nonnegligible contribution. During concrete production, transportation, varying around 120g/m³ of concrete (Fig. 13b), is the largest source of PM10, followed by cement production, which decreases steadily with decreasing wPC content. In concrete industry, PM10 emissions are usually related to grinding, and handling of materials and the use of fuels, which is in accordance with the observation that the processing of limestone and DE is inconsequential to PM10 emissions in both mortar and concrete mixes; moreover, it has been reported that due to need for grinding RHA and fuel demand for drying FA, both SCMs result in much higher PM emissions compared to LS at similar mass per unit-volume concrete (Gursel, 2014).

Due to the lack of the information of VOC emissions from transportation activities, only VOC emissions from material production and processing are modeled. For the mortar production (Fig. 14a), the cement production contributes the most to VOC emissions, between 32 and 65 g/m³ of mortar, because of the

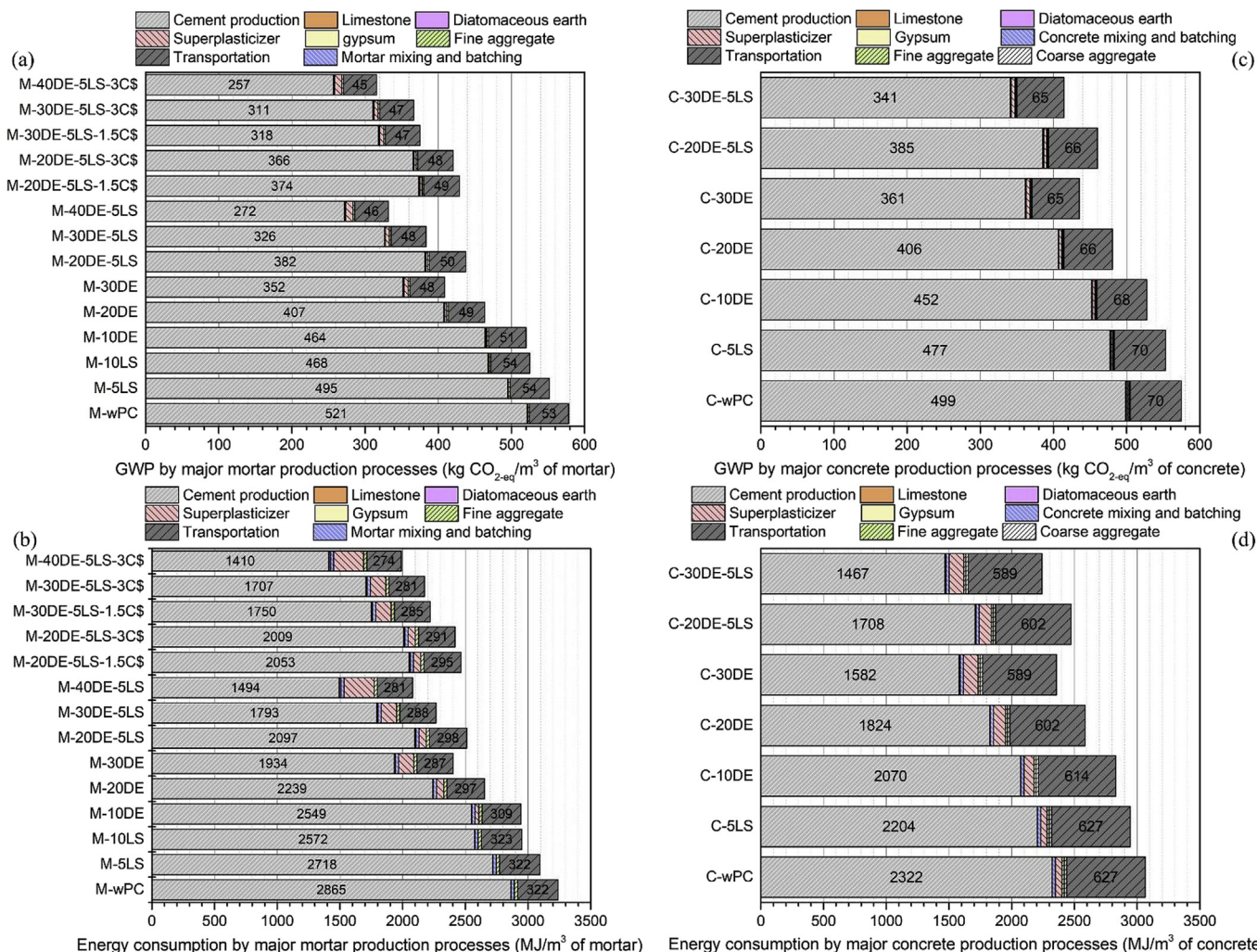


Fig. 7. Distribution of (a) GWP emissions/(b) energy consumption due to major processes during mortar production; distribution of (c) GWP emissions/(d) energy consumption due to major concrete production processes.

volatile impurities in the cement ingredients which escape from the solid phases under intensive heating during pyroprocessing; superplasticizer production, up to 3.8g/m³ of mortar, follows because of its organic nature. A similar trend is seen from the concrete mixes (Fig. 14b), despite a lower contribution of PCE production to total VOC emissions due to material production and processing during concrete production. In both mortar and concrete mixes, despite the increasing emissions during superplasticizer production at a higher substitution level of DE, the overall VOC emissions are nevertheless decreasing from the M-wPC and C-wPC references due to the extremely high emission rate from cement pyroprocessing. Furthermore, with up to 30% DE, the PCE-incurred VOC emissions are minimal. The production of aggregates and the processing of SCMs have a negligible contribution to the total VOC emissions.

4.6.2. Results of alternative scenarios with different transportation distance and means

The study is suited for DE usage in California, assuming the most possible transportation modes and comparatively near locations of material sources. It would provide a more comprehensive insight into the environmental impact of DE usage by examining alternative transportation modes (e.g., using Class 2b truck instead of Class

8b) and longer distance from DE source to concrete plant (e.g., considering DE in Washington and Nevada instead of California). Due to limited space, mixes C-wPC, C-10DE, C-20DE, and C-30DE are selected for a simple and clear demonstration of the effect of changing transportation conditions on GWP emissions and energy consumption. The scenarios in Table 12 are compared with the true scenario of this study with assumptions summarized in Table 4. In scenarios 1–3, the transportation modes are modified; in scenarios 4 and 5, the source of DE is modified and thus its transportation distance varies.

Tables 13 and 14 presents results of modelling of GWP and energy consumption in each scenario with the GWP intensity and energy intensity normalized to the 28-day compressive strength of the corresponding mixes. Switching from class 8b truck to class 2b truck or from barge to class 8b truck significantly rises the total GWP for each mix. Nevertheless, for each scenario, increasing substitution with DE decreases the total GWP of concrete production compared to C-wPC mix, by more than ~19% for changing-transportation-mode scenarios and for more than ~25% for changing-DE-source scenarios. When normalized to 28-day strength, the DE-containing mixes exhibit greater advantage against C-wPC reference, C-30DE showing more than 40% reduction in GWP intensity in all studied scenarios.

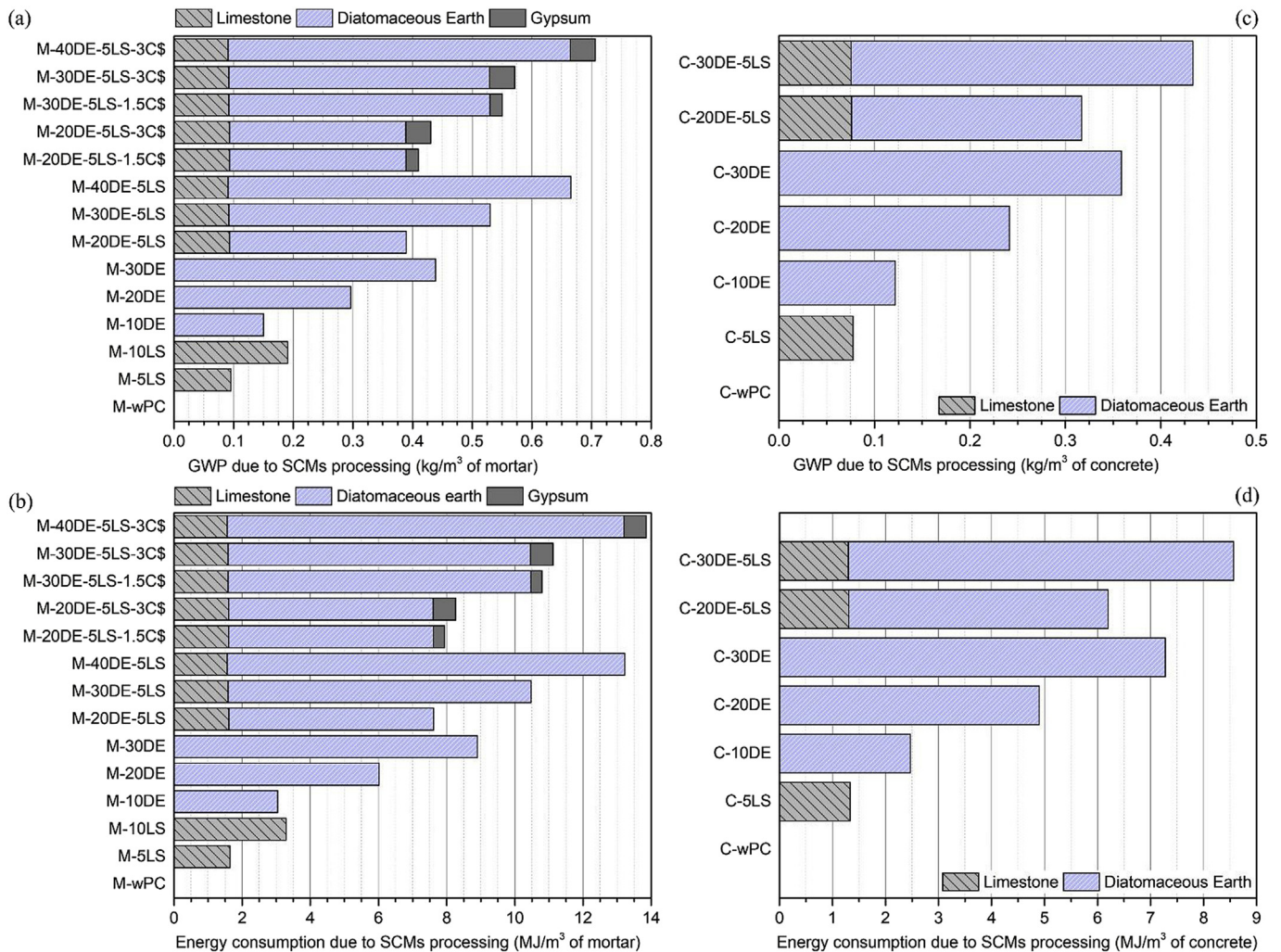


Fig. 8. Distribution of (a) GWP emissions/(b) energy consumption due to SCM processing during mortar production; distribution of (c) GWP emissions/(d) energy consumption due to SCM processing during concrete production.

Table 11

Criteria air pollutants and volatile organic compounds (VOC) emissions per unit volume of mortar.

	CO (kg/m ³)	NOx (kg/m ³)	PM10 (kg/m ³)	SO ₂ (kg/m ³)	Lead (kg/m ³)	VOC (kg/m ³)
wPC	3.20E-01	2.07E+00	2.23E-01	1.14E+00	2.47E-02	6.51E-02
5LS	3.14E-01	2.01E+00	2.17E-01	1.08E+00	2.35E-02	6.19E-02
10LS	3.07E-01	1.94E+00	2.12E-01	1.03E+00	2.23E-02	5.87E-02
10DE	3.01E-01	1.90E+00	2.07E-01	1.02E+00	2.20E-02	5.85E-02
20DE	2.82E-01	1.74E+00	1.91E-01	9.10E-01	1.93E-02	5.19E-02
30DE	2.66E-01	1.58E+00	1.75E-01	8.07E-01	1.67E-02	4.60E-02
20DE-5LS	2.76E-01	1.67E+00	1.86E-01	8.56E-01	1.82E-02	4.88E-02
30DE-5LS	2.61E-01	1.51E+00	1.70E-01	7.56E-01	1.55E-02	4.31E-02
40DE-5LS	2.50E-01	1.37E+00	1.55E-01	6.65E-01	1.29E-02	3.81E-02
20DE-5LS-1.5C\$	2.72E-01	1.64E+00	1.83E-01	8.39E-01	1.78E-02	4.78E-02
20DE-5LS-3C\$	2.68E-01	1.61E+00	1.80E-01	8.22E-01	1.74E-02	4.68E-02
30DE-5LS-1.5C\$	2.57E-01	1.49E+00	1.68E-01	7.37E-01	1.52E-02	4.20E-02
30DE-5LS-3C\$	2.53E-01	1.46E+00	1.65E-01	7.21E-01	1.48E-02	4.11E-02
40DE-5LS-3C\$	2.43E-01	1.31E+00	1.50E-01	6.32E-01	1.22E-02	3.62E-02

*Note: The lead and VOC emissions do not include emissions due to transportation.

In the model for energy consumption, despite increased energy consumption with respect to the original scenario due to using less eco-friendly transportation modes and DE from farther sources, compared to C-wPC, all DE-containing mixes lead to lower total energy consumption except in scenario 5 where the more practical

delivery methods of DE over a distance of 1285 km is by the more energy-saving rail instead of by truck as assumed. Nevertheless, when considering the energy intensity, the higher strength gain of DE-containing mixes promises decreased value for all mixes in all scenarios being studied, with more than 40% reduction in scenarios

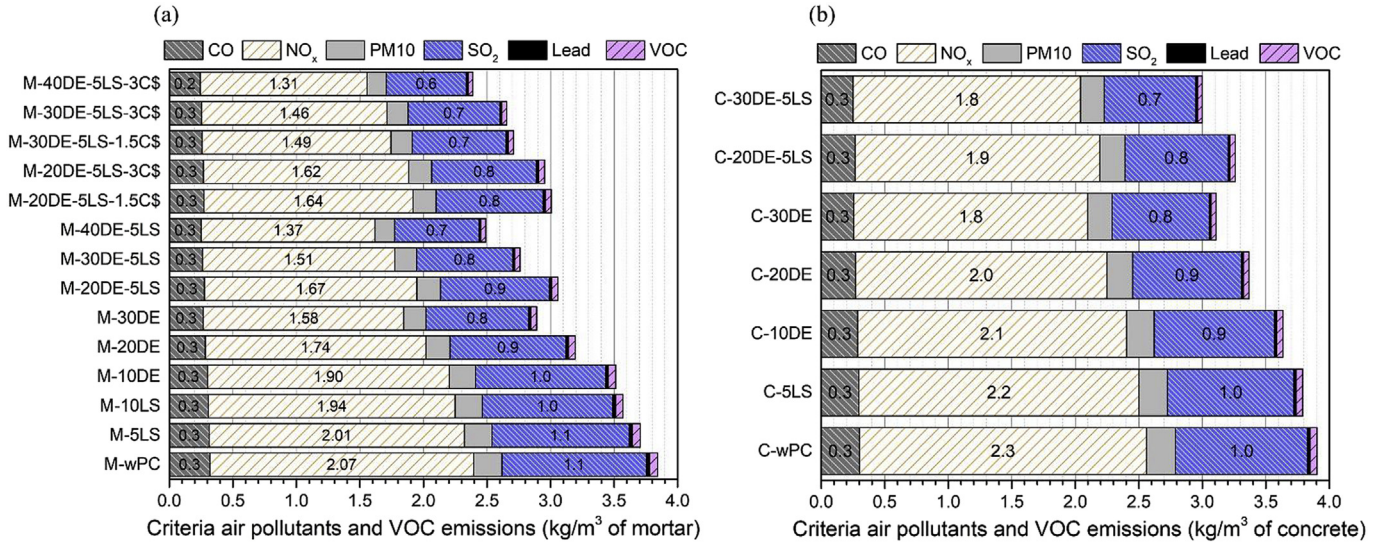


Fig. 9. Air pollutant emissions due to (a) mortar and (b) concrete production.

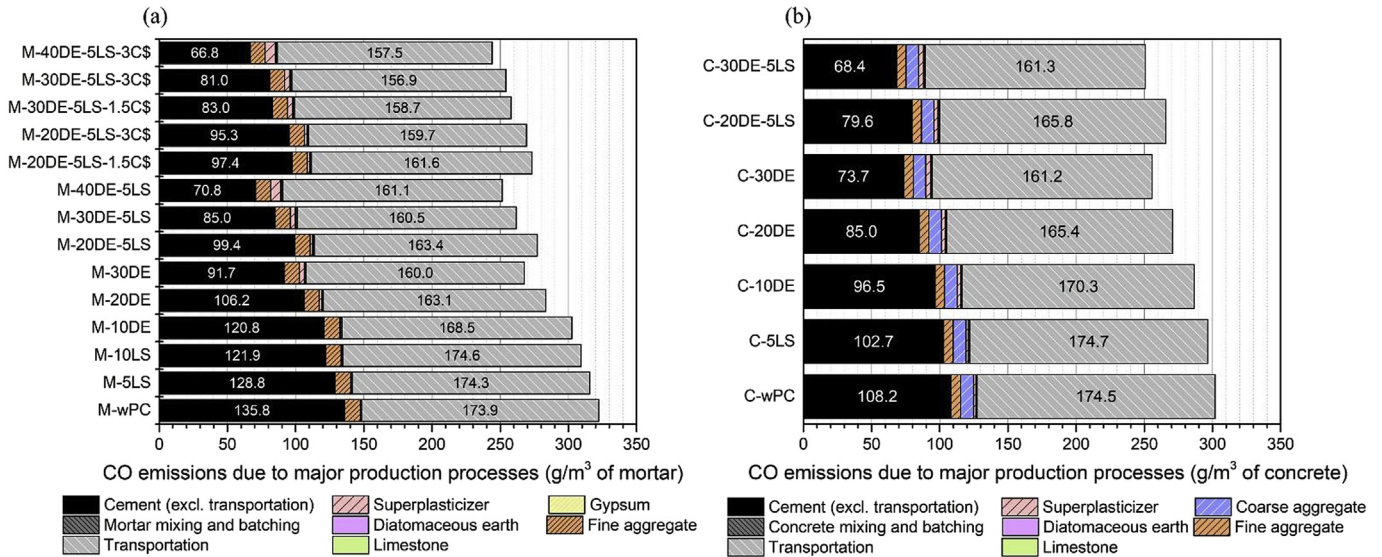


Fig. 10. Distribution of CO emissions by processes of (a) mortar and (b) concrete production.

1 to 4 and ~26% in scenario 5.

Therefore, the study of the alternative scenarios indicates the importance of selecting greener transportation modes and nearer source locations; failure of either could result in over 50% in GWP emissions and energy use. Though, shown by the 28-day intensities, increase in mechanical properties from DE makes it environmentally worth even to import from distant sources.

5. Discussion

All studied mortar mixes with DE both improve strength gain and reduce GWP. However, the optimal mix design regarding strength and environmental impacts is of 30 wt% DE level, despite that the 40 wt% DE mixes produce less environmental impacts; the high energy demand and VOC emissions from the production of the high dose of PCE for 40 wt% should not simply be neglected.

For mixes with most common pozzolan (e.g., fly ash), 90- or 365- day strengths (Celik et al., 2014b, 2015) are preferred to 28-

day strength for the calculation of normalized energy and GWP intensity due to low early strength development of these pozzolan-containing mixes. However, DE-containing mortar and concrete have no such limitation because even the 28-day strengths are higher than that of the wPC reference due to the high reactivity of DE. The 28-day GWP intensity of 30 wt% DE concrete mix is 42% reduced compared to that of wPC concrete reference while the GWP intensity of 30% FA mix is less than 10% reduced at 28-day and ~25% reduced at 365-day compared to PC concrete reference (Celik et al., 2015) (Gursel et al., 2016). Therefore, mixes with considerable substitution levels of DE have high potential to be practically utilized in column design without increasing the cross-section area and self-weight of the column to secure the demanded capacity.

In the real-scenario study, the GWP, energy use, and emissions due to transportation activities during concrete production, despite the reduced amount of wPC usage, are not appreciably lower than those during mortar production since the environmental impacts from long-distance transportation of coarse aggregate is significant.

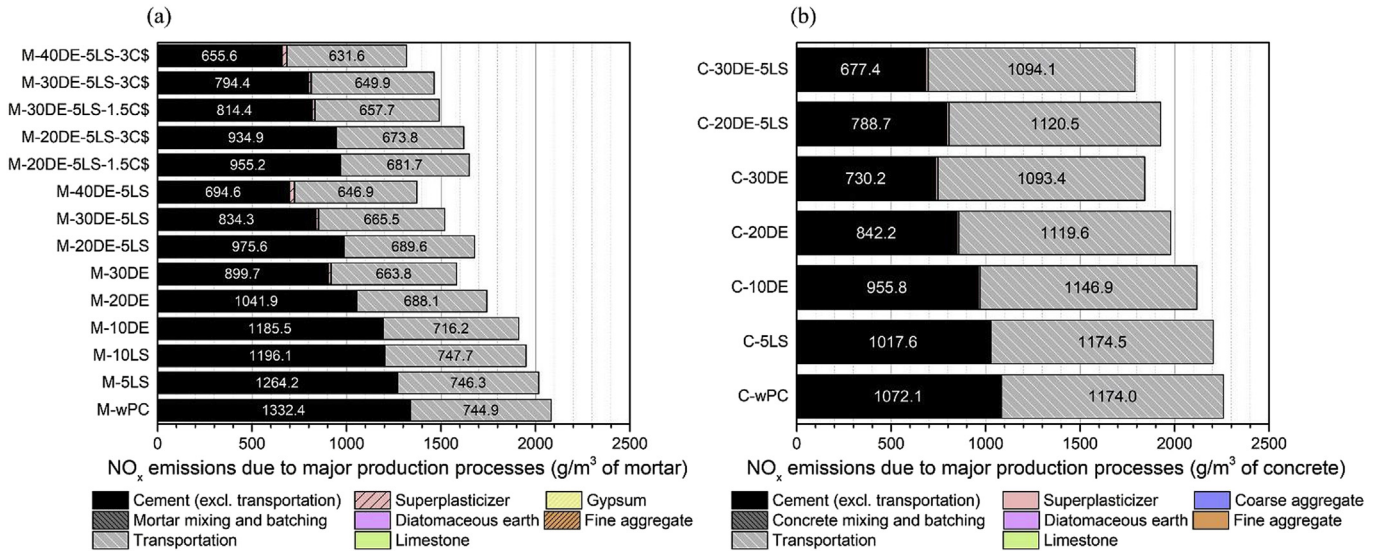


Fig. 11. Distribution of NO_x emissions by processes of (a) mortar and (b) concrete production.

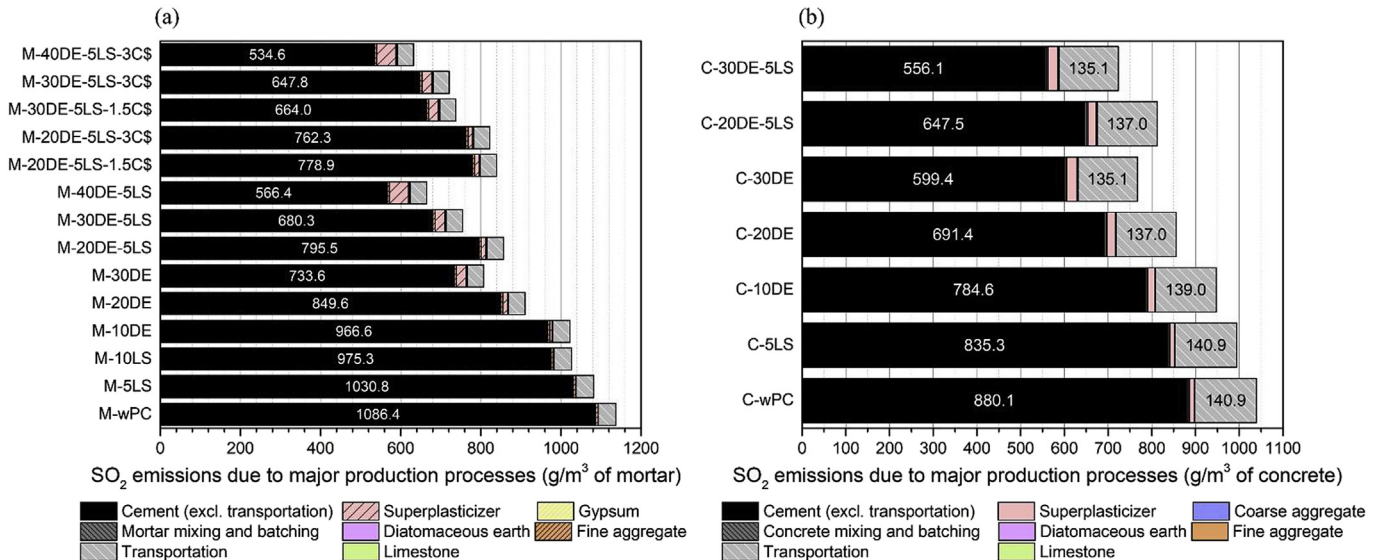


Fig. 12. Distribution of SO₂ emissions by processes of (a) mortar and (b) concrete production.

For SCMs and aggregate, the influence from distance and means of delivery may outweigh the environmental benefits attained by the clean electricity mix at the location of the material source. This observation exhibits the importance of the material origins to the overall environmental impact due to environmental expense of delivery and notable electricity grid mix difference, more insights on which are shown in the five alternative scenarios. Thus, the environmental impact of a mix design can be implicitly (i.e., with no change in mix proportions) ameliorated by selecting materials of higher local availability and more environmental-friendly means of transportation and manufacturing.

The advantages of DE from the LCA perspective is most valid in the western U.S. and in countries, e.g., China, Czechia, and Turkey, where DE source is also abundant. For other regions (e.g., Singapore), where lack direct DE (as well as cement) supply, the transportation between DE deposits (e.g., Thailand or Malaysia) and mixing plant should be taken into extra consideration, in a similar logic to using FA and GGBFS for concrete mixing in California where

there are few sources. The current LCA case study is geographically limited, based on assumptions of distances, transportation methods, and energy mix, etc. specific to California, which may not accurately represent the scenario in other DE-rich regions with different energy mix and characteristic transportation mode or in regions relying on imported DE manifesting higher impact from transportation. Therefore, future LCA study can be modeled for scenarios, e.g., in DE-abundant China where coal is of significantly higher component in fuel mix than in CA or in DE-wanting African countries also short of cement production.

Only a number most commonly examined environmental impactors are modeled in our study; in addition, due to inadequacy of the database, transportation information is lacking for the emissions of VOC and lead. The absence of data should be fixed for future study especially when modelling for non-local DE use where impacts from transportation are of major emphasis. It should be admitted that, despite great benefits shown from the reduction in GWP, energy use, and criteria air pollutant emissions by the

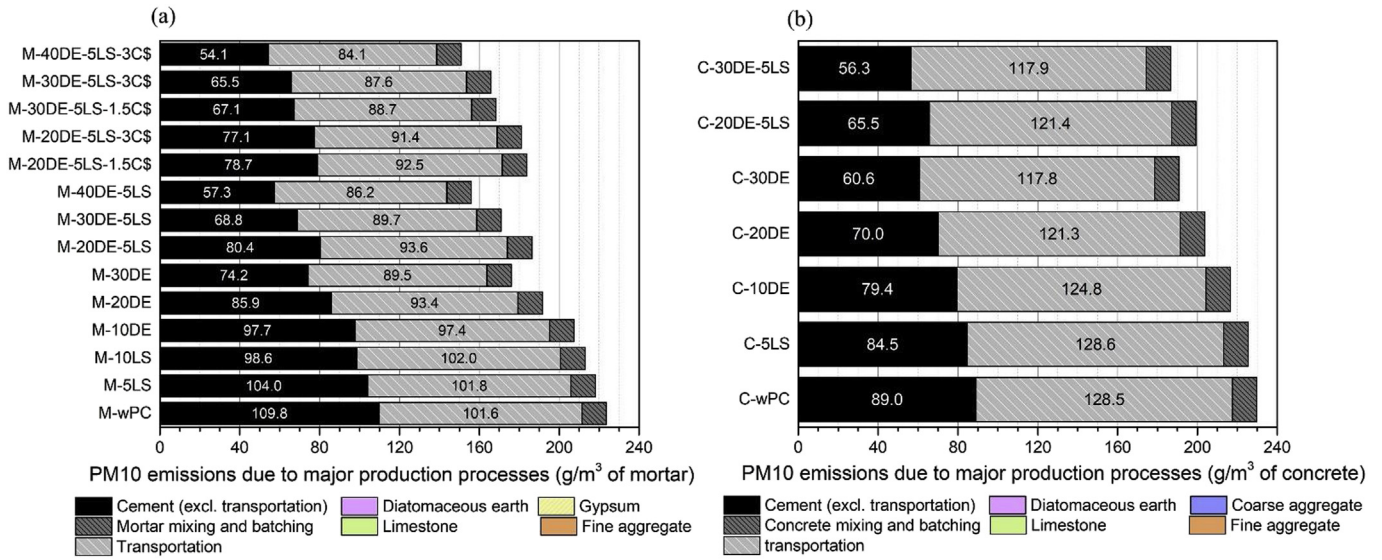


Fig. 13. Distribution of PM emissions by processes of (a) mortar and (b) concrete production.

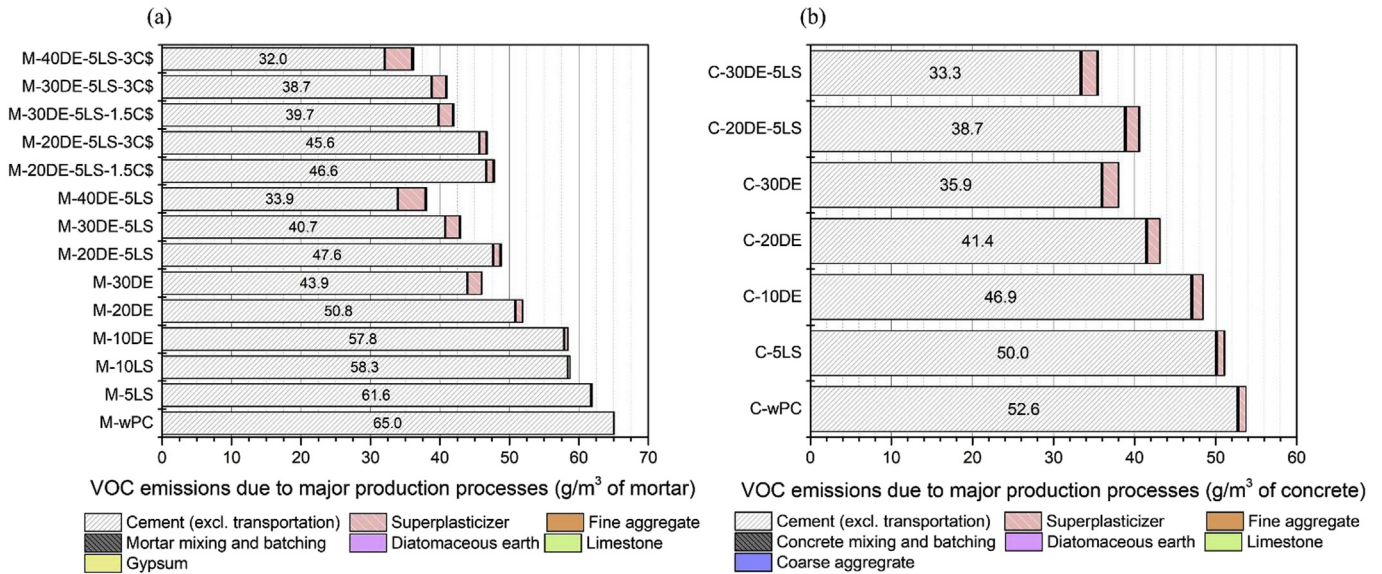


Fig. 14. Distribution of VOC emissions by processes of (a) mortar and (b) concrete production.

Table 12

Assumptions of transportation distance and mode for alternative scenarios.

	Scenario 1		Scenario 2		Scenario 3		Scenario 4 ^a		Scenario 5 ^b	
wPC	Truck 8B	637 km	Truck 2B	637 km	Truck 2B	637 km	Truck 8B	637 km	Truck 8B	637 km
DE	Truck 8B	391 km	Truck 2B	391 km	Truck 2B	391 km	Truck 8B	568 km	Truck 8B	1285 km
Fine aggregate	Truck 8B	53 km	Truck 2B	53 km	Truck 2B	53 km	Truck 8B	53 km	Truck 8B	53 km
Coarse aggregate	Truck 8B	1840 km	Truck 2B	40 km	Truck 2B	1840 km	Truck 8B	40 km	Truck 8B	40 km
PCE	-	-	Barge	1800 km	-	-	Barge	1800 km	Barge	1800 km
	Rail	4463 km	Rail	4463 km	Rail	4463 km	Rail	4463 km	Rail	4463 km
	Truck 2B	5 km	Truck 2B	5 km	Truck 2B	5 km	Truck 2B	5 km	Truck 2B	5 km

^a Scenario 4 assumes a DE source from Medford, Nevada.

^b Scenario 5 assumes a DE source from Seattle, Washington.

substitution with DE, there is the possibility of increasing environmental burdens in other unexamined criteria. As has been modeled by previous LCA studies on different commercial cement types, a more comprehensive list of environmental impacts is

suggested for future LCA of DE-containing materials: ozone depletion potential, photochemical oxidant formation potential, terrestrial acidification potential, eutrophication potential, human toxicity potential, ecotoxicity, and land use, etc. (Stafford et al.,

Table 13
GWP (kg CO₂-eq/m³) and GWP intensity (kg CO₂-eq.m⁻³/MPa) of concrete mixes.

	Original		Scenario 1		Scenario 2		Scenario 3		Scenario 4		Scenario 5	
	GWP	Intensity	GWP	Intensity	GWP	Intensity	GWP	Intensity	GWP	Intensity	GWP	Intensity
C-wPC	499	14.0	655	18.4	524	14.8	779	21.9	499	14.0	499	14.0
C-10DE	452	11.1	607	14.8	476	11.6	729	17.8	453	11.1	457	11.2
C-20DE	406	8.7	560	12.0	429	9.2	680	14.6	408	8.7	416	8.9
C-30DE	361	7.4	514	10.5	384	7.8	632	12.9	364	7.4	376	7.7

Table 14
Energy consumption (MJ/m³) and energy intensity (MJ.m⁻³/MPa) of concrete mixes.

	Original		Scenario 1		Scenario 2		Scenario 3		Scenario 4		Scenario 5	
	Energy	Intensity	Energy	Intensity	Energy	Intensity	Energy	Intensity	Energy	Intensity	Energy	Intensity
C-wPC	3061	86.2	5008	141.1	4172	117.5	9261	260.9	3061	86.2	3061	86.2
C-10DE	2825	69.1	3538	86.5	3891	95.1	8933	218.4	2831	69.2	3497	85.5
C-20DE	2583	55.3	3291	70.5	3607	77.2	8604	184.2	2595	55.6	3300	70.7
C-30DE	2354	48.1	3055	62.5	3335	68.2	8287	169.5	2371	48.5	3115	63.7

2016) (Moretti and Caro, 2017), as public concerns on ozone layer depletion, photochemical smog, and acid rain, etc. are as intensive. Furthermore, the LCA study can be expanded from cradle-to-gate to cradle-to-grave by examining the durability of DE-containing mixes. High durability leads to low cost for operation and maintenance and long useful life, and vice versa. Environmental impacts at decommissioning and disposal are also concerned in a cradle-to-grave approach.

Last but not least, due to the natural formation of DE in a variety of environment, the amorphous-silica content, impurities, the specific surface area and other properties of the DE are source-specific; thus, further study is encouraged to compare the reactivity of the differently sourced DE and the resulted different mechanical properties of the DE-containing concrete mixes. Moreover, spent DE recycled from other industries is also a potential subject for study.

6. Conclusions

This paper studies the early-age behavior, mechanical properties, and environmental impacts of different “green” mortar and concrete mixtures containing DE and/or LS, and the main points are:

1. With a low dose of superplasticizer, paste, mortar, and concrete mixtures containing DE and LS with 35 wt% cement substitution can be efficiently produced. Setting time, slump, and soundness of blends containing up to 30 wt% DE are comparable to white Portland cement reference at the same water-to-cementitious materials ratio. The density of PC-DE-LS blends decreases as DE content increases.
2. Mortars with DE level of 10–30 wt% have higher compressive strengths compared to wPC control at 7, 28, and 90 days. Strengths of mortar with 20% DE at 28 and 90 days are 70 and 80 MPa, respectively, 40% higher than those of wPC mortar. The 28- and 90- day strengths of mortars with 40 wt% DE and 5 wt% LS are still 20%–30% higher than wPC. Adjusting gypsum content by 1.5–3 wt% benefits the strength gain of mortars with high DE levels.
3. The concrete mixtures containing 10–30 wt% DE attain 15–38% higher compressive strength than wPC control at 7 and 28 days. 28-day strength of 30DE-5LS concrete attains 50 MPa, 40% higher than wPC control.
4. In the production of mortar and concrete mixes, cement production and transportation contribute to ~80–90% and ~10–15% of the total GWP emissions, respectively, while contributing to ~70–80% and ~10–25% of total energy use. For the production of mixes with as much as 30% DE substitution level, the processing of SCMs and aggregates and the production of PCE do not have significant influence to total GWP and energy use.
5. For 10–30 wt% DE mortar mixes with total substitution level of 10–38 wt%, total GWP is reduced by 10–37% and 28d-GWP intensity is reduced by 14–56%; total energy use is reduced by 9–33% and energy intensity is reduced by 13–53%.
6. For 10–30 wt% DE concrete mixes with total substitution level of 10–35 wt%, total GWP is reduced by 9–32% and GWP intensity is reduced by 21–52%; total energy use is reduced by 7–23% and energy intensity is reduced by 19–46%.
7. In mortar and concrete production, cement production has the greatest contribution to SO₂ emissions due to high sulfur content in pyroprocessing fuel, followed by transportation, which has the greatest contributions to CO and PM emissions due to incomplete combustion of fuel, followed by cement production. Cement production and transportation are the first and second largest emission source of NO_x in mortar production while the sequence is reversed in concrete production due to long transportation distance of coarse aggregate. SCMs processing has inconsequential contributions to emissions in every circumstance.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2019.03.077>.

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