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Eco-friendly mortar with high-volume diatomite and fly ash: Performance and life-cycle assessment with regional variability

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ABSTRACT

Replacing Portland cement with supplementary cementitious materials (SCMs) reduces the high environmental impacts of cement production. However, in western U.S., such reduction is limited by the shortage of local SCMs. The use of diatomite (DE), abundantly deposited in western U.S., into cementitious materials can mitigate this local shortage. In this study, up to 60% cement was replaced with DE, fly ash (FA), and limestone, for reduced environmental impacts of mortar production. The workability and compressive strength of mortars with high-volume SCMs were comparable to 100%-cement mortar at optimized mix proportions. The energy consumptions, global warming potential (GWP), and air pollutants emissions of the production of 13 mixes were evaluated with cradle-to-gate life-cycle assessment in a California-based scenario. For a more comprehensive understanding of the regionally-variable greenness of the mixes, scenarios of mortar mixing in 11 U.S. states were modeled to estimate the energy consumptions and GWP from different processes, under the consideration of source locations of cement, FA, and DE in 18 states/provinces in U.S. and Canada and the geographically-dependent electricity grids and transport distances. In all cases, the environmental impacts are proven greatly reduced with the green mixes. Through optimization of transportation modes and electricity grids at resource origins, environmental impacts of producing green cement-based materials are further reduced in SCMs-lacking regions. The combined use of DE and FA in blended cement is promising for both mechanical and environmental benefits even in regions without their local deposits.

1. Introduction

The production of Portland cement (PC) in 2018 generated ~9% of global anthropogenic CO_2 emissions (USGS, 2019) and is the third-largest industrial energy consumer with 7% annual contribution (Diaz-Loya et al., 2019). The demand of PC continues to rise, with a projected 50% increase in annual production of PC by 2050 (Monteiro et al., 2017). PC production is under momentous pressure to lower its carbon footprint (Juenger et al., 2011).

Partial replacement of PC with supplementary cementitious materials (SCMs) is a common strategy to reduce CO_2 emissions and energy consumptions during concrete production (Fan and Miller, 2018). SCMs are typically pozzolanically reactive (alumino)siliceous powders (Snellings et al., 2012), some of which, e.g., blast furnace slag (BFS) and high-Ca fly ash (FA), have cementitious reactivity as well (Li and Li, 2015). The concrete industry traditionally relied on the stream of industrial byproducts or wastes as the primary source for SCMs, e.g., BFS and silica fume (Miller et al., 2015). Unfortunately, the projected supply of these SCMs is dwindling significantly, not satisfying the prospective increasing demand of the construction industry (Juenger et al., 2019). Thus, investigating potential sources of SCMs is crucial in the development of modern concrete industry. Moreover, since many of currently-used SCMs are not geographically well distributed (e.g., BFS is rarely produced in western states in U.S.) (Juenger et al., 2019), the development of new types of SCMs can mitigate the local shortage.

Diatomite (diatomaceous earth, DE) with over 300 Mt deposited in western North America (Wallace et al., 2006) can partially offset the local deficiency in SCMs in the region. Low-grade DE, which intermixes with impurities, cannot be used in other applications

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(i.e., brewery, winery, and distillery) (Letelier et al., 2016). Although raw DE is not an industrial byproduct, the CO_2 emission of mining and processing of low-grade DE is significantly lower than that of manufacturing PC (Yilmaz and Ediz, 2008). The porous nature of DE results in high water-demand in concrete production, limiting its replacement level of PC to ~10% (Starnatakis et al., 2003). With the use of superplasticizers, the replacement level can reach 40% (Li et al., 2019), and the strengths of PC-DE blends were significantly improved. But the high superplasticizer demand increases the cost, lowering the industry's interest. PC-DE-FA blends have two potential advantages over PC-DE blends and high-volume FA cement: A) FA lowers the superplasticizer-demand and the cost, and B) DE may mitigate the low early strength of high-volume FA cement. Therefore, PC-DE-FA blends may maintain strength at higher replacement levels with limited superplasticizer demand and thus achieve even lower CO_2 emissions and energy consumptions in its manufacture and lower cost compared to PC-DE blends.

DE and FA activated by alkali activators have been used to prepare alkali-activated materials (AAMs) for construction use (Sinsiri et al., 2012). However, DE-FA AAMs and DE AAMs often exhibit significantly low strengths, limiting its industrial application (Arbi et al., 2013). PC-DE-FA blends seem to be more practical due to potential high strengths, which remain to be validated – The physical and mechanical properties of PC-DE-FA mixes have not been explored yet, neither have the environmental impacts of manufacturing PC-DE-FA blends. Only those of manufacturing DE-containing mortars (FA-free) have been studied to a limited extent: the energy consumptions and global warming potential (GWP) are found lower than PC reference (Ahmadi et al., 2018); however, the influences of transportation from and varying energy grids at different raw material sources on the environmental impacts have not considered (Ahmadi et al., 2018). In addition, the working hypotheses and the scope of the life cycle assessment in the existing literature (Ahmadi et al., 2018) are not very clear.

Western U.S., with the largest reserve of DE and conventional SCMs shortage in a few states, is most relevant to study the effectiveness of DE-containing blends. Because the greenness of electricity grids and transportation approaches significantly differs over the Western U.S. region, the geographically varying electricity grid and the availability of DE, FA, and PC among states should be carefully considered when the environmental impacts of blends production are evaluated. For example, coal contributes to over 90% of Wyoming's electricity grid while two thirds of electricity resources in Oregon is renewable (Gursel, 2014). The uncertainty in how transportation and electricity grid influence the overall environmental impacts of the DE, FA-containing mixes should be eliminated. This information is useful for fine-tuning and maximizing the greenness of utilizing this new formulation of cement-based materials.

In this study, green binders of PC replaced with high volume of DE and FA were prepared to validate the high strength of PC-DE-FA blends. The early-age properties (setting times, workability, and soundness) of the pastes were determined. With cradle-to-gate Life-cycle assessment (LCA) approach, energy consumptions, air pollutant emissions, and GWP were assessed for replacement levels up to 60%. A Berkeley-based LCA case study was conducted to demonstrate the efficacy of PC-DE-FA in terms of eco-performance (combined environmental and mechanical benefits), followed by a more comprehensive LCA study to evaluate the potential regional variability in the environmental impacts of a green mix design, regarding the non-uniform resource distribution of PC, DE, and FA. Mortars mixed in 11 western-U.S. states with optimized material source selection were modeled to analyze the impact of transportation and different energy grids on the total energy consumptions and GWP. Therefrom, the optimized mortar mix proportions were proposed.

This study fills the gap between the existing sporadic LCA case studies on DE-containing mixes and the more generalized understanding of the efficacy of the high-volume green blends, by examining in detail the variation in total environmental impacts from different transportation approaches and energy grids over the vast region of western U.S. This study demonstrates the approach to maximize the environmental benefits through producing green cement-based materials in regions with certain SCMs shortage. Relevant international policy, regulation, and approaches are also discussed.

2. Materials and methods

2.1. Materials and mixtures

This study used ASTM Type I Portland cement with 57.3% alite, 30.1% belite, 4.6% C_3A, and 3.1% calcium sulfates. As-received diatomite containing ~83% amorphous silica and ~9% montmorillonite was used. Limestone powder (LS, 99% calcite) was used. Class-C fly ash with 83.2 wt% amorphous and 1.6% anhydride was used. The chemical compositions and physical properties of PC, DE, FA, and LS are listed in Table 1. The specific gravity of quartz sand was 2.65.

Mix proportions of the blends are listed in Table 2. All samples were prepared with a water/binder ratio (w/b) of 0.48. Mortars were prepared with a sand/binder ratio of 2.75. A polycarboxylate ether (PCE) superplasticizer with 40 wt% solid was used to adjust the workability of samples when necessary.

2.2. Methods

2.2.1. Setting time, slump, consistency, and volume soundness

Pastes were mixed at 18 °C using a 5L Hobart mixer for 2 min at 136 RPM and another 2 min at 281 RPM. The water demand for normal consistency of blends was tested per (ASTM C187-16, 2016). Initial and final setting time of pastes were measured by Vicat needle penetration tests (ASTM C191-19, 2019). Volume soundness of the pastes was determined using Le-Chatelier method (EN 196-3: 2016, 2016). For mini-slump test, a cone with downscaled Abrams cone geometry (3.5 mm top diameter, 6 mm bottom diameter, and 6 mm height) was used (Tan et al., 2017). The test was conducted by placing a cone on a borosilicate glass sheet. Freshly mixed pastes were poured into the pre-wetted cone, followed by compaction. The cone was lifted, and the spread of the pastes was recorded at

Table 1
Chemical composition (wt.%) and particle size distribution of PC, FA, DE, and LS.

	PC	DE	FA	LS
SiO_2	20.55	85.63	38.71	1.09
Al_2O_3	4.42	3.96	18.88	0.22
Fe_2O_3	0.26	1.04	6.56	0.15
CaO	66.28	0.57	22.94	55.09
MgO	0.86	0.46	4.04	0.50
SO_3	4.10	<0.01	1.01	<0.01
Na_2O	0.04	<0.01	2.1	0.02
K_2O	0.20	0.14	0.5	0.01
TiO_2	0.07	0.26	1.18	0.02
P_2O_5	0.04	0.09	2.2	0.04
MnO	0.01	0.02	–	0.01
SrO	0.04	0.01	0.54	0.01
LOI	1.64	7.74	0.39	42.83
Free CaO	0.2	–	0	–
D10 [μm]	1.45	1.24	1.05	0.92
D50 [μm]	10.94	6.77	9.96	2.98
D90 [μm]	36.85	18.41	40.8	8.16
Specific surface area [m^2/g]	1.18	27.8	1.47	2.81

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

	PC [wt.%]	DE [wt.%]	FA [wt.%]	LS [wt.%]	PC replacement [wt.%]
PC	100	–	–	–	–
15FA	85	–	15	–	15
30FA	70	–	30	–	30
15DE	85	15	–	–	15
30DE	70	30	–	–	30
30DE-15FA	55	30	15	–	45
15DE-30FA	55	15	30	–	45
30DE-10LS	60	30	–	10	40
30DE-15FA-5LS	50	30	15	5	50
30DE-30FA	40	30	30	–	60
30DE-25FA-5LS	40	30	25	5	60
25DE-30FA-5LS	40	25	30	5	60
30DE-20FA-10LS	40	30	20	10	60

30s.

2.2.2. Compressive strength

All mortars were mixed at 18 °C, then cast in 50 mm cubic molds and cured in lime-saturated water at 18 °C until measurement. The cubes were compressed using a hydraulic testing machine at loading rate of 0.5 MPa/s (ASTM C109/C109M-16a, 2016). Additional cubes were casted and cured at 26 °C and 35 °C and tested under the same laboratory conditions.

2.2.3. Life cycle assessment

2.2.3.1. LCA methodology and scope. LCA consists of A) definition of goal and scope, B) life cycle inventory analysis (LCI) within the scope, C) life cycle impact assessment (LCIA) based on LCI, and D) interpretation of A-C (ISO-14040, 2006). The goal and scope are pivotal to the LCA results (Saynajoki et al., 2017; Peters, 2016). In this study, a process-based LCA is conducted by a cradle-to-gate approach (Fig. 1) with 1-m^3 ready-mixed mortar (containing the 13 different blended binders) as the functional unit – extraction or processing of raw materials as the cradle and mixing and batching as the gate to the use stage (Gursel et al., 2016).

The study is conducted with the GreenConcrete LCA Tool database (Gursel, 2014). The tool gathers North American-based information about inputs (e.g., energy and fuel) and outputs (e.g., emissions) of individual processes (e.g., cement pyroprocessing and transportation) for LCI and is embedded with TRACI for mid-point oriented LCIA (Bare, 2011).

2.2.3.2. LCI assumptions. In the study, the LCI results include air pollutant (CO, SO_2, NO_X, PM, and VOC) emissions and energy consumptions for every process during mortar production. Technological alternatives in cement production and transportation modes/distances are critical LCI assumptions inputted to the GreenConcrete LCA Tool (Table 3). “Various” listed for material supplier locations and transportation distances is for case-to-case specification in next sub-sections. The energy input (electricity grid mix) of each region is listed in Table S1 and in (Gursel, 2014).

Assumptions for the Berkeley-based LCA study. For the scenario of mortar mixing in Berkeley, CA, the sources and transportation of the raw materials are summarized in Table 4. Short-distance delivery (<250 km) is modeled by truck (Strocko et al., 2014); for long distance transportation, 50 km of the total distance is modeled as truck delivery while the rest as rail carriage. The electricity grids in California, Arizona, and Ohio (Table S1) are incorporated in the LCA calculation for the production/processing of raw materials and mixing and batching of mortar.

Assumptions for the LCA study based on 11 western U.S. states. Beside the Berkeley-based case study, the possibilities of mixing the 13 mortars in Table 2 in 11 western states –Arizona (AZ), California (CA), Colorado, Idaho (ID), Montana (MT), Nevada (NV), New Mexico (NM), Oregon (OR), Utah (UT), Washington (WA), and Wyoming (WY)– are examined. The relevant regions (the 11 states, and neighboring states/provinces) are possible sources of material supply. LS has wide distribution (Fig. 2), DE is exclusively clustered in western states (Wallace et al., 2006), and Class-C FA is mainly supplied from Wyoming and Texas.

For each mix design and for each state of concern, a location within the state is selected for 1-m^3 mortar mixing, where the total delivery distance from the raw materials is minimized; and another location is selected to maximize the distance. In both scenarios, the location of one raw material closest to the mixing site is assumed to be the sole supplier for that material in the 1-m^3 mortar batch. The mixing site maximizing the delivery distance implies the site's geographical isolation from the resources, i.e., it is not by deliberately omitting closer resources to forge a maximum delivery distance. Due to the wide distribution of fine aggregate sources (USGS, 2011), the transportation distance of fine aggregate is assumed to be 50 km regardless of the mixing location. The assumptions for transportation modes are consistent with the Berkeley case study.

2.2.3.3. LCIA. Global warming is selected as the impact category of concern – the global warming potential (GWP) is characterized from the LCI and outputted by the tool for every process during mortar production. Other environmental impact indicators have been considered in LCA studies on different cements, e.g., alkali-activated materials. However, for LCA of PC concrete and mortar, most studies restrict their scope to GWP, characterization method of which from LCI results is direct and widely agreed. Therefore, considering that (a) the major well-known environmental issue of PC concrete is high CO_2 emissions, (b) no alkali activators (of much more intense contribution to, e.g., ozone depletion than PC production), and (c) the well-established characterization method of GWP with low uncertainty, GWP is the only utilized indicator in our LCIA.

Furthermore, combined eco-performance indicators are utilized to interpret the LCA results, by normalizing the total energy consumptions or the total GWP of a mix to its strength at multiple ages (Fort et al., 2018). This approach improves the assessment of the green binders' competence in practical construction.

3. Results

3.1. Setting time, consistency, slump and soundness

The w/b ratio for normal consistency increased with an increased DE level (Table 5) due to the high water-demand of

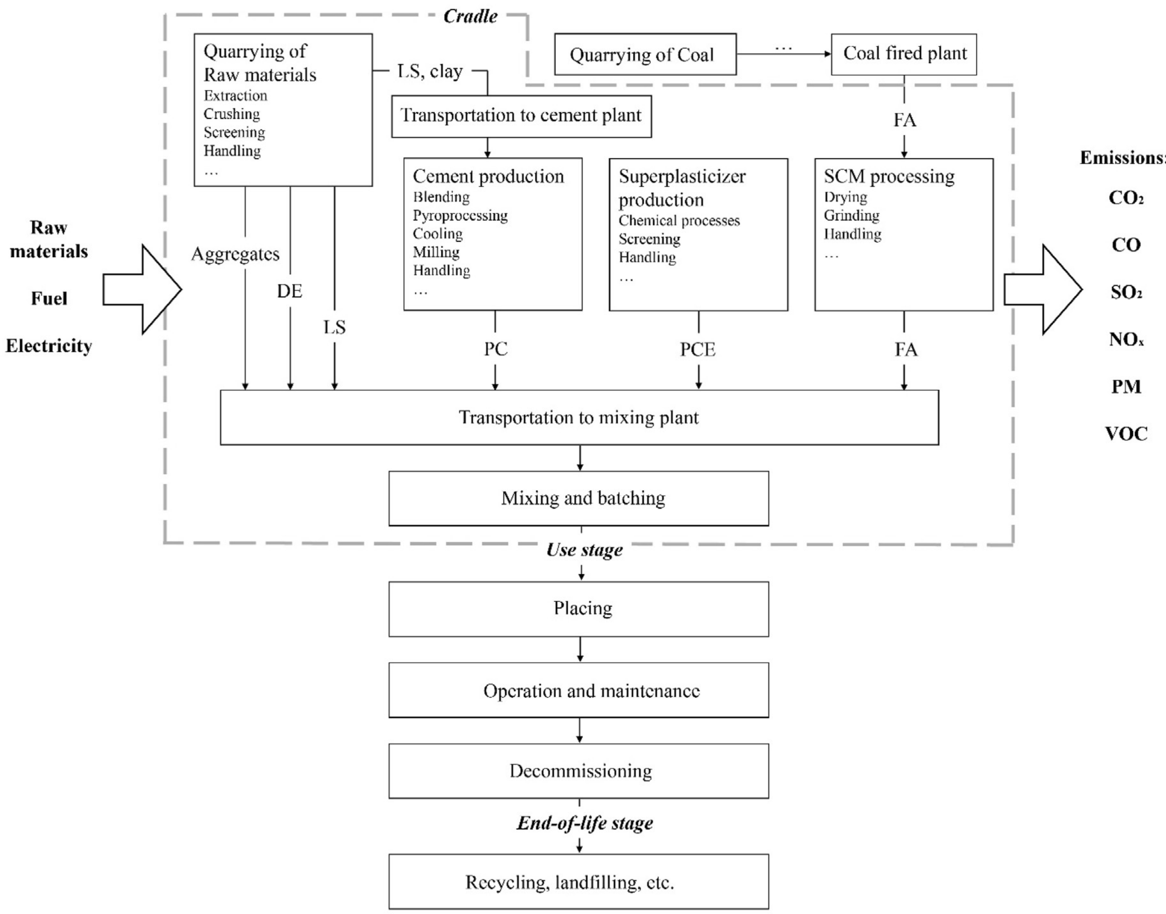

Fig. 1. Process flow of the mortar production system. The scope of this study is in the dash-line box.

Table 3
Assumptions used in LCA calculations.

Processes and material sources	Location	
Mixing plant	Various in western U.S.	
Cement production	35 plants in various locations listed in Supplementary information (SI)	
Fine aggregate processing	The same state/province of mixing plant	
Limestone	Various (see Fig. 2)	
Diatomite	Washington, Oregon, California, etc. (see Fig. 2)	
Class C Fly ash	Texas, Wyoming, North Dakota, etc. (see Fig. 2)	
PCE production	Cleveland, Ohio, U.S.	
Transportation	**Mode**	**Distance (km)**
Raw materials to cement plant	Class 8B truck (0.76 MJ/ton-km)	1
Gypsum to cement plant	Class 8B truck	100 km
Cement to mixing plant	Class 8B truck (and Rail)	Various
FA to mixing plant	Class 8B truck (and Rail)	Various
DE to mixing plant	Class 8B truck (and Rail)	Various
LS to mixing plant	Class 8B truck (and Rail)	Various
PCE to mixing plant	Rail (0.24 MJ/ton-km) and Class 2B truck (3.86 MJ/ton-km)	Various
Fine aggregate to concrete plant	Class 8B truck	50 km
Technology options	**Selected technology**	
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	Preheater	
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	
Batching plant loading/mixing	Mixer loading	
Batching plant PM control	Fabric filter	

Table 4
Source locations, distances to the mixing plant, and transportation methods.

	Source	Distance	Transportation
Portland cement	Cupertino, CA	86 km	Truck 8B
Fly ash	St Johns, AZ	1455 km	1405 km by rail; 50 km by truck 8B
Diatomite	Richmond, CA	20 km	Truck 8B
limestone	Richmond, CA	20 km	Truck 8B
PCE	Cleveland, OH	3950 km	3900 km by rail; 50 km by truck 2B
Fine aggregate	Pleasanton, CA	53 km	Truck 8B

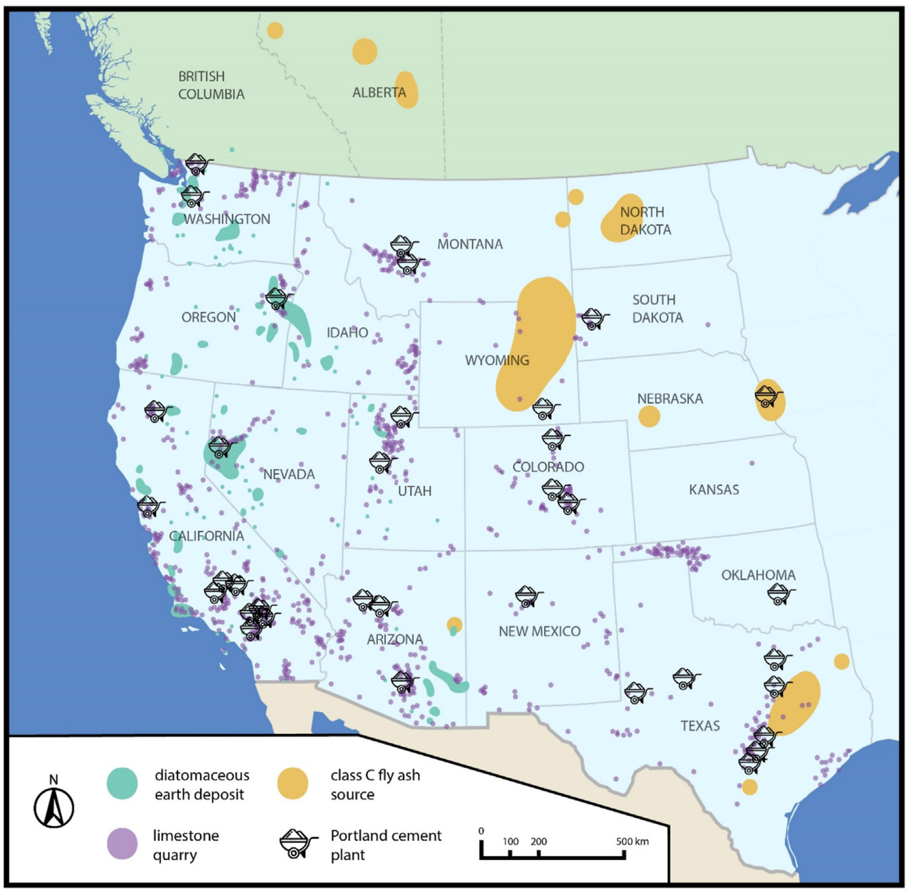

Fig. 2. Resource map of western United States and British Columbia and Alberta. Locations of PC plants are from (CemNet.com, 2018), locations of DE are from (Wallace et al., 2006), locations of limestone are from (USGS, 2011), and the areas with class-C FA production are colored yellow (Hoffman, 2002; Slag Cement Association, 2001; U.S. Energy Information Administration, 2017). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 5
Water-to-binder ratio, setting time, soundness of mixes with normal consistency.

	w/b for normal consistency	Initial setting (min)	Final setting (min)	Final setting − Initial setting (min)	Soundness expansion (mm)
PC	0.29	63	82	19	<1
15FA	0.252	69	92	23	1
30FA	0.191	79	109	30	2
15DE	0.403	67	84	17	<1
30DE	0.553	75	100	25	<1
30DE-15FA	0.497	87	113	26	1
15DE-30FA	0.366	84	119	35	1
30DE-10LS	0.566	78	110	32	<1
30DE-15FA-5LS	0.499	90	122	32	1
30DE-30FA	0.479	115	158	43	1
30DE-25FA-5LS	0.488	105	150	45	1
25DE-30FA-5LS	0.47	110	150	40	1
30DE-20FA-10LS	0.498	106	151	45	1

porous DE (Kocak and Savas, 2016). Water demands of 15 wt% DE and 30 wt% DE substitution were 39% and 91% greater than that of the PC reference, respectively. The w/b for normal consistency decreased with an increased FA level due to the spherical shape of FA particles (Bae et al., 2014). Water demands of 15 wt% FA and 30 wt% FA substitution were 13% and 34%, lower than that of the PC reference, respectively. The additional replacement of PC by FA from PC-DE and PC-DE-LS blends slightly reduced their water demands. Thus, FA has a great potential for reducing the water demand of DE-containing blends.

Mixes with normal consistency containing both DE and FA experienced retarded setting due to the dilution of the pastes. The initial and final setting time of PC-DE and PC-FA systems were both retarded at high replacement levels. The replacement of PC by over 45 wt% of FA and DE significantly deferred the setting by at least 20 min due to the dilution of the hydration system. At replacement level of 60 wt%, the time differential between initial and final setting was extended by ~20 min. The soundness expansion of all pastes was not greater than 2 mm, conforming to the standard.

For pastes with w/b of 0.48, the slump of PC-FA increased with an increased FA level (Table 6). PCE dosage was adjusted by up to 0.9 wt% of binder in DE-containing pastes to fix pastes at slumps of ~110 mm. The PCE dosage increased at increased DE level due to the high water-demand of DE. The addition of FA in DE-containing pastes lowered the water-demand and reduced the PCE dosage. FA retarded the pastes due to dilution effect (De Weerdt et al., 2011) and possibly intermixed anhydride in FA. Although PCE can retard PC hydration (Winnefeld et al., 2007), the fine DE particles accelerated the PC hydration in PC-DE pastes and dominated the competition of hydration at replacement level below 50 wt%. For replacement over 50 wt%, the setting was delayed due to low PC content. The initial setting at replacement level of 60 wt% was retarded by at least 50 min. The final setting of the pastes at high replacement level (above 50 wt%) was extended by 30–100 min, and the time differential between initial and final setting at replacement level of 60 wt% was extended by 30 min. The soundness expansion of all pastes at w/b of 0.48 was not greater than 2 mm, conforming to the standard.

3.2. Compressive strength

For mortar cast and cured at 18 °C, all blended mortar exhibited lower compressive strength at 1 day than the reference due to their low cement content and low hydration degree (Table 7 and Fig. 3a). At 7 days, only DE or FA with replacement level of 15% and 30% exhibited comparable strength to the reference due to the pozzolanicity of DE (Jud Sierra et al., 2010) and FA and self-cementing property of FA while mortars with higher replacement level exhibited 12%–62% lower strength due to low cement content. At 28 days, 15DE and 30DE mortars exhibited comparable strength to the reference due to intense pozzolanicity of DE (Yimaz, 2008), and the 28d-strength of 30DE-10LS was comparable to that of 30DE due to pore-refining of fine LS (Celik et al., 2015). The 28d-strength of 15FA and 30FA was 13%–16% lower than the reference due to the low reactivity of FA (Li et al., 2017). Mortars with replacement level above 45% exhibited 13–60% lower strengths at 28 days than reference due to the low cement content. The strength of 30DE-30FA at 56–90 days was 34% lower than the reference while other quaternary-system mortars with LS also at 60 wt% replacement level exhibited comparable strength to reference at 90 days (±3 MPa). The considerable difference in strength can be explained by the FA-LS reaction and the pore-refining of fine LS (Celik et al., 2015). The strength of PC-DE was higher than PC-FA at 56-90 d at the same replacement level due to the large surface of DE.

For mortars cast and cured at 26 °C and 35 °C (Fig. 3b), the 1d- and 7d-strength were significantly higher than that at 18 °C,

Table 7
Compressive strength of mortar.

	Compressive strength (MPa)				
	1 day	7 days	28 days	56 days	90 days
Cast and cured at 18 °C					
PC	19.6	32.6	53.9	56.5	57.4
15FA	13.8	30.9	45.9	53.1	59.8
30FA	11.2	32.9	47.6	59.8	66.8
15DE	14.4	32	54.8	60.1	64.9
30DE	10.4	33.2	55.6	66	72.7
30DE-15FA	9.2	20.6	40.5	52	59.1
15DE-30FA	8.5	28.1	48	57.9	62.9
30DE-10LS	10	28.9	52.1	62.1	69.8
30DE-15FA-5LS	10.1	23.8	42.9	51.7	60.9
30DE-30FA	4.4	12.3	20.8	30.9	38.2
30DE-25FA-5LS	5.1	18.8	37.7	51.2	59.4
25DE-30FA-5LS	5	19.2	35.6	45.9	53.5
30DE-20FA-10LS	5.9	20.3	38.1	46.1	52
Cast and cured at 26 °C					
PC	23.8	42.9	51.9	53	54.7
30FA	21	41.8	51.1	60	67.1
30DE	21.4	45.6	63.3	67	69.8
30DE-10LS	19.9	48.7	65.2	67.9	71.2
30DE-25FA-5LS	11.1	29.9	43.9	54.8	63.9
25DE-30FA-5LS	12.8	30.2	42.2	53.9	60
Cast and cured at 35 °C					
PC	29.9	47.9	51	51.2	51.9
30FA	24.4	46	58.1	62.6	63.2
30DE	25.9	49.9	64.9	66.4	67.8
30DE-10LS	24.1	52.8	64.1	67	68.3
30DE-25FA-5LS	18.1	31.9	46.8	55.8	63
25DE-30FA-5LS	19.9	33.1	47.4	56.8	64.1

Table 6
Superplasticizer dosage, setting time, slump and soundness of mixtures at w/b = 0.48.

	PCE dose of binder (%)	Initial setting (min)	Final setting (min)	Final setting − Initial setting (min)	Slump (mm)	Soundness expansion (mm)
PC	0	146	169	23	102	1
15FA	0	153	179	26	119	1
30FA	0	161	200	29	142	2
15DE	0.3	129	159	30	105	<1
30DE	0.9	112	143	31	102	<1
30DE-15FA	0.67	133	161	28	110	1
15DE-30FA	0.13	139	169	30	114	1
30DE-10LS	0.87	110	147	37	108	<1
30DE-15FA-5LS	0.6	160	199	39	109	<1
30DE-30FA	0.39	210	273	63	123	1
30DE-25FA-5LS	0.47	201	253	52	118	<1
25DE-30FA-5LS	0.36	204	257	53	125	1
30DE-20FA-10LS	0.51	199	239	40	120	1

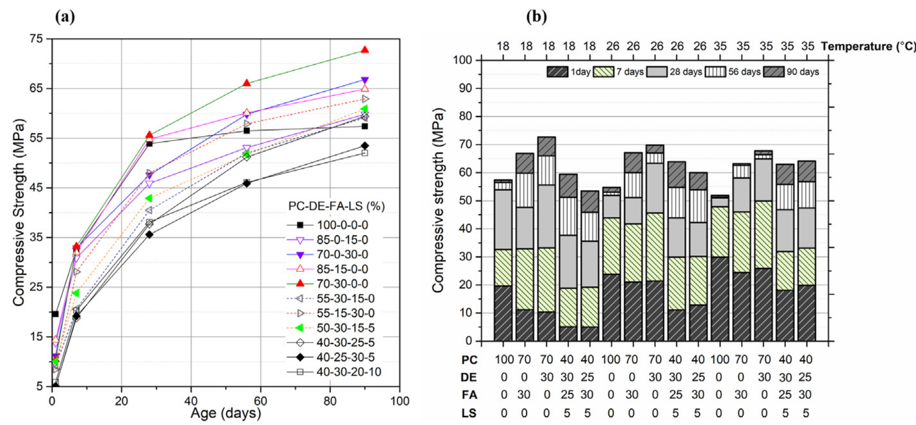

Fig. 3. Compressive strength of mixes cast and cured at (a) 18 °C and (b) various temperatures.

resulted from accelerated hydration at elevated temperature. The strength of PC reference at 26 °C and 35 °C at 28–90 days was 4%–10% lower than that at 18 °C; the slow strength gain can be explained by the intrinsic defects (e.g., micro-crack) during high-temperature casting (Mehta and Monteiro, 2013). The strength at 28–56 days of the blended mortars at 26 °C and 35 °C was higher than that of blended mortars at 18 °C due to accelerated reaction (Rossen et al., 2015). At 90 days, the strength of 30DE-25FA-5LS and 25DE-30FA-5LS at elevated temperatures were higher than that at 18 °C due to accelerated reaction. Thus, casting and curing at high temperature benefits the strength gain of samples with high replacement levels.

3.3. LCA results

3.3.1. Berkeley case study

PC mix results in the highest total energy consumptions of 2836 MJ/m^3 mortar produced (Fig. 4a); the 30DE-20FA-10LS mix, results in the lowest total energy consumptions of 1344 MJ/m^3 (~47% of the maximum). In general, the total energy consumptions monotonically decrease with the increased replacement level. Using DE leads to slightly higher reduction than using FA at the same replacement level. Cement production contributes to most of the total energy consumptions, at maximally 93% in the PC reference and minimally 73% in the 30DE-30FA mix. The reduction in GWP with increased replacement levels has a similar pattern (Fig. 4b). From 100 wt% to 40 wt% PC, the total GWP are decreased by 58% in 30DE-30FA while the contribution from cement production drops moderately from 96% to 88%. The slighter decrease is explained that the SCMs contribute less to total GWP than to total energy consumptions since cement pyroprocessing dominates CO_2 emissions.

Energy intensity and GWP intensity are useful eco-performance indicators evaluating both the environmental impacts and the mechanical properties (Fort et al. 2018). As the replacement level increases from 0 to 40%, the 28d energy intensity decreases considerably (Fig. 5). At replacement level of 40%–60%, the 28d

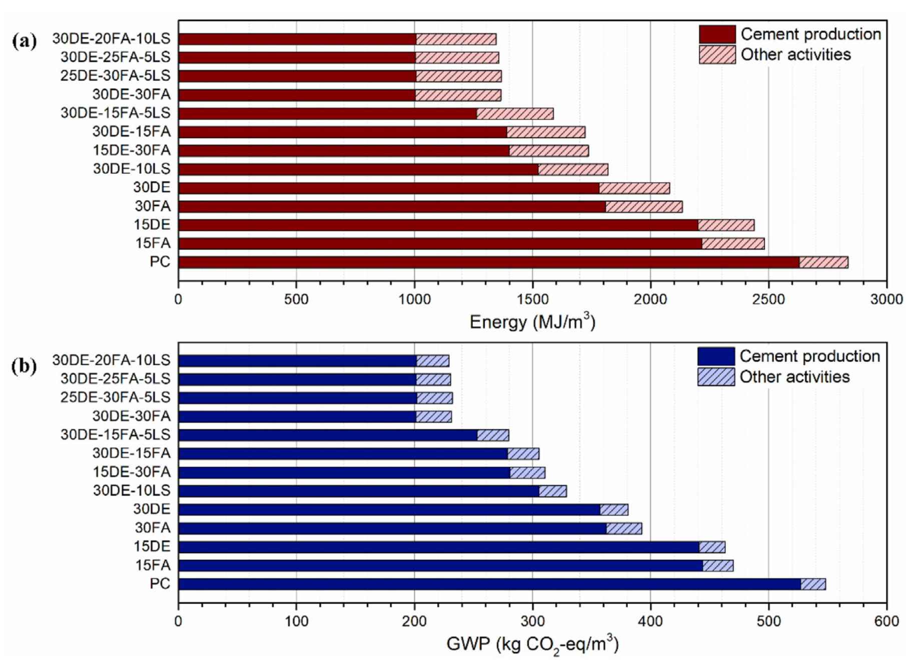

Fig. 4. (a) Energy consumptions and (b) GWP due to cement production and other activities (including transportation, processing/production of other raw materials, and mortar mixing and batching) in the Berkeley-based scenario.

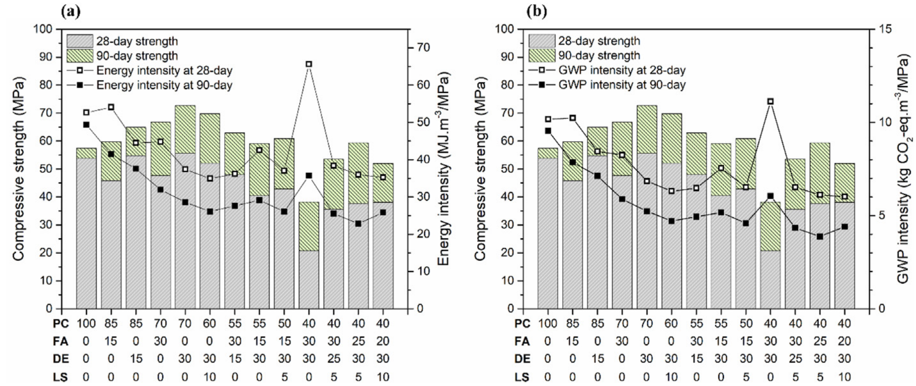

Fig. 5. 28-day and 90-day compressive strength of the mortar mixes and their corresponding (a) energy and (b) GWP intensity normalized to 28-day and 90-day strength in the scenario of Berkeley, CA.

energy intensity mostly plateaus at ~38 MJ m^{-3}/MPa (~70% of that of the PC mix), with an exceptional fluctuation at the 30DE-30FA due to its slow strength development. As a result of higher long-term strength gains of the blended mixes compared to pure-PC mix, the 90d energy intensity stabilizes to ~25 MJ m^{-3}/MPa at high replacement levels (~60% of that of PC mix). Besides, the undesired high energy intensity of the mix 30DE-30FA is ameliorated at 90 days owing to its long-term strength gains.

The GWP intensity generally follows the same trend as the energy intensity at both ages (Fig. 5b). As the replacement level increases to 60%, the maximum decrease in GWP intensity is achieved by the mix 30DE-20FA-10LS at 28 days as 6.0 kg m^{-3}/MPa (~60% of the PC mix) and by the mix 30DE-25FA-5LS at 90 days as 3.9 kg m^{-3}/MPa (~40% of the PC mix). It is noteworthy that the mix 30DE-25FA-5LS gains greater strength at 90-day than the PC mix. Therefore, 30DE-25FA-5LS effectively enhances both mechanical and environmental benefits.

Apart from cement production, the major contributors to energy consumptions and GWP during mortar production are mixing and batching, transportation from material sources to processing plants, and the processing/production of raw materials (Fig. 6). The energy consumptions of mixing and batching and fine aggregate processing are independent of mix proportions, each amounting to ~53 MJ/m^3 (~2–4% of total energy consumptions). Due to the shorter transportation distances of LS and DE and the much longer distance of FA compared to that of PC, the energy consumptions from transportation activities are minimized in the mix 30DE-10LS at 88.5 MJ/m^3 and maximized in 30FA-15DE at 139.5 MJ/m^3; in all mixes, transportation contributes to ~3.5–9.9% of the total energy consumptions. At the highest replacement level, FA processing contributes ~5% of total energy consumptions, DE processing contributes ~1% of the total amount, and LS processing leads to <0.4% contribution. SCMs in all mix contribute to <7% of total energy consumptions. Due to the energy-intensive nature of PCE production, PCE production in 30DE is the second most energy-consuming process after cement production.

Transportation is the second dominant contributor to GWP after cement production (Fig. 6b), varying between ~62 and 78% of GWP from processes other than cement production. During processes other than cement production, transportation's share in GWP is higher than its share in energy consumptions because transportation is more GWP -intensive than energy-intensive. Similar to energy consumptions, the GWP from mixing and batching and aggregates processing is independent of mix proportions. The contributions of all SCMs in each mix to the total GWP are <2%. While PCE production is ~4 times as energy-consuming as cement production, it yields 20% GWP compared to an equal amount of cement produced. Thus, PCE shows less remarkable share in total GWP, e.g., ~1.1% of total GWP in 30DE-10LS.

At increased replacement levels, the emissions of air pollutants decrease (Table 8). The greatest reduction occurs in 30DE-20FA-10LS (60% replacement level) with ~30% reduction in CO emissions, ~49% reduction in NOx emissions, ~57% reduction in SO_2 emissions, and ~23% reduction in PM10 emissions.

CO emissions from transportation and cement production are comparably dominant as a common result of incomplete combustion of fuels (Fig. 7). At higher replacement levels, especially at higher usage of distantly-sourced FA, the contribution from

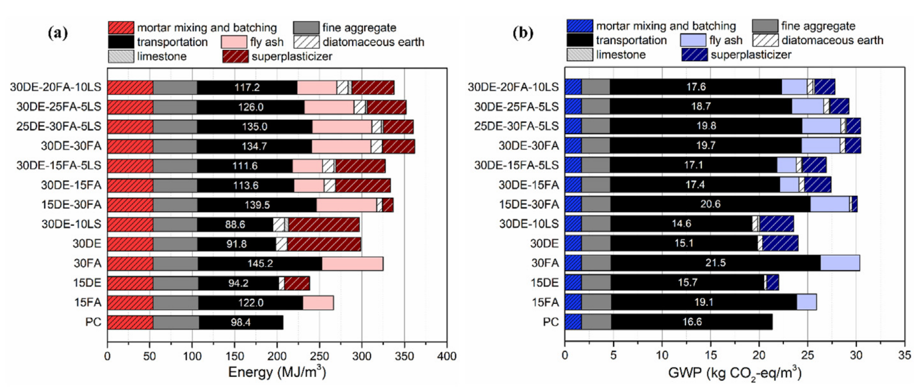

Fig. 6. Distribution of (a) energy consumptions and (b) GWP due to major processes during mortar production other than cement production.

Table 8
Emission rates (kg/m^3 mortar produced) of air pollutants from mortar production.

	CO	NOx	SO_2	PM10	VOC[a]
PC	0.292	1.551	1.062	0.0776	0.0635
15FA	0.292	1.395	0.919	0.0776	0.0540
15DE	0.258	1.329	0.899	0.0704	0.0537
30FA	0.292	1.242	0.779	0.0776	0.0447
30DE	0.228	1.119	0.745	0.0634	0.0446
30DE-10LS	0.207	0.983	0.643	0.0594	0.0386
15DE-30FA	0.258	1.027	0.619	0.0705	0.0351
30DE-15FA	0.227	0.969	0.606	0.0637	0.0353
30DE-15FA-5LS	0.216	0.901	0.554	0.0615	0.0322
30DE-30FA	0.225	0.820	0.467	0.0636	0.0260
25DE-30FA-5LS	0.226	0.822	0.468	0.0638	0.0261
30DE-25FA-5LS	0.215	0.802	0.463	0.0616	0.0261
30DE-20FA-10LS	0.205	0.784	0.457	0.0594	0.0262

[a] Due to lack of data, the VOC emissions from transportation is not included.

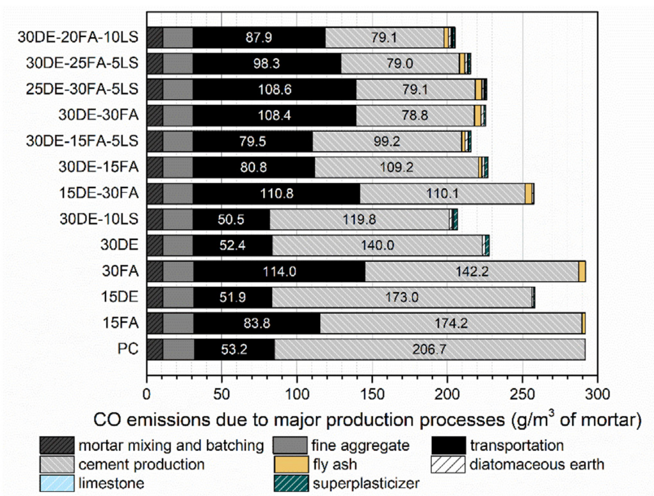

Fig. 7. Distribution of CO emissions by major processes in mortar production.

transportation to CO emissions exceeds that from cement production because fuel burning during transportation has a relatively high rate of CO emissions. In general, cement production is responsible for 35–71% of total CO emissions during mortar production, transportation for 18–48%, aggregate production for 7–10%, mixing and batching for 3.7–5.2%, while the processing/production of SCMs and PCE altogether accounts for <3.5% of total CO emissions.

Cement production and transportation are the first (~61–85%) and the second (~15–37%) largest source of NOx emissions during mortar production due to the formation of thermal NOx and fuel NOx in high-temperature pyroprocessing kilns and diesel engines (Fig. 8a). Mixing and batching, processing of aggregate and SCMs, and PCE production have minimal contributions due to the low NOx emissions rate from California's electricity generation. Like NOx, the main source of SO_2 is the combustion fuel. Differently, the automobile fuel has a significantly suppressed content of volatile sulfur. Thus, almost all SO_2 emissions come from cement production, and transportation has a much smaller share while other processes are insignificantly contributors (Fig. 8b). VOC emissions display a pattern similar to SO_2. From major processes in mortar production except transportation (Fig. 8c), cement production dominates the total VOC emissions due to the escaping of volatile impurities during pyroprocessing. Other activities result in negligible VOC emissions.

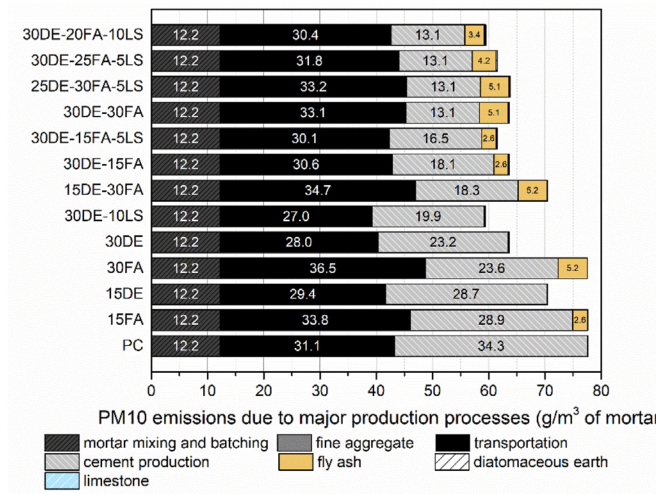

Fig. 9. Distribution of PM10 emissions by major processes in mortar production.

As both a primary pollutant created directly during processing and a secondary pollutant formed from small airborne particles, PM10 is emitted among the major processes of mortar production (Fig. 9). Transportation is responsible for ~40–52% of total PM10 emissions during mortar production, cement production for ~21–44%, mixing and batching for ~16–21%, and FA processing for 3.4–8.0%, while the processing of aggregate, DE, and LS for <0.6%.

3.3.2. Mixing scenarios in 11 states in the western U.S

In each state, the location of mixing plant is varied so that the

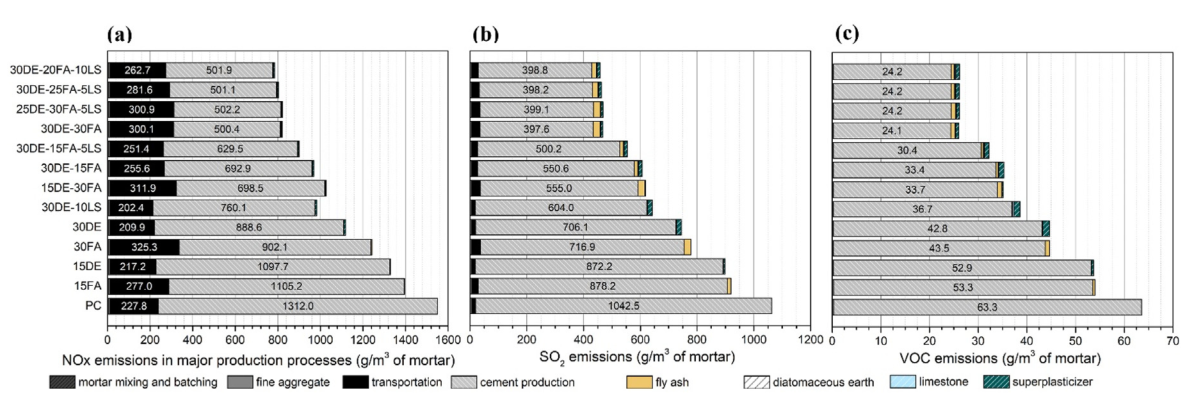

Fig. 8. Distribution of (a) NOx, (b) SO_2, (c) VOC emissions by major processes in mortar production. Note: VOC emissions from transportation is not included.

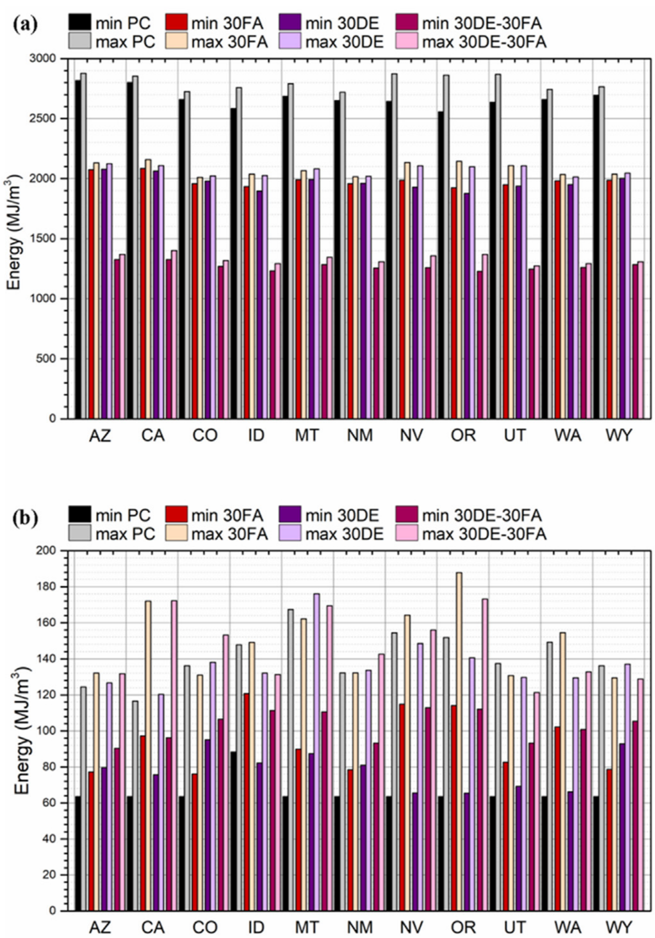

Fig. 10. (a) Total energy consumptions and (b) energy consumptions due to transportation during the production of mortar of the mix PC, 30FA, 30DE, and 30DE-30FA for the scenarios of minimum and maximum delivery distances in each state. The eight columns ascribed to each state, from the left to the right, are for minimum- and maximum-distance PC mix, minimum- and maximum-distance 30FA mix, minimum- and maximum-distance 30DE mix, and minimum- and maximum-distance 30DE-30FA mix.

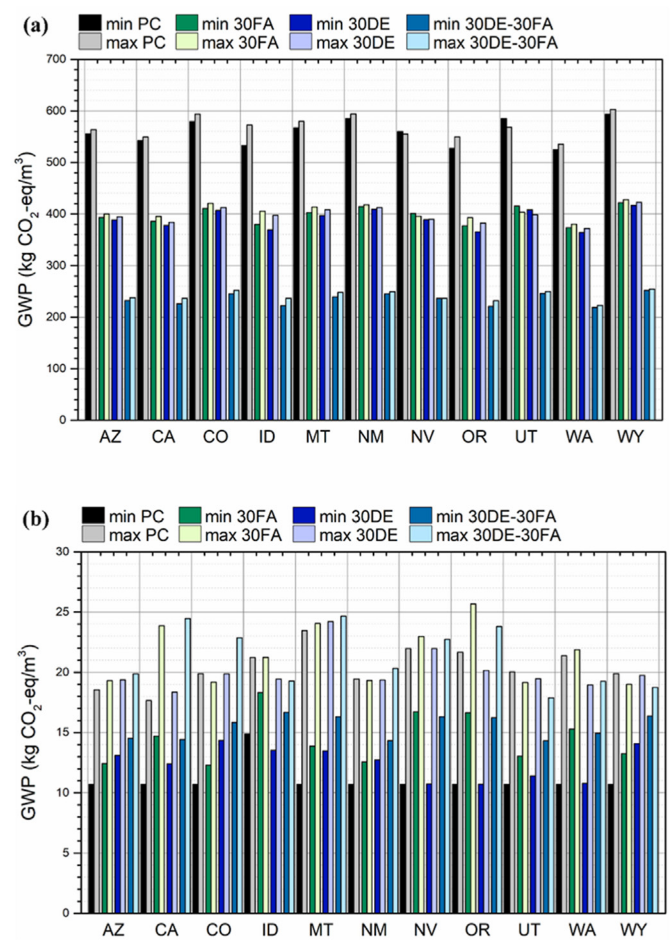

Fig. 11. (a) Total GWP and (b) GWP due to transportation during the production of 1-m^3 mortar of the mix PC, 30FA, 30DE, and 30DE-30FA for the scenarios of minimum and maximum delivery distances in each state of concern. The eight columns ascribed to each state, from the left to the right, are for minimum- and maximum-distance PC mix, minimum- and maximum-distance 30FA mix, minimum- and maximum-distance 30DE mix, and minimum- and maximum-distance 30DE-30FA mix.

total transportation distance from the closest raw materials sources is minimized/maximized. Energy consumptions (Table S2) and GWP (Table S3) are calculated for each case with state-specific electricity grids. Results from PC, 30FA, 30DE, and 30DE-30FA are selected to demonstrate the effect of high-replacement level (the influence of LS is not discussed due to its wide existence and typical vicinity to cement plants).

In all cases, 30% and 60% replacement levels result in respectively ~26% and ~52% reduction in total energy consumptions (Fig. 10a). This consistency substantiates that replacement with SCMs effectively mitigates the environmental impacts from cement industry. Nevada, Oregon, and Utah show the greatest energy consumptions differences between the minimum- and maximum-distance case of each mix, due to the uneven resource distribution around the states and the significant difference between the adjacent states' electricity grid. In contrast, for Arizona, New Mexico, and Wyoming, the small energy consumptions difference between the minimum- and maximum-distance case is mostly attributable to differential delivery distances. In any case, this energy consumptions difference between the minimum- and maximum-distance case of a mix is ~1.8−10.7% of the total value.

In minimum-distance cases, energy consumptions from transportation minimize at the PC mix, increase in various extent at the 30FA and 30DE mixes, and culminate at the 30DE-30FA mix due to the higher local availability of PC than SCMs (Fig. 10b). The energy consumptions from transportation in a maximum-distance case range ~1.2−2 times of that in the corresponding minimum-distance case. Generally, for both the minimum- and maximum-distance cases: the westmost states (California, Oregon) have higher energy consumptions from transportation in 30FA and 30FA-30DE than other states due to their longer distance to FA sources. For the maximum-distance cases: Arizona has low energy consumptions from transportation for all four mixes, as the only state with local PC, DE, and FA sources; in contrast, Montana has high values due to the sparse resource distribution. In the extreme, mixing 30FA-30DE at the upper coastline in Oregon results in the highest possible contribution (12.6%) to the total energy consumptions from transportation.

The trends for GWP are similar to those for energy consumptions. FA and/or DE effectively reduces the total GWP by ~29% at 30% replacement level and by ~58% at 60% replacement level (Fig. 11a). Note that in Nevada and Utah, the total GWP for the minimum-distance cases exceed the total GWP for the maximum-distance cases. This singularity is explained that while local cement sources are used in the minimum-distance cases, cement sources

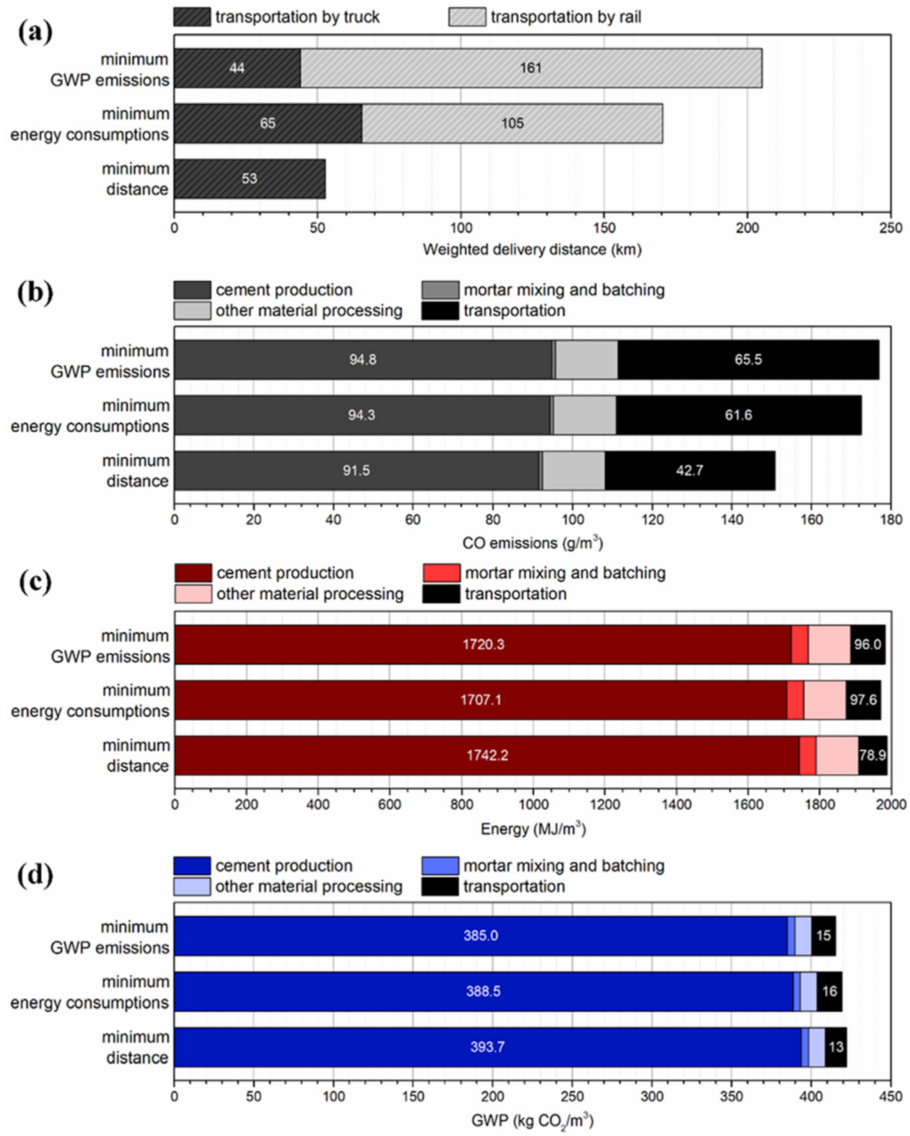

Fig. 12. (a) Weighted delivery distances, (b) CO emissions, (c) energy consumptions, and (d) GWP of the minimum-distance case, the minimum-energy consumptions case, and the minimum-GWP case based in Wyoming.

from neighboring states (California and Arizona, respectively) with greener (i.e., of lower GWP-rate) electricity grid are used in the maximum-distance cases. When the supply of raw materials is optimized, transportation distances hardly govern the total environmental impacts of a mix but the "greenness" of the electricity grid in the cement production location matters. Trends of transportation's contribution to the energy consumptions apply well to that to GWP (Fig. 11b).

Comparison between the three "minimums". The minimum-distance case does not match the minimum-energy consumptions or the minimum-GWP case when the greener electricity grid in the adjacent state more than compensates for the impacts from cross-state transportation. Transporting cement from Oregon (of extraordinarily green fuel mix) to southern California (~1600 km) and mixing in California results in comparable total energy consumptions as mixing right next to a local cement plant in California. Nevertheless, typically the rates of energy consumptions and GWP in neighboring states' electricity grid do not differ by significant amounts.

A closer examination of mixing 30FA in Wyoming allows better understanding of the subtle difference between the three minimums. The weighted delivery distances (Fig. 12a) of the three cases are calculated as 70% delivery distance of PC plus 30% delivery distance of FA. The total CO emissions in the three cases differ by ~15%, mostly attributable to delivery distance difference (Fig. 12b). The minimum-distance case has the least CO-emissions because diesel combustion during transportation is CO emission-intensive. Similar trends are found for NOx and PM10 emissions, also significantly contributed by transportation.

The electricity grid in Wyoming (cement source in the minimum-distance case) is less green than that in Utah (cement source in the minimum-energy consumptions case) and Colorado (cement source in the minimum-GWP case), owing to the heavy use of coal in Wyoming. Consequently, the minimum-distance case has total energy consumptions 0.8% higher than the minimum-energy case and total GWP 1.7% higher than the minimum-GWP case (Fig. 12c and d) since the influence of the cement production dominates. That the minimum-distance case has the least in air

Table 9
Electricity grid in the selected states (Gursel, 2014).

	Arizona	Oregon	Wyoming
Coal	35%	6%	91%
Natural gas	31%	28%	1%
Nuclear	27%	–	–
Hydro	6%	58%	2%
Biomass	–	1%	–
Wind	–	6%	5%

pollutant emissions and almost the least energy consumptions and GWP validates the use of minimum-distance case in the study.

Comparison between different electricity grid. The minimum-distance cases of mixing 30DE-30FA in Arizona (close to FA and DE), Oregon (close to DE and remote to FA), and Wyoming (close to FA and remote to DE) demonstrate the primary influence of the electricity grid at the PC source on the total energy consumption and GWP. The three states have internal PC production with the respective electricity grid (Table 9) (Gursel, 2014).

Despite the local availability of both SCMs in Arizona, it has the highest total energy consumptions during the mortar production. While it saves 21.7 and 15 MJ/m^3 energy from transportation compared to Oregon and Wyoming, its energy consumptions for cement production are 87.8 and 41.3 MJ/m^3 higher, respectively (Fig. 13a). This fact is explained by their different electricity grids: the lifecycle energy use for electricity generation by nuclear power is ~6% higher than by coal, 34% higher than by natural gas, and 91.5 times higher than by hydropower (Gursel, 2014). Moreover, 1-kg PC production demands ~ 5 MJ energy, which allows 1-kg PC delivered by class 8B truck for thousands of kilometers. Thus, the local power grid, rather than transportation, governs the total energy consumptions from mortar production.

Differently, the GWP from mortar production in Arizona are lower than that in Wyoming (Fig. 13b), explained by the differential relative embedded energy use and GWP rates in each fuel type. Despite of the higher embedded energy, nuclear power has low lifecycle GWP, ~81% of that for hydropower and ~2.4% and 2.6% of that for coal and natural gas, respectively (Gursel, 2014). Therefore, even though electricity from nuclear power results in high energy consumptions in Arizona, it mitigates the GWP impacts. Oregon's dependence on renewable sources ensures both low energy consumptions and GWP. Admitted that transportation distance is concerned for monetary and labor costs and air pollutant emissions, it is not the primary determinant in the energy consumptions and GWP during mortar production.

4. Discussion and limitations

4.1. Environmental considerations

PC-DE-FA-LS mixes with replacement level of 60% can exhibit comparable strength to the reference at 90d and even higher strength at elevated temperature. However, the high-temperature condition could depend on curing with heating accommodations, in the LCA study of which, the additional energy consumptions and consequent emissions should be included. The combined eco-performance indicators (i.e., energy intensity and GWP intensity) are suggested as appropriate indices to evaluate the effectiveness of heat curing.

The high clay content in the raw DE is a reason for the high PCE demand of DE-containing mixes, owing to the PCE-clay adsorption (Ng and Plank, 2012). Therefore, calcination of DE is a potential solution to the problematic workability of high-DE mixes. A tradeoff between degree of DE calcination and the dosage of PCE could be discovered to optimize the environmental benefits of incorporating DE so that the total energy consumptions and emissions from DE calcination and PCE production is minimized. In addition, the silica content in DE increased from 88% to 97% after 5-h calcination at 400 °C, positively influencing the strength gain of the DE-containing sample (Bagci et al., 2017). Therefore, the effect of calcination can be similarly assessed with the eco-performance indicators as that of heat curing.

From the previous study (Li et al., 2019), long-distance transportation would overshadow the reduced energy consumptions

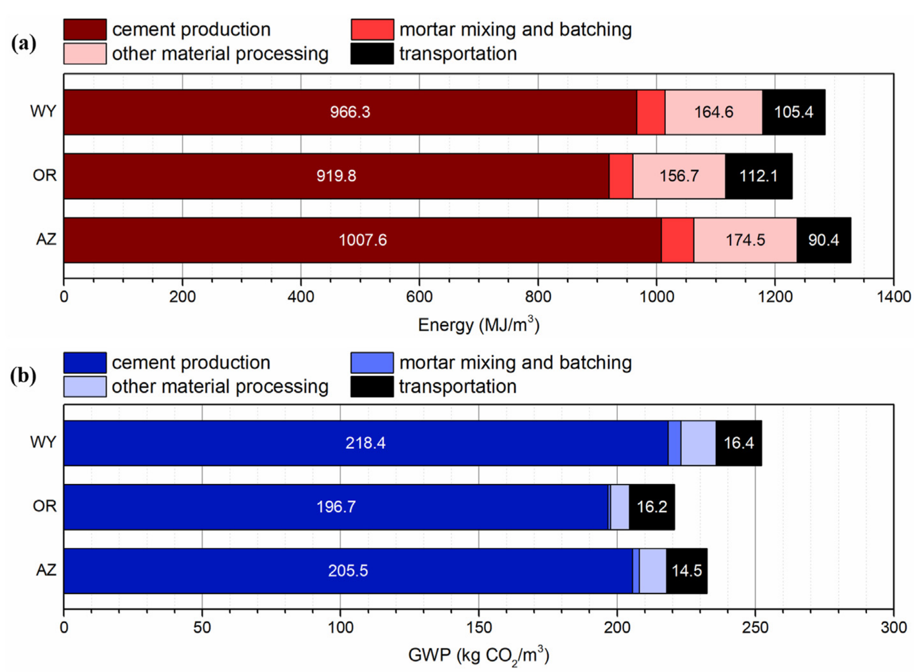

Fig. 13. Distribution of (a) energy consumptions and (b) GWP of the minimum-distance 30FA-30DE mix produced in the selected states (Arizona, Oregon, and Wyoming).

and emissions from using SCMs if one preordained resource location was used for far-apart mixing sites. Now with optimized material sources, for a mix, the energy consumptions/GWP difference between the minimum- and maximum-distance cases based in the same state is usually smaller than the difference between states, manifesting the importance of the greenness of the electricity grid in the state of cement production over transportation. With ideal transportation modes, the electricity grid of material production dominates the overall greenness of a binder.

A more careful examination should not overlook the practicability of exploiting a raw DE deposit (e.g., some DE deposits require inconvenient transportation) and the reactivity of DE from various sources. Mortars at 40% replacement of PC with DE can maintain most strength (Ahmadi et al., 2018) while alkali-activated DE can exhibit substandard strength due to the high impurity of DE (Sinsiri et al., 2012) and due to low alkalinity of the activator (Arbi et al., 2013). Thus, different DE likely results in different values of the combined eco-performance indicators due to their purity and reactivity. Another direction is to include spent DE (byproduct from other industries), the embedded environmental impacts of which should be reassessed due to different pre-processing and the change in its footprint history. Spent DE for filtering in brewery was calcined at 400 °C to remove the organic yeast and then dissolved in sodium hydroxide solution to produce an alkali-activator as an alternative to sodium silicate (Mejía et al., 2016). The need for calcination is left for discussion. Nevertheless, the disadvantaging additional environmental impacts from calcination may be balanced by the advantaging enhanced reactivity.

The current LCIA is limited to the most used category of global warming with GWP as the indicator. For a bigger picture of the environmental impacts, ozone depletion (ODP), acidification potential (AP), eutrophication potential (EP), abiotic depletion for fossil fuels (ADPF), and photochemical oxidants formation (POF) are other frequently used environmental indicators in the LCIA of blended cement (Moretti and Caro, 2017), alkali-activated concrete (Salas et al., 2018), and SCMs-containing concrete (Teixeira et al., 2016). From these studies, blended cement with either LS, BFS, or silica fume and concrete using coal/biomass FA have reduced environmental impacts in each category compared to pure PC references; water-glass activated natural zeolite concrete has greatly reduced GWP, comparable AP, EP, ADPF, and POF, and significantly increased ODP compared to the plain concrete of comparable strength, due to the chemical emissions-intensive chlor-alkali process in NaOH production. Given no need for alkali-activator and the use of FA and LS, our green binder PC-DE-FA-LS is expected to, overall, have lower-than-reference environmental impacts since DE is not expected to generate significant impacts from its little processing required. The only uncertainty lies in PCE − future study should more comprehensively examine its environmental impacts.

Striving for reduced emissions over time, the state of California has enacted the bill of Buy Clean California Act, which requires the maximum acceptable GWP for multiple categories of construction materials to be established and published, having started in 2019 and renewing after every 3 years (California Legislative Information, 2017a). The bill requires a successful bidder to submit the Environmental Product Declaration demonstrating that any eligible materials involved in the project comply to the determined GWP maximums, where the eligible materials regulated in the bill include carbon steel rebar, flat glass, mineral wool board insulation, and structural steel, but not cement-based materials (California Legislative Information, 2017b). The exclusion of cement-based materials in the first three-year of the Act may be related to potentially significant increase in concrete cost or the great variability in cement compositions, which creates great resistance or difficulties in effective implementation of the Act. Nevertheless, once the Act extends to the concrete industry in future cycles, with California's annual cement consumption of 10.7 Mt, (Portland Cement Association, 2019), great incentive in using the green binders will rise to relieve concrete industry from the high emissions from PC production.

4.2. International situation and perspective of previous studies

The present study has shown the great significance of the influence of transportation and various regional electricity grid on the overall environmental impacts of producing PC-DE-FA blends. In (Ahmadi et al., 2018), Locations of cement plant and mines being unknown, the contributions of transportation and different electricity grids of plants and mines to the environmental impacts of the production of DE-containing mortar were not considered. These contributions should not have been omitted in the previous study because 1) the reserve of diatomite in Iran is negligible (Iran's share is below 0.5% of the global reserve (China Geological Survey, 2017)); 2) the distance between Iran and the nearest large diatomite mine (Anatolia, Turkey) is at least 1700 km; 3) the environmental outputs from electricity grids of Turkey and Iran are distinct—Turkey's electricity grid contains 37% coal (Republic of Turkey Ministry of Energy and Natural Resources, 2016) while Iran does not use coal for electricity generation (Bekhrad et al., 2020). If these contributions have been considered in the calculation of life-cycle assessment of the production DE-containing mortars in (Ahmadi et al., 2018), the environmental benefits of cement replacement with DE would be much lower (e.g., relative high CO and PM10 emissions).

Although the present study focuses on solving the local shortage of SCMs in western U.S. by using FA and/or DE, the partial replacement of cement with DE and FA is as valid in international situations. After western U.S., Czechia and Denmark both account for 15% DE production, China with 14%, Argentina with 7%, and Peru with 4% (USGS, 2019). Other countries (e.g., Turkey, France, Spain, Japan, Korea, and Mexico) also produce fair quantities of DE. Thus, the replacement of cement with DE can be considered in western Latin America, east Asia, and the majority of Europe. For the replacement of cement with DE in regions (e.g., central Asia, east Latin America, and Africa) distant from large DE reserves, the decision makers should not underrate the environmental impact (e.g., energy consumptions and CO emission) from long distance of DE transportation.

So far, there is no enforced policy about the use of diatomite. Department of labor in the United States has determined raw DE with less than 1% crystalline silica as non-carcinogenic (US Department of Labor, 1999). Although DE used in this study is crystalline silica-free, the DE mined from other sources in some existing literatures contains a non-negligible amount of crystalline silica (Degirmenci and Yilmaz, 2009; Papadakis and Tsimas, 2002). Meanwhile, amorphous silica, the major phase in DE, may only causes a mild irritant of the upper respiratory tract, eyes, and the skin because of its drying properties. Chronic health effects have rarely been reported for amorphous silica (EN481, 1993). Therefore, regulations shall be established regarding the handling of raw DE in considerations of safety and health of labors.

A few laws and regulations on fly ash have been applied due to its potential toxicity (i.e., heavy metal ions). Improper handling of fly ash can be harmful to water (surface and ground) and air. The regulations established by the environmental protection agency in different countries show certain variations in rules, but the required parameters for leaching test of coal fly ash are similar in U.S., Australia, and Japan. For example, the leaching limits for As, Ba, Cd, Hg, and Pb ions are strictly applied (Cho et al., 2019). However, for instance, the leachate standard in South Korean does not define the limit for Ba, and the limit for other elements in South Korea is significantly higher than in the other countries. FA not complying to the specific regulations should not be used in the production of PC-DE-FA mixes in the respective country. Especially, the use of FA certified in South Korea should be carefully inspected because the toxicity limits in South Korea are less strict than in other countries.

The present study has demonstrated the intense water/PCE demand of DE which can be mitigated with additional replacement of PC with FA. However, if FA with excessive unburnt carbon is used, the unburnt carbon can absorb the superplasticizer and increase net water/PCE demand of the blended mixes. ASTM C618 sets the upper limit of unburnt carbon content in FA to be 6%. But given the above concern, for PC-FA-DE mixes, the limit of unburnt carbon content in FA should be more rigorous. Otherwise, either the PC-FA-DE mixes would be poorly workable without sufficient amount of superplasticizer, or the mixes would be highly viscous with overdosage of superplasticizer. Furthermore, recalling that the clay mineral intermixed in diatomite absorbs PCE as well (Ng and Plank, 2012), not only should the standard on unburnt carbon content in FA used for PC-FA-DE mixes be more stringent, the standard on clay content in DE used in cement-based materials should also be more rigorous.

As a future direction, the durability of the green binders will be examined, which may shed light on their cradle-to-grave LCA study. Promising long-term properties of the green binder could further enhance its environmental-impact reduction, due to the decreased energy and material inputs in maintenance and extended service lives. From the economic aspect, the trade-offs between cost and environmental impacts of the different mixes need to be considered.

5. Conclusions

This paper aims to study the early properties, mechanical properties, and cradle-to-gate LCA of green mortar mixes with high-volume DE, FA, and/or LS for mitigating the local shortage of SCMs in western U.S. The work has great implication in the use of DE into cementitious materials in DE-rich regions (e.g., western U.S.) for reducing the environmental impacts of the materials production. The important points are:

1. The intermix of FA with DE considerably decreases the high water- and PCE-demand of DE, therefore ensuring good workability at 30% replacement level with DE in mixes of maximum total replacement level of 60%.
2. Despite lower early strength development of the green mixes, mixes at 40–60% replacement levels can obtain compressive strength of ~60–70 MPa at 90 days, higher than the reference, due to high pozzolanic reactivity of FA and DE.
3. In Berkeley-based LCA study, the total energy consumptions, GWP, and the air pollutant emissions, majorly contributed by cement production and transportation, significantly decreased through the replacement with DE, FA, and LS. The 90-day energy intensity and GWP intensity are maximumly reduced by ~60% and ~40%, respectively, in the mix 30DE-25FA-5LS compared to the reference.
4. From the LCA study of mixing in 11 states for regional variability, with source locations optimized, 30% and 60% replacement levels reduced the total energy consumptions by ~26% and ~52% and GWP by ~29% and ~58%, respectively, regardless of the mixing location. In both the minimum- and maximum-distance scenarios, transportation accounts for at most ~10% of total energy consumptions and GWP. The greenness of local cement production, determined by the electricity fuel mix, matters the most in controlling the greenness of a specific binder.
5. For future study, A) raw DE from different sources and spent DE from different industries should be compared to qualify the efficacy of DE, B) other impact categories (e.g., acidification, eutrophication, ozone depletion) should be considered to achieve a more comprehensive LCIA of the green binder, C) durability properties should be examined to elucidate the cradle-to-grave LCA approach, and D) economic dimension – the trade-offs between cost and environmental impacts of different mixes.

Author contributions

Jiaqi Li: Conceptualization, Methodology, Investigation, Data Curation, and Writing - Review & Editing.

Wenxin Zhang: Methodology, Investigation, and Writing - Original Draft.

Chen Li: Conceptualization, Methodology, and Investigation.

Paulo J M Monteiro: Conceptualization, Project administration, and Funding acquisition.

Declaration of competing interest

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

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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.2020.121224.

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