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Environmental life-cycle assessment of concrete produced in the United States

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ABSTRACT

Concrete is a primary material in infrastructure projects and is a significant contributor to global climate emissions. However, there is a lack of readily available cement and ready-mix concrete inventory data for evaluating the environmental performance of the industries. This study describes the development of cradle-to-gate inventories for U.S. ready-mix concrete and gate-to-gate inventories for portland cement production technologies. These life-cycle inventories provide baselines for the environmental releases associated with concrete that is used for major infrastructure projects. The inventories are incorporated into the publicly available Greenhouse gases, Regulated Emissions, and Energy use in Technologies (GREET) model. The life-cycle inventories are created using facility-level environmental release data from U.S. datasets normalized to activity levels which are based on production capacity and utilization data provided by Portland Cement Association (PCA) and the U.S. Geological Survey Minerals Yearbook. Unit processes for limestone quarrying, sand and gravel quarrying, and wet-mix concrete batch plants are developed on the basis of national total point-source environmental releases and production statistics, coupled with corresponding flows associated with off-road fuel consumption and other non-point-source emissions. Midpoint impact assessment results are normalized to provide insight into their relative significance in the context of U.S. total impacts. These findings show that advanced calcination technologies help reduce greenhouse gas emissions, but the full set of releases also highlights the significance of metal releases and particulate-matter emissions generated by non-combustion-related activity.

1. Introduction

Concrete is a primary structural material central to the construction projects throughout the world. It is the second most-consumed material in the world, after water (Miller et al., 2018b). Cement is the binder that holds concrete aggregates together, enabling structural performance. Global cement production of about 4.1 billion tonnes year after year since 2013 (IEA, 2021). Of this, 102 million tonnes, or 2.5%, were consumed in the U.S. Concrete can be transported, pumped, and poured to fill frames in a wide range of forms, and once cured, it is rigid and durable, making it ideal for structural elements, especially distributed transportation infrastructure. As a result, concrete is used in a wide range of construction projects. The Portland Cement Association (PCA) reports that in 2015, 30% of the cement use in the U.S. was for transportation infrastructure, 28% for residential buildings, 18% for nonresidential buildings, 13% for public utilities and water/wastewater systems, and 10% for other applications (PCA, 2016).

The cement industry is a significant source of greenhouse gas (GHG) emissions (Bernstein et al., 2007), with the 2019 U.S. production of 88.5 million tonnes of cement (USGS, 2020) contributing approximately 69 million tonnes CO_2-eq. (USGS, 2020; U.S. EPA, 2019a). These GHG emissions are primarily from the production of portland cement, which uses significant amounts of fuel and releases CO_2 from limestone during calcination. Calcination alone contributed an estimated 41 million tonnes of CO_2-eq. in 2019, which was the second highest contribution of non-fuel-based emissions from the U.S. industrial sector (U.S. EPA, 2020b).

Given concrete's importance in the construction and transportation sectors, environmental assessments of concrete production systems are critical for benchmarking environmental impacts and measuring the

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effectiveness of mitigation measures. Previous environmental assessments of concrete production have found that significant energy and GHG reductions can be achieved by shifting from a wet-kiln system to more advanced dry-kiln systems (Benhelal et al., 2013; Galvez-Martos and Schoenberger, 2014). In its 2006 study, PCA found that the pyroprocessing step, which includes the combustion of fuels for the calcination of limestone at the cement kiln and preheaters, accounts for 91% of the total energy use during ordinary portland cement production (Marceau et al., 2006). The Intergovernmental Panel on Climate Change (IPCC) has focused on cement production as a major source of GHGs, and has developed three methods for calculating CO_2 emissions from the calcination of limestone to support the environmental evaluation of the industry: estimating clinker production using cement production data, using direct clinker production data, or using carbonate input data (Hanle et al., 2006).

Studies exploring a wider range of environmental impacts associated with concrete production have highlighted significant releases of particulate matter (PM) (Schuhmacher et al., 2004; Van den Heede and De Belie, 2012) as well as metals, dioxins, and furans resulting from fuel combustion (Gursel et al., 2014; Marceau et al., 2006; Rovira et al., 2010; Van den Heede and De Belie, 2012). Miller et al. (2018a) found that concrete production accounted for 9% of global industrial water withdrawals in 2012. Mack-Vergara and John (2017) suggest that the available inventories for water use are inconsistent and are influenced by localized factors including water scarcity, quality, and local climatic conditions. These studies emphasize the other potentially significant environmental effects associated with cement production at facilities located near populations that are exposed to air emissions.

Life-cycle assessments (LCAs) of concrete and cement have primarily focused on process improvement through the use of alternative fuels (Georgiopoulou and Lyberatos, 2018; Zhang and Mabee, 2016) and the replacement of portland cement with alternative materials, but do not provide reliable insight into the current operations of cement production facilities. Gutiérrez et al. (2017) utilized data gathered directly from a cement manufacturing facility in Cuba, while Biswas et al. (2017) gathered data directly from a concrete manufacturer in Qatar, but these studies are limited to individual facilities and are not representative of a wider range of production technologies. Gursel et al. (2014) presented a review of previous LCAs for concrete and raw materials and highlighted three key areas for improvement: 1) provide “holistic assessment of environmental impacts”—in particular, going beyond just energy use and GHG metrics; 2) apply “regional and technological variations,” including differentiating production technologies and differences in local supply chains; and 3) do not neglect aspects deemed insignificant on the basis of assumptions or past studies, as these could still be significant in the context of global production.

There are efforts by PCA and the National Ready Mixed Concrete Association (NRMCA) to produce environmental product declarations (EPDs) for the industries they represent (NRMCA, 2021; PCA, 2021), but these industry-average summaries are limited in the data they provide. The PCA inventories are based on the Global Cement and Concrete Association (GCCA) EPD tool, which was designed for manufacturers who want to generate EPDs for their products (GCCA, 2020). While this is a useful tool that enables cement and concrete manufacturers to generate internally coherent EPDs within the industry, it does not provide the transparency or accessibility to be useful for research efforts. Thus, there is a gap in the availability of transparent and representative environmental performance data for cement production.

The purpose of the present study is to produce an objective, transparent, and complete baseline life-cycle inventory (LCI) for concrete production, focusing on U.S. conditions, and with sufficient subprocess detail to support location-specific assessments based on case-specific parameters. The inventories developed for this study are suitable for characterizing concrete in industry-average modeling efforts and provide a tool for quantifying the impacts associated with specific cement and concrete production facilities or technologies. This study represents the industry on the basis of its publicly reported environmental releases, updating previous inventories (Argonne National Laboratory, 2017; Marceau et al., 2006, 2007) in accordance with the most recently reported values across the same data year, facility capacities, and production volumes. This consistency is important, as kiln technologies are improving and demand patterns are changing. Critically, this approach will provide researchers with baseline inventories so they can generate comparative assessments to traditional portland cement production.

The model produced for this study is made publicly available as part of the Greenhouse gases, Regulated Emissions, and Energy use in Technologies (GREET) model and as unit-process datasets downloadable from the Federal LCA Commons (Federal LCA Commons, 2021). The new unit-process datasets developed for this study consider the full range of environmental releases to support life-cycle impact assessment across impact categories using the Tool for Reduction and Assessment of Chemicals and Other Environmental Impacts (TRACI) (U.S. EPA, 2014) or other life-cycle impact assessment methods. The cradle-to-gate results presented here illustrate the potential of the inventories to provide accounting for emissions including volatile organic compounds (VOCs), CO, NO_x, PM_{10}, $PM_{2.5}$, SO_x, black carbon, organic carbon, CH_4, N_2O, and CO_2 based on supply-chain datasets from GREET. Gate-to-gate results are also presented for TRACI impact-assessment metrics and normalized U.S. totals (Ryberg et al., 2014) (Tables SI–1).

2. Methods

2.1. Scope, functional unit, and system boundary

Two functional units are used for this study: (1) 1 kg of dry portland cement and (2) 1 kg of wet mixed concrete at a ready-mix batching plant. The system boundary is cradle-to-gate, from extraction of raw materials to wet concrete mix at a batch mixing facility ready for delivery to a construction site (Fig. 1). This includes the upstream raw materials production of gypsum, clay, sand, and aggregate, and their transportation. Results include those for U.S. average concrete as well as technology-specific pathways for wet- and dry-kiln systems, classified following the convention used by the PCA’s U.S. Labor-Energy Input Survey 2016 (Sullivan et al., 2016): wet, dry with no preheater, dry with preheater, and dry with a combined preheater-precalciner system. These technologies are henceforth referred to as “wet,” “dry,” “preheater,” and “precalciner,” respectively. The GREET 2019 model is used to represent the supply chains of inputs to concrete production that are not explicitly modeled here, and the cradle-to-gate results include all metrics tracked by GREET 2019, including GHG emissions, criteria air pollutant (CAP) emissions, water consumption, and energy use by type. New unit process data were created for portland cement production, ready-mix concrete batch plants, and limestone, gypsum, and aggregate quarrying. GREET 2019 is used for the supply chains of inputs to these processes. The unit processes created for this study also include a more complete inventory of releases to air, water, and land based on environmental inventory datasets described in the next section. In addition to the cradle-to-gate LCI results, gate-to-gate impact assessment results are presented for portland cement production based on the more complete gate-to-gate inventory and using TRACI to characterize potential life-cycle impacts. The foreground datasets created for this study generally reflect 2016 datasets, as those were the most recent energy and facility capacity estimates available from the PCA’s surveys. Major energy supply chains and emissions factors in GREET are updated annually, generally reflecting current conditions.

2.2. Data sources

Environmental release data for the unit processes identified for this study were sourced from publicly available datasets following an approach similar to that described by Cashman et al. (2016). These data were compiled to provide a representative inventory for U.S. cement

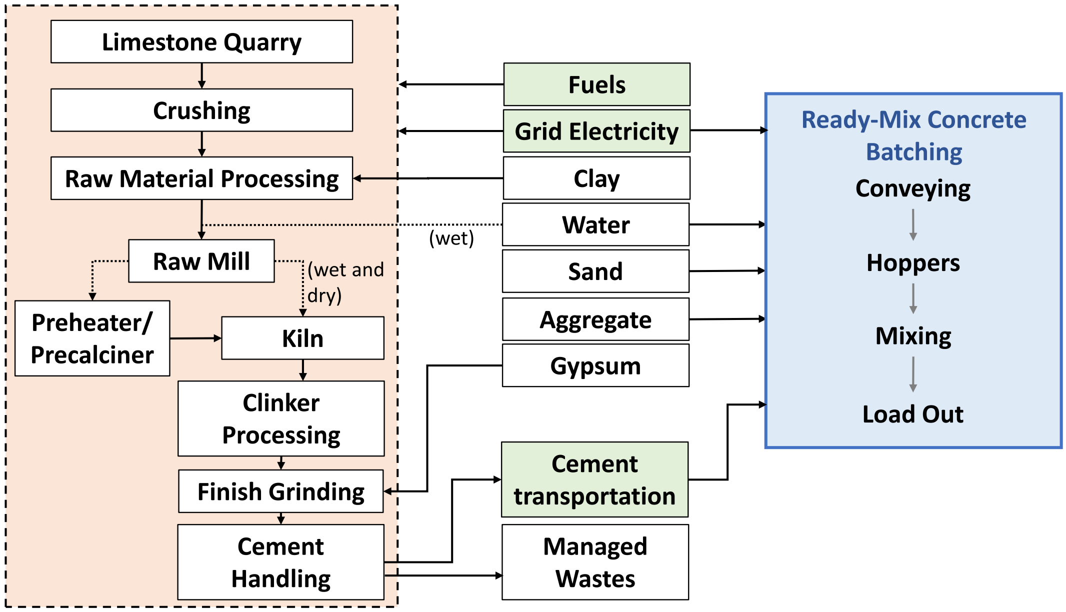

Fig. 1. System boundary diagram including subprocesses for the primary foreground processes, cement and concrete production. Supply chains from GREET are shaded green and include fuel combustion emissions and on-site vehicle operation. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

production based on facility-level emissions data identified using the North American Industry Classification System (NAICS) code for Cement Manufacturing, 327310 (US Census Bureau, 2019). Environmental releases from cement production facilities in 2016, including those from on-site limestone quarrying, were sourced from the National Emissions Inventory (NEI) (U.S. EPA, 2018a), Greenhouse Gas Reporting Program (GHGRP) (U.S. EPA, 2018b), Toxic Release Inventory (TRI) (U.S. EPA, 2018c), and Discharge Monitoring Reports (DMR) (U.S. EPA, 2016a).

The 2016 NEI v1 is a special inventory prepared for modeling purposes and includes the larger point-source facilities as well as smaller facilities and some records carried forward from the 2014 NEI. The Air Emissions Reporting Rule requires state, local, and tribal air agencies to submit emissions inventory data to EPA every year for larger point-source facilities and requires all sources of emissions to be reported every three years. In the NEI, facilities report releases of CAPs and hazardous air pollutants (HAPs) at specific emissions sources by Source Classification Codes. Releases of HAPs are also reported in the TRI. Releases to water are reported as facility totals to the TRI and DMR. Some TRI flows are reported as managed wastes rather than environmental releases. Under the Greenhouse Gas Reporting Rule (40 CFR Part 98), regulated facilities report releases of GHGs to the U.S. EPA, and these data are published annually in the GHGRP. Relevant releases are reported by cement producers under Subpart H - Cement Production, and Subpart C - General Stationary Fuel Combustion Sources.

2.3. Cement production processes

Cement production begins with the quarrying of limestone and clay (Fig. 1), which are the primary raw constituents of portland cement. Typical cement production facilities are located at or adjacent to the limestone quarry itself. Clay and gypsum may be available on-site, depending on local geology (this is the case for 54% of total U.S. cement by mass), or are quarried off-site and transported to the cement production facilities via trucks (42%) or barge (4%); see Tables SI–5 for details (Marceau et al., 2006). These raw materials are handled, processed, and stored at cement facilities.

Crushing, a two-stage process to reduce the size of the quarried stone, precedes raw-material processing, which includes storage, handling, and grinding of the primary ingredients of clinker (i.e., "kiln feed") prior to pyroprocessing (i.e., preheaters, precalciners, and kilns). Wet facilities also add water to the kiln feed to create a slurry which is fed into the kiln. Facilities using dry feed may have preheaters and precalciners which preprocess the feed by utilizing waste heat from combustion in the kilns (PCA, 2018a). This technology improves energy efficiency at cement plants and enables greater throughput (Marceau et al., 2006). Some older facilities have not been updated to take advantage of these systems; in these cases, the raw mill feeds directly into the kiln.

The kiln is the site of the primary combustion process at a cement production facility and can use a variety of fuels, including coal and natural gas as well as alternative fuels like waste oil, solvents, tires, and other solid wastes. As the kiln feed passes through the kiln, it is exposed to increasing temperatures, eventually peaking around 1400–1450 °C. As the temperature of the kiln feed increases, chemical reactions take place, driving CO_2 out of the limestone (primarily as $CaCO_3$) and producing clinker, which consists of various calcium silicates including alite, Ca_3SiO_5 with impurities, and belite, as well as other compounds such as tricalcium aluminate and calcium aluminoferrite (Kosmatka, 2012). The clinker is then cooled and is passed through finish-grinding operations, at which point additional limestone and gypsum are added to improve the properties of the cement mix, such as particle size distribution, to prevent flash setting. The cement powder is screened and siloed for storage prior to shipment to concrete batch plants.

2.4. Determining cement production rates and CO_2 emissions for facilities

Annual facility cement production values (Tables SI–3) were calculated by first adjusting clinker production capacity data for each facility from PCA reports, using regional capacity utilization rates from the Minerals Yearbook published by the U.S. Geological Survey (USGS, 2015). These values were then rebalanced using the PCA U.S. Labor-Energy Input Survey 2016 (Sullivan et al., 2016) utilization rates by technology type and adjusted to match total national capacity from the U.S. Portland Cement Industry: Plant Information Summary (PCA, 2018b). The clinker production energy per mass of clinker is calculated on the basis of the amounts of each fuel and electricity divided by the clinker production based on the values for each kiln technology presented in the PCA U.S. Labor-Energy Input Survey 2016 (Sullivan et al., 2016). These values are then scaled up to the U.S. total energy use using the average cement-to-clinker ratio of 0.906 ton/ton and the total annual cement production for each kiln technology, estimated as just described.

GREET profiles for fuel combustion were used in conjunction with fuel usage reported by PCA to determine combustion CO_2 emissions. Waste fuels are credited with avoidance of methane emissions associated with the anaerobic decomposition of wastes in landfills, considering U.S. average landfill conditions, degradation rates of waste components, timing of landfill capping, landfill gas capture, and flaring rate after capping (Lee et al., 2017). The CO_2 emissions from combustion of waste fuels are estimated on the basis of their carbon content. Separate emission factors (EFs) based on reported GHGRP data served as a comparison with the fuel carbon content approach.

The calcination rate used for cement was 553 kg CO_2 per tonne of cement, in accordance with an earlier LCI of cement performed by PCA (Marceau et al., 2006). For validation, the calcination rate was estimated based on the difference between reported GHGRP emissions and GREET fuel combustion data, yielding an estimated national average calcination rate of 498 kg CO_2 per tonne of cement. This rate corresponds to other cited calcination rates, although regional variation in cement types may affect these estimates (Ke et al., 2013). Other estimates place the value at 517 kg CO_2 per tonne of cement (R. Bohan, personal communication, 2021). The cited number from the previous PCA study of 553 kg CO_2 per tonne, based on stoichiometric estimates and publicly reported by PCA, is used here and reflects a more conservative estimate.

Table 1 presents the total production of cement, fuel shares, and CO_2 emission rates by technology type in 2016. The sum of the rebalanced cement production totals by technology type fall within one percent of U.S. total cement production (USGS, 2019). The cement production values by technology type are allocated to facilities, resulting in a ±0.6% deviation from the nationally adjusted USGS regional clinker utilization rates, maintaining the relative contribution for each region. The EFs from combustion and calcination are compared to those calculated from GHGRP in the bottom two rows of the table. CO_2 emissions from fuel combustion, using GREET fuel profiles combined with the estimated calcination rate from PCA, are about 8% higher than emissions calculated on the basis of GHGRP reporting.

2.5. Creation of emissions factors

Emissions factors for the processes involved in the production of concrete were created by normalizing environmental releases reported in the NEI, TRI, and DMR datasets by production levels for each activity. CO_2 from cement production. For upstream quarrying processes (e.g. clay and gypsum), releases were aggregated nationally and EFs are calculated based on total production of each material. Emissions factors for downstream concrete mixing processes were performed at the state level and the method is described in the following section. For the cement production process, emissions factors were developed at the facility level, and for air emissions, at the subprocess level by facility. To do this, the annual releases of each substance at each facility were divided by its estimated annual cement production. Air emissions factors were also calculated for subprocesses within individual facilities using an approach of standardizing the process flows for cement (shown in Fig. 1) and matching with subprocess information provided by the eight-digit Source Classification Codes reported in the NEI. The method used here follows the approaches developed for and used in our prior work (Sun et al., 2019a,b; Young et al., 2019a,b). As both the NEI and TRI report air emissions, instances of duplicate reports for the same substance from the same facility were resolved by preferring the NEI data. Where the total of the emissions of a given substance at a facility reported by TRI exceed the sum of subprocess emissions reported by NEI, the difference is retained and accounted at the facility level. Releases to air, water, and soil associated with managed waste flows were estimated based on the management practices specified by the TRI using a method described previously (U.S. EPA, 2015; Young et al., 2019).

Annual facility production amounts were estimated by adjusting clinker capacity data with capacity utilization rates. Clinker capacity data for 2015 and 2016 are provided by the PCA's U.S. Portland Cement Industry: Plant Information Study (PCA, 2018b). Regional clinker capacity utilization rates are reported by the USGS (USGS, 2015) for 2015. The regional capacity utilization for 2016 was estimated by adjusting the 2015 capacity utilization to reflect the change in clinker capacity from 2015 to 2016. The total cement output for each facility was estimated using these values and the ratio of clinker to cement in the primary mix produced by each facility. These estimates were further adjusted so the facility-level cement production values match the totals by technology type reported by PCA (PCA, 2018b).

2.6. Parameterization of cement data

Results were calculated with 90% prediction intervals for releases on the basis of the regression of facility-level estimates for each elementary

Table 1
Annual cement production rates, total energy use in cement production, and CO_2 releases from fuel combustion during cement production.

			Wet		Dry		Preheater		Precalciner	Totals
Technology Contribution to Nat. Avg.			3%		5%		12%		80%	100%
Cement Production (Mtonnes)			2.60		4.49		9.92		65.7	82.7
Total Energy Use at Kiln (10^6 MMBtu)			17.5		24.0		42.5		249	333
Fuels	*CO_2 EFs (g/MMBtu)*	*Share*	*CO_2 (ktonnes)*	*Share*	*CO_2 (ktonnes)*	*Share*	*CO_2 (ktonnes)*	*Share*	*CO_2 (ktonnes)*	
Resid. Oil	85,081	0.1%	1.49	0%	–	0.4%	14.5	0.4%	84.8	
Diesel	78,199	0.2%	2.74	1.9%	35.7	0.6%	19.9	0.8%	156	
Gasoline	76,839	–	–	–	–	–	–	–	–	
Pet Coke	106,976	14%	261	61.2%	1,570	7.6%	346	16.1%	4,290	
Natural gas	59,413	12%	120	5.9%	84.2	22.2%	561	15.5%	2,290	
Coal	89,920	21%	334	19.0%	410	45.5%	1,740	40.9%	9,160	
Waste	145,882	17%	430	0.7%	24.5	6.9%	428	3.9%	1,420	
Tire Fuel	60,876	4.4%	47.0	1.6%	23.4	5.6%	145	3.9%	591	
Solvents	72,298	25%	316	–	–	–	–	5.4%	972	
Waste Oil	77,758	0.4%	5.45	–	–	–	–	0.9%	174	
Renewables	0	0.1%	0	0.0%	–	0.0%	–	0.9%	0	
Fuel CO_2 Subtotal (Mtonnes)			1.52		2.15		3.25		19.1	26.0
Calcination CO_2 (Mtonnes)			1.44		2.48		5.49		36.3	45.8
Total Calculated CO_2 (Mtonnes)			2.95		4.64		8.74		55.5	71.8
CO_2 EFs GHGRP (tonnes per tonne)			0.96		0.87		0.85		0.78	0.80
Total CO_2 GHGRP (Mtonnes)			2.51		3.89		8.45		51.5	66.3

*Fuel combustion emissions factors in "CO_2 EFs" are based on lower heating values (LHVs).

flow, consistent with methods employed in prior research (Young et al., 2019). The prediction intervals are evaluated as a function of the standard error of the prediction around the expected releases at the mean for each kiln combustion technology used in U.S. cement production facilities. The parameters of the distribution are calculated such that the expected value of the log-normal distribution is set equal to the EF. These parameters were used to populate the uncertainty distributions in openLCA software (GreenDelta, 2018) for the inventories of each of the four distinct kiln technologies, enabling stochastic modeling to assess uncertainty. The openLCA software has the capability of running Monte Carlo simulations, which are a statistical approach to modeling that uses randomly selected data points for variables with defined distributions, generating a series of results for many iterations of the model and providing statistically relevant data for probabilistic interpretation. For this study, the model was iterated using ten thousand runs per technology type.

Inventory data for other processes, not reported as point-source emissions, were sourced from the PCA's cement LCI (Marceau et al., 2006). These include on-site water use, diesel and gasoline consumption in mobile equipment, and electricity demand for upstream processes and at the ready-mix facility. All upstream fuel production, on-site mobile equipment, electricity, and transportation parameters use default values from GREET 2019.

2.7. Concrete mixing

Concrete, in its most basic form, is a combination of water, aggregate, and cement, which serves as a binder throughout cured concrete pavement. Examples of aggregate include sand, gravel, crushed stone, and iron blast-furnace slag. Raw materials are delivered to a concrete mixing plant by rail, truck, or barge. The constituents are fed by gravity or screw conveyor to weigh hoppers, which combine the proper amounts of each material. Additional chemical admixtures and supplementary cementitious materials (SCMs) may be added to specific concrete blends. Because these additional materials vary widely and are not generalizable to modeling that characterizes national concrete production, they are not included in the inventory described here. The mixture is sent out by specialized ready-mix concrete trucks as needed to job sites.

Concrete mixing can occur at a variety of plant types tailored to different applications. Owing to data availability, this study uses data for ready-mix facilities and assumes these data to be a reasonable approximation of concrete production at mobile batch plants, which are often used for large projects. Data reported to the NEI, TRI, and DMR for ready-mix concrete facilities were compiled and harmonized. These were then aggregated to the state level and matched with ready-mix concrete production volumes by state from USGS (2019) and NRMCA (2015), as facility-specific production volumes could not be estimated. To correct for incomplete reporting, state-level ready-mix concrete production amounts were multiplied by the percentage of ready-mix facilities reported by the release inventory datasets compared with those reported by the Bureau of Labor Statistics (BLS, 2019). State-level EFs were only included for states where the share of ready-mix facilities reporting to the release inventory datasets exceeds 50% to avoid spurious results. Details are provided in Tables SI–10.

The production levels for states that met the threshold for inclusion were calculated by combining state-level production shares from NRMCA (2015) and the 2016 production volume of ready-mix concrete in the U.S., which was reported as 342.9 million cubic yards (262.2 million cubic meters) (The Concrete Producer Staff, 2017). These state-level production volumes were adjusted by applying the percent of reporting facilities in NEI based on total facilities according to BLS (2019). Ready-mix facilities do not report to GHGRP, so a different approach was needed to estimate GHG emissions. GHGs were calculated by applying ratios from GREET combustion profiles to nitrogen dioxide releases reported to the NEI. EFs using TRI and DMR data were developed using all the available records and applying total state-level and adjusted production rates to create uncertainty parameters. For quarried sand and gravel, just as with clay and gypsum in the cement system, releases are aggregated, and EFs are industry-wide. These impacts are then allocated on a mass basis for the contribution of 35% sand and 49% gravel to ready-mix concrete production, with the remaining contributions of 9% cement and 7% water to represent a general-use concrete (Marceau et al., 2007). GREET enables the user to alter the material shares in the concrete if a specific mix is preferred. As with cement production facilities, upstream processes were accounted for using GREET profiles in the concrete supply chain.

The cradle-to-delivery inventories include ready-mix concrete delivered to a job site, but do not include any site prep, forming, rebar, rolling, or finishing that may be associated with the use of concrete in transportation infrastructure projects.

2.8. Cement and concrete transportation

Transportation distances for cement from the production site to ready-mix facilities were developed using geographic coordinates. ArcGIS software (Esri, 2019) was used to map the latitude and longitude for all cement facilities and all reporting ready-mix facilities. The average distance between each ready-mix facility and the nearest cement facility was 112 km (70 mi). Three modes of transportation were included: truck, rail, and barge. The parameterized distances were used to model the truck and rail shipments. This approach assumes that cement is sourced from the closest production location. The distance for barge shipments as well as the share of each mode of transportation are provided in the 2006 PCA cement LCI (Marceau et al., 2006).

3. Results and discussion

The presentation of the results begins with total system-wide emissions by major concrete-related processes from a cradle-to-delivery perspective. These results highlight the significance of cement production to the overall impacts associated with delivered concrete for nearly all the emissions reported as GREET metrics. While cement is one of the smallest inputs to concrete production by mass (9%), cement production dominates most of the GREET metrics associated with cradle-to-delivery of the production system. The focus then shifts to the production of cement on a gate-to-gate basis for each technology type, including uncertainty parameters for U.S. facilities utilizing TRACI impact categories for the complete inventory. The impacts of cement production from precalciner facilities are then assessed on a subprocess basis, affirming the dominance of the kiln process, which includes emissions from fuel combustion and calcination. This subprocess-level evaluation is presented in terms of both the GREET emission profile and the TRACI impact categories.

3.1. Contributions by life-cycle stage to concrete production

The cement production stage drives most of the GREET metrics for the cradle-to-delivery production of concrete (Fig. 2). The other life-cycle stage driving releases across categories is the ready-mix production process. For methane releases, upstream fuel production also plays an important role. Pyroprocessing and calcination emissions are the primary contributors to on-site emissions of CAPs and GHGs. Dust is the major contributor to PM_{10} and $PM_{2.5}$, coming primarily from ready mix facilities and limestone quarrying activities. NO_x emissions result from pyroprocessing and are the main flows contributing to photochemical oxidant formation potential (POFP) across technology types. Cement pyroprocessing shows a negative value for methane releases, owing to avoided landfill-gas production as a result of combusting wastes that would otherwise enter landfills. The other processes within the concrete production life cycle (transportation, delivery to site, gypsum and clay production) are minimal contributors compared to the cement categories and ready-mix production. Transportation accounts for delivery

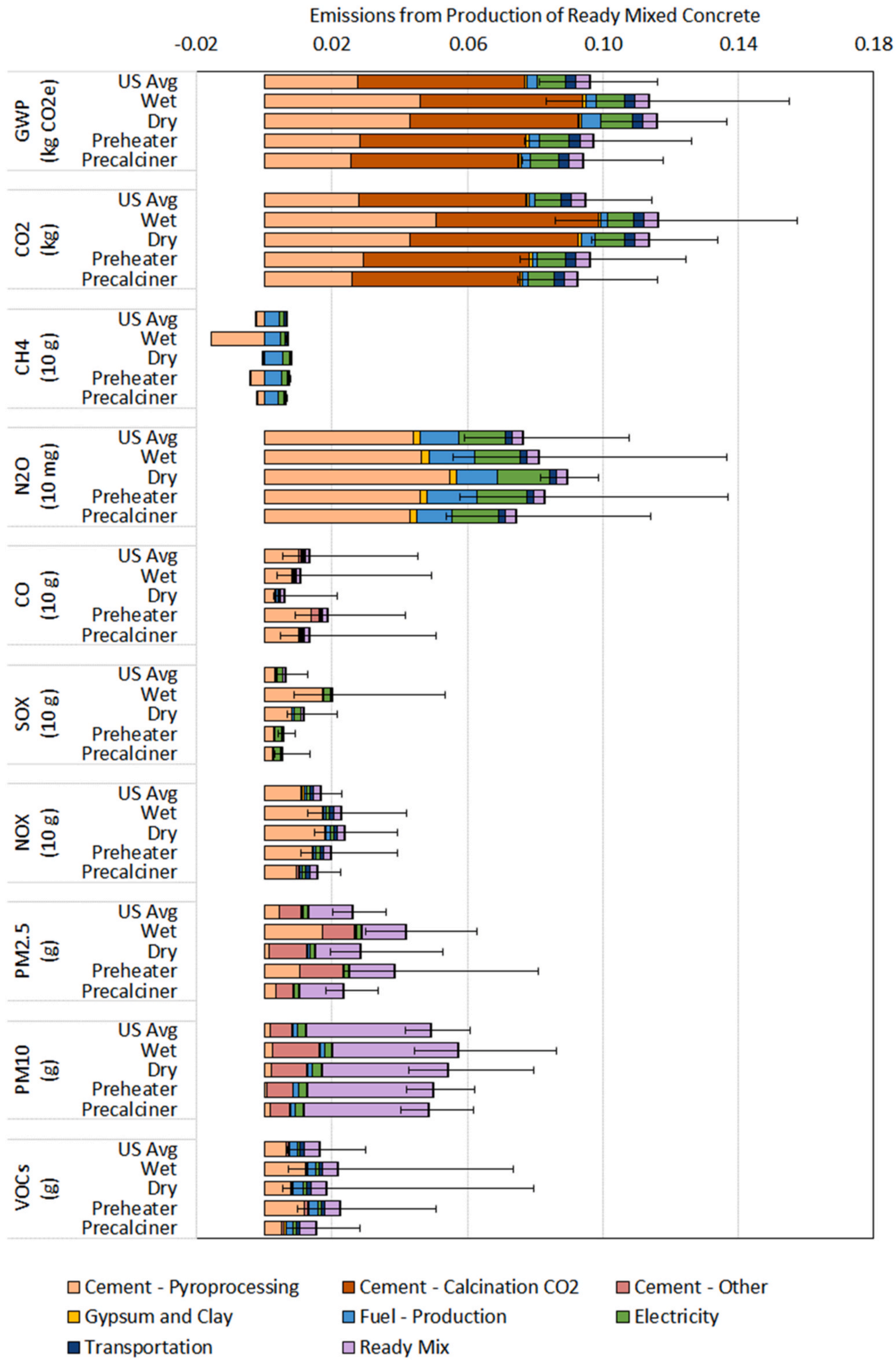

Fig. 2. GHG and criteria air pollutant emissions for concrete production by kiln technology used to produce the portland cement. *The stacks show contributions of life-cycle stages, and whiskers show the 10*th *and 90*th *percentile of the Monte Carlo results generated in openLCA (*Tables SI–7*).*

of all inputs to portland cement production, vehicle use within the facility gates, and delivery of inputs to the ready-mix concrete facility. Fig. 2 demonstrates the ability of these inventories to provide detailed, impact-specific results on a subprocess level with probability ranges, including cement production, transportation, and ready-mix operations.

3.2. On-site, gate-to-gate emissions for cement production

Precalciner kilns dominate the national cement production profile, accounting for ~80% of U.S. cement production (Table 1). The results presented in Fig. 3 show that cement production at precalciner facilities results in fewer impacts at the production site with respect to global warming potential (GWP), acidification potential (AP), particulate matter formation potential (PMFP), and POFP. Normalized impacts in

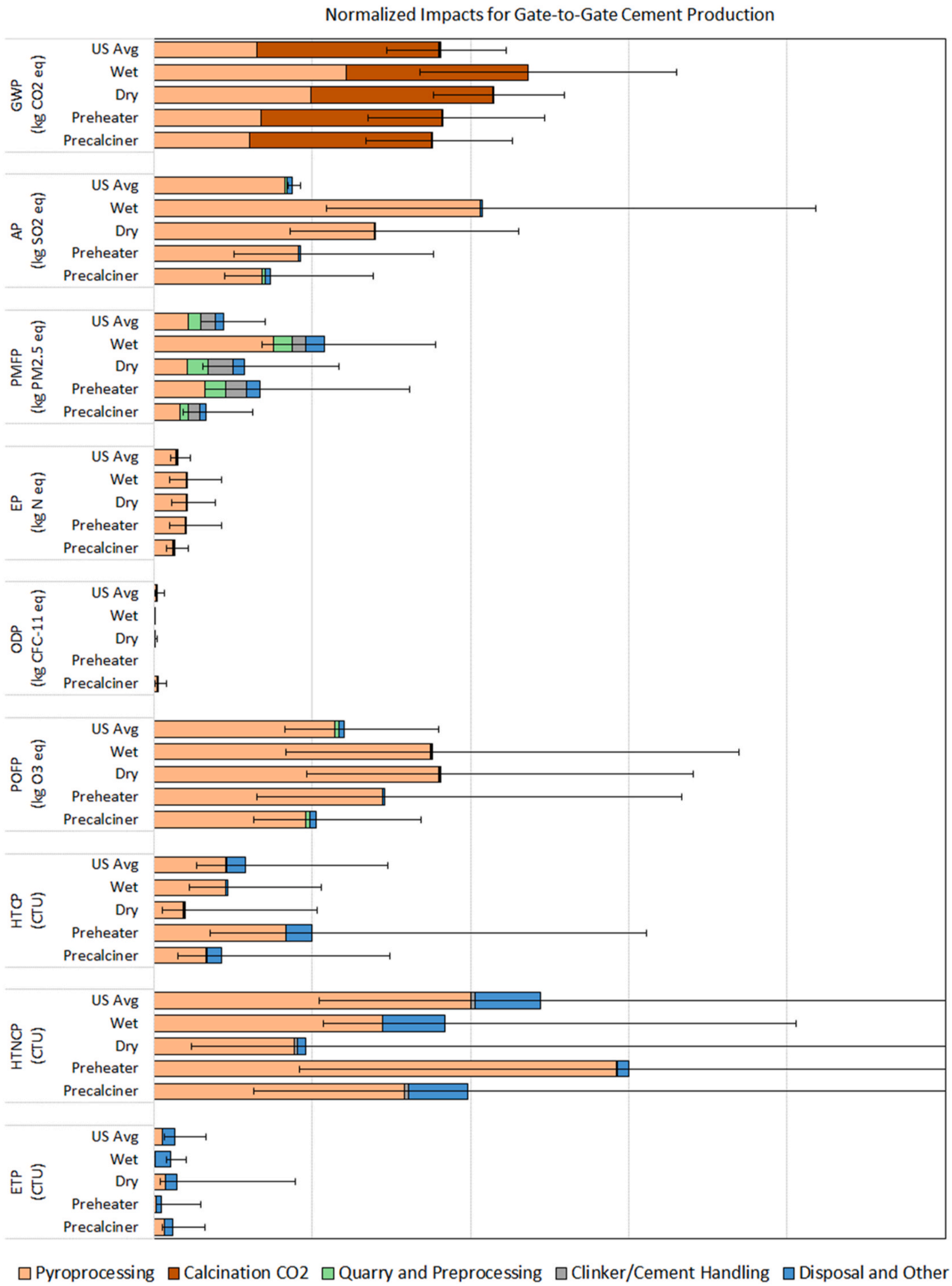

Fig. 3. Process contributions to the normalized gate-to-gate midpoint impacts of cement production. *Stacks show process contributions and whiskers show the range of Monte Carlo results from the* 10th *to* 90th *percentiles. GWP: global warming potential, AP: acidification potential, PMFP: particulate matter formation potential, POFP: photochemical oxidant formation potential, EP: eutrophication potential, ODP: ozone depletion potential, HTCP: human toxicity – cancerous potential, HTNCP: human toxicity – non-cancerous potential, ETP: ecotoxicity potential. The* 90th *percentiles, which have been cut off for HTNCP and all other values, are available in* Tables SI–8.

Fig. 3 represent contributions by each impact category to total U.S. impacts per capita in 2008 (Ryberg et al., 2014). Precalciner kilns are more efficient, requiring less energy per unit of cement. Given the values in Table 1, the MMBtu of fuel used per tonne of cement is 6.75 for wet, 5.35 for dry, 4.29 for preheater, and 3.79 for precalciner pathways. Fig. 3 demonstrates the ability of these inventories to provide detailed, impact-specific results on a subprocess level with probability ranges for cement production.

The impact categories that do not follow this expected trend are human toxicity – cancerous potential (HTCP), human toxicity - non-cancerous potential (HTNCP), and ecotoxicity potential (ETP). For these three impact categories, the results for the precalciner facilities are mixed. The mercury from limestone used in precalciner facilities likely drives the increased releases; emissions of mercury are responsible for the increase in HTNCP. Mercury emissions are a function of raw material and fuel inputs and can be correlated directly to local geology. Mercury emissions are known to occur at cement production facilities because of the mercury content of limestone. In the absence of mercury control devices on the kiln, the mercury in limestone can volatilize and be released to the air (Kosmatka, 2012). Mercury is explicitly named in a series of EPA regulations of the portland cement industry which have been implemented over the past twenty years to help limit air toxic emissions (U.S. EPA, 2019b). Precalciner facilities seem to be associated with more mercury-laden feedstocks than facilities with other kiln technologies.

Fig. 3 also provides insight into which contributing subprocesses are the primary sources of impacts. The kiln contributes the majority of emissions for every impact category, with GWP being split between calcination (65%) and fuel combustion emissions (35%) at the kiln for precalciner facilities. The only categories that show major contributions from non-kiln emissions are PMFP and HTNCP. The particulate emissions are associated with on-site dust, while the human health impacts are primarily associated with mercury emissions to air. The non-combustion emission contributions to the human toxicity categories are primarily associated with mercury and zinc emissions to water, while the pyroprocessing impacts are associated with the same metals being released to air (Tables SI–9). There are also significant impacts associated with human and ecotoxicity resulting from zinc emissions to air, especially in cement production at wet kilns.

3.3. Comparison with previous inventories

The results published in this study differ from previous studies from the PCA because they are independently calculated by reconciling emissions reported by individual cement and concrete facilities to federal reporting programs with independent bottom up calculations based on GREET emission factors. PCA's methods for generating LCIs of cement and concrete, published in 2006 and 2007, respectively, do not incorporate emissions reported by facilities and incorporate other emission factors (Marceau et al., 2006, 2007).

The comparison in Table 2 is based on a U.S. average concrete mix for the inventory developed for this research, while the PCA results reported are for what PCA calls “Mix 3” in the 2007 LCI (Marceau et al., 2007), which is approximately the same mix of concrete ingredients. While these are the results generated for comparison here, the GREET-based model and LCI enable users to define specific mixes or introduce additional chemical admixtures or SCMs.

Table 2
Nationally averaged LCI results for cement and concrete calculated in this study, compared to the PCA-developed inventories for cement and concrete (Marceau et al., 2006, 2007) and the more recent PCA Portland Cement EPD (PCA, 2021).

		Cement cradle-to-gate (tonne cement)			Concrete cradle-to-gate (tonne concrete)	
		Calculated National Average	PCA (2006)	PCA EPD (2016)	Calculated National Average	PCA (2007)
CO_2-eq.	kg	959	927	1040	97.6	88.0
CO_2	kg	938	927	-	95.3	87.9
CH_4	g	364	4.76	-	44.9	3.65
N_2O	g	6.93	-	-	0.748	-
SO_X	g	417	1,660	-	47.3	120
$PM_{2.5}$	g	128	0.0911	-	26.9	0.0153
PM_{10}	g	235	296	-	77.0	215
NO_X	g	1,360	2,500	-	168	237
CO	g	1,260	1,100	-	135	113
VOCs	g	109	50.2	-	81.5	6.33

The results of this comparison generally reflect improvements in the environmental profile of the industry between the time of PCA's inventory and this study, as well as some areas where this study improves on the earlier estimates. The PCA's inventory is based on energy data for 2002. The GWP presented as CO_2-eq. can be compared to both the PCA LCI and PCA's more recent EPD, with the results generated here falling between the reported PCA values and within 8% of each. The largest flow, which is CO_2, is within 10% in both the cement and concrete inventories and, together with CO, is the emissions value that changed least between PCA's 2006 inventory and the present study. The reductions in NO_x, SO_x, and PM_{10} are likely due to changes in the fuel mix and combustion technologies (e.g., low-NO_x burners and selective non-catalytic reduction) since 2002. As an example of shifts in the fuel mix, the PCA cement inventory shows 60% coal use, while the present study estimates approximately 40% coal use. Natural gas and waste fuels have provided the additional fuel to offset the reduction in coal use. Other factors accounting for differences across these inventories may include differences in system boundaries and discrepancies in the concrete mix. For example, the largest difference is for $PM_{2.5}$; the reason is that the method PCA used to determine particulates from the quarrying process, which is the primary source of these emissions, did not provide speciation of $PM_{2.5}$ from PM_{10}. As a result, the total $PM_{2.5}$ releases reported in the PCA inventory are also much lower than the reported values in the NEI. The increase in methane emissions is caused by the increased use of natural gas, which went from 4% of the energy inputs for U.S. average portland cement on an energy basis in PCA's study to 16% in the present study. Most of these methane emissions are direct from the cement production process, as reported by the GHGRP, presumably due to methane slip.

The inventories presented here provide updated metrics that serve as objective benchmarks, based on publicly accessible data, which can be used for comparison with the product category rules underlying EPDs. The results generated using the inventories demonstrate the benefits of inventories containing environmental release data that have subprocess resolution for technology-specific cement and concrete production systems. The inventories are used for modeling transportation infrastructure in GREET and provided for general use as LCA datasets through the U.S. Federal LCA Commons. Both the GREET emissions data and openLCA inventories enable editing of both the cement production technology mixes and the fuel mix used in each technology category, which aids in developing temporally appropriate results.

When the results are scaled to the 88.5 million tonnes of cement used in the U.S. in 2019 (USGS, 2020) and assuming an average cement content of 9% in concrete (excluding any SCMs), the resulting total CO_2 emissions from U.S. concrete consumption is estimated to be 95 million tonnes CO_2-eq. This is 12% of U.S. domestic industrial GHG emissions and 1.4% of the total U.S. domestic GHG emissions in 2018 (the most recent year reported). Considering that 30% of U.S. cement use is for transportation infrastructure (PCA, 2016), the emissions from the production of concrete for transportation infrastructure is roughly 1.5% of the total GHG emissions from fuel combustion for road transportation (U.S. EPA, 2020b). Emissions from concrete production for transportation infrastructure also amount to 2% of NO_x, 8% of $PM_{2.5}$, 10% of PM_{10}, and 60% of SO_x emissions from fuel combustion for road transportation, on the basis of the 2017 NEI (the most recent year reported) (U.S. EPA, 2020a). This is, of course, a rough calculation based on coarse estimates of the concrete mixes used; nonetheless, it provides a useful sense of scale for gauging the significance of emissions from concrete production.

The scope of this study and these inventories is limited to cradle-to-gate for cement and concrete production. Previous LCAs have explored a wide range of potential changes in the concrete supply chain to improve environmental performance. Georgiopoulou and Lyberatos (2018) and Zhang and Mabee (2016) evaluated alternative fuel mixes. Miller et al. (2018a,b), Ruan and Unluer (2016), Tait and Cheung (2016), Anastasiou et al. (2015), and Gartner (2004) assessed technologies to replace concrete or cement ingredients. Some studies have addressed the potential for cement to capture carbon during the production process (Sanjuán et al., 2020b; Sanjuán et al., 2020). These comparative LCA studies highlight the potential for process improvement in the concrete industry and the need for reliable baseline inventories, which the present research has delivered. Other studies address the potential for CO_2 absorption from the atmosphere by concrete over its lifetime (Galan et al., 2010; Pade and Guimaraes, 2007; Sanjuán et al., 2020a). The life-cycle impacts of concrete construction is left for future studies that could leverage the inventories developed here.

4. Conclusions

This study provides transparent emissions data for infrastructure modeling within the GREET framework, as well as robust inventories for openLCA modeling, available for free download on the Federal LCA Commons. The four primary kiln technologies employed to produce cement are all available, in addition to the U.S. national average blend, enabling specification of blends that may be more regionally appropriate for specific LCA studies. The ready-mix concrete inventory is appropriate for commonly available truck-delivered concrete and is also adaptable for specified cement blends, including adjustments for modeling batch plants that may be set up for larger infrastructure projects. These data enable transparent assessments of concrete applications with complete inventories across a full suite of life-cycle impact categories. Researchers and decision-makers can use GREET or the data on the Federal Commons to assess the impacts associated with major concrete infrastructure projects.

The inventories calculated in this study provide some notable improvements on previous life cycle inventories published for portland cement and concrete. First, the inventories produced by this study are the first to be based on the facility-level environmental release data publicly-reported by the U.S. Environmental Protection Agency. Using this information, this study increases the scope of environmental releases included in the gate-to-gate inventory to include consideration of over 300 substances reported by the NEI and TRI, refines the resolution of air emissions to include subprocesses within the cement production and concrete mixing facilities, and provides quantitative uncertainty information based on variation across facilities. This study serves as an independent confirmation of results presented by the PCA (Table 2). Results for carbon dioxide emissions and greenhouse gas emissions for portland cement do not differ significantly, the results from this study are larger by 1% and 3% respectively. The results from this study provide valuable updates to the values reported by the PCA for methane, nitrous oxide, PM2.5, and VOCs where it appears the previous PCA study may have been underreporting these emissions. This study also provides significantly lower results for emissions of sulfur oxides and nitrogen oxides compared with the PCA study. These differences are due to a combination of the incorporation of the effect of emissions control technologies in the values reported to the NEI and improvements in the industry since the PCA study. Finally, this study provides high-resolution, publicly-available data across subprocesses and impact categories that can be used to support future assessments of infrastructure projects.

CRediT authorship contribution statement

Troy Hottle: Formal analysis, Validation, Visualization, Writing – original draft, Writing – review & editing, and review and editing. **Troy R. Hawkins:** Conceptualization, Methodology, Supervision, Formal analysis, Validation, Writing – original draft, Writing – review & editing, and review and editing, Project administration. **Caitlin Chiquelin:** Formal analysis, Visualization, Writing – original draft. **Bryan Lange:** Formal analysis. **Ben Young:** Formal analysis, Visualization, Writing – original draft, Validation, Writing – review & editing, and review and editing, Project administration. **Pingping Sun:** Validation, Writing – original draft, Writing – review & editing, and review and editing. **Amgad Elgowainy:** Conceptualization, Funding acquisition, Supervision, Validation, Writing – review & editing. **Michael Wang:** Conceptualization, 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.2022.131834.

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