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RESEARCH ARTICLE

Uncertainties in greenhouse gas emission factors: A comprehensive analysis of switchgrass-based biofuel production

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Abstract

This study investigates uncertainties in greenhouse gas (GHG) emission factors related to switchgrass-based biofuel production in Michigan. Using three life cycle assessment (LCA) databases—US lifecycle inventory (USLCI) database, GREET, and Ecoinvent—each with multiple versions, we recalculated the global warming intensity (GWI) and GHG mitigation potential in a static calculation. Employing Monte Carlo simulations along with local and global sensitivity analyses, we assess uncertainties and pinpoint key parameters influencing GWI. The convergence of results across our previous study, static calculations, and Monte Carlo simulations enhances the credibility of estimated GWI values. Static calculations, validated by Monte Carlo simulations, offer reasonable central tendencies, providing a robust foundation for policy considerations. However, the wider range observed in Monte Carlo simulations underscores the importance of potential variations and uncertainties in real-world applications. Sensitivity analyses identify biofuel yield, GHG emissions of electricity, and soil organic carbon (SOC) change as pivotal parameters influencing GWI. Decreasing uncertainties in GWI may be achieved by making greater efforts to acquire more precise data on these parameters. Our study emphasizes the significance of considering diverse GHG factors and databases in GWI assessments and stresses the need for accurate electricity fuel mixes, crucial information for refining GWI assessments and informing strategies for sustainable biofuel production.

KEYWORDS
cellulosic biofuel, global warming intensity, greenhouse gas emission factor, LCA database, sensitivity analysis, switchgrass, uncertainty analysis

1 | INTRODUCTION

Understanding and addressing the uncertainties in life cycle assessment (LCA) is critical, particularly for biofuels, given their potential role in reducing greenhouse gas (GHG) emissions. LCA studies require extensive data, including material and energy inputs, emissions, and life-cycle data for processes involved in the foreground system.

This comprehensive data requirement, coupled with varying methodologies and assumptions, inherently introduces uncertainty into LCA results. Several studies have explored this uncertainty, examining how variations in input parameters and the selection of life cycle inventory (LCI) databases influence the outcomes of LCA studies (Baral et al., 2020; Heijungs & Lenzen, 2014; Hsu et al., 2010; Huijbregts et al., 2003; Igos et al., 2019; Kalverkamp et al., 2020; Miranda Xicotencatl et al., 2023; Mullins et al., 2011; Pauer et al., 2020; Shi & Guest, 2020; Spatari & MacLean, 2010; Wang et al., 2020; Zhao et al., 2019).

To address these uncertainties, the International Standard for Life Cycle Assessment (International Organization for Standardization, 2006) recommends performing uncertainty analysis. The Monte Carlo method, which involves random sampling, is widely used for this purpose. This method requires a probability distribution for each input parameter, with the lognormal distribution being a popular choice in LCA studies (Ciroth et al., 2016; Imbeault-Tétreault et al., 2013; Muller et al., 2016). Performing many Monte Carlo simulations, typically around 10,000 runs, is essential to obtain a statistically robust estimate of variance (Heijungs & Lenzen, 2014; Igos et al., 2019).

Moreover, Monte Carlo simulations have been instrumental in quantifying uncertainties in the lifecycle GHG emissions of biofuels, underscoring their significance within the realm of biofuel policies (Mullins et al., 2011; Spatari & MacLean, 2010). Utilizing beta, lognormal, triangle, and uniform distributions, researchers have modeled input parameters, as well as CO_2 emissions associated with land-use change (both direct and indirect) and soil emissions (i.e., CO_2, CH_4, and N_2O), in Monte Carlo simulations aimed at capturing uncertainties inherent in the biofuel system (Baral et al., 2020; Locker et al., 2019; Mullins et al., 2011; Spatari & MacLean, 2010; Wang et al., 2020). However, there are comparatively few studies that incorporate distributions for GHG emission factors into Monte Carlo simulations, indicating a potential area for further research and methodological refinement in biofuel impact assessments.

In conjunction with uncertainty analysis, sensitivity analysis plays a pivotal role in assessing the impact of input variations on LCA results to prioritize future research efforts aimed at reducing uncertainties. Two types of sensitivity analysis are available: local sensitivity analysis (LSA) and global sensitivity analysis (GSA) (Igos et al., 2019; Wolf et al., 2017; Zhao et al., 2019). Local sensitivity analysis focuses on evaluating how small changes in input parameters affect LCA results under specific conditions, with the goal of identifying the parameters that have the most significant impact on the model output under specific, local conditions. LSA provides insights into the immediate effects of changes in individual parameters. In contrast, GSA aims to assess how variations in input parameters across their entire ranges impact LCA results, considering interactions and dependencies among inputs, namely identifying the parameters that are most important in influencing the model output across the entire range of parameter values. The objective of GSA is to provide a comprehensive understanding of how changes in input parameters, both individually and collectively, affect LCA results across the entire range of inputs, offering a holistic perspective on LCA results under various conditions.

LCA studies conducted by four Bioenergy Research Centers (BRC), funded by the US Department of Energy [Center for Advanced Bioenergy and Bioproducts Innovation (CABBI), Center for Bioenergy Innovation (CBI), Great Lakes Bioenergy Research Center (GLBRC), and the Joint BioEnergy Institute (JBI)], have employed three different LCA databases to assess GHG emissions associated with biofuel production systems (see Table 1). These databases include the US lifecycle inventory (USLCI) database (National Renewable Energy Laboratory, 2012), GREET (Argonne National Laboratory, 2023), and Ecoinvent (Ecoinvent, 2023). As seen in Table 1, some studies relied exclusively on GHG emission factors from the GREET model, while others used emission factors from all three databases. When employing the unit process data from Europe or the rest of the world in the Ecoinvent database, the lifecycle emissions associated with products using those unit processes are estimated based on US electricity fuel mixes (Baral et al., 2019; Kim et al., 2019, 2020; Kim, Dale, Basso et al., 2023; Kim et al., 2023a; Neupane et al., 2017).

The USLCI database, developed by the National Renewable Energy Laboratory (NREL) in 2001, contains detailed process data for approximately 600 unit processes within the United States. However, to calculate the lifecycle emissions for materials, energy, and fuels, additional steps such as matrix inversion are required. In contrast, the GREET model, developed as a spreadsheet-type model by the Argonne Laboratory, provides unit process data and lifecycle emission information for materials and energy within the United States and select international regions. The Ecoinvent database, originating in Switzerland, is the most comprehensive of the three, and includes unit process data and lifecycle emission/impact data for over 18,000 processes, covering a substantial portion of the world.

The two recent studies (Chen, Blanc-Betes et al., 2021; Kim et al., 2023a) indicate that the global warming intensity (GWI, well-to-wheel GHG emissions) of ethanol produced from switchgrass varies between −9.4 and 1.7 gCO_2 MJ^{-1}, influenced by factors such as technologies used, spatial and temporal boundaries, and GHG emission factors. In those studies, changes in soil organic carbon (SOC)

TABLE 1 Life cycle assessment (LCA) databases used in literature from the DOE-funded Bioenergy Research Centers (BRCs).

BRCs	System	Database	References
CABBI	Ethanol derived from sugarcane	USLCI, GREET, Ecoinvent, FORWAST	Shi and Guest (2020)
	Corn ethanol	GREET	Khanna et al. (2020)
	Bioenergy derived from miscanthus and switchgrass	GREET	Chen, Blanc-Betes et al. (2021)
	Corn ethanol and cellulosic ethanol	GREET	Chen, Debnath et al. (2021)
	Ethanol and biodiesel from sweet sorghum and sugarcane	GREET, Ecoinvent	Cortés-Peña et al. (2022)
CBI	Ethanol derived from switchgrass	GREET	Field et al. (2020)
	Reductive catalytic fractionation oil from hybrid poplar	DATASMART life cycle inventory database (based on a combination of US LCI)	Bartling et al. (2021)
GLBRC	Ethanol derived from corn stover	USLCI, GREET, Ecoinvent	Kim et al. (2019)
	Ethanol derived from corn stover and switchgrass	USLCI, GREET, Ecoinvent	Kim et al. (2020)
	Chemicals derived from corn stover	GREET	Huang et al. (2021)
	Ethanol derived from switchgrass	GREET	Martinez-Feria et al. (2022)
	Ethanol derived from switchgrass	USLCI, GREET, Ecoinvent	Kim et al. (2023a)
	Ethanol derived from switchgrass	USLCI, GREET, Ecoinvent	Kim (2023c)
JBEI	Ethanol derived from corn stover	USLCI, GREET, Ecoinvent	Neupane et al. (2017)
	Jet fuel blendstock derived from biomass sorghum	USLCI, GREET, Ecoinvent	Baral et al. (2019)
	Biomass sorghum	USLCI, GREET, Ecoinvent	Baral et al. (2020)
	Ethanol derived from biomass sorghum	USLCI, GREET, Ecoinvent	Yang et al. (2020)
	Renewable natural gas derived from organic municipal solid waste	GREET, Ecoinvent	Nordahl et al. (2020)
	Jet fuel blendstock derived from biomass sorghum	USLCI, GREET, Ecoinvent	Baral et al. (2021)

and excess electricity emerge as significant contributors to GHG emissions in switchgrass-based ethanol production (Chen, Blanc-Betes et al., 2021; Kim et al., 2023a). The discrepancies in SOC changes between the two studies can be attributed to variations in biogeochemical models, spatial and temporal boundaries, simulation resolutions, among others. Similarly, the primary contrast in GHG credits associated with excess electricity stems from the use of different GHG emission factors: 0.598 $kgCO_2\ kWh^{-1}$ in the study by Kim et al. (2023a) compared with 0.549 $kgCO_2\ kWh^{-1}$ in the work of Chen, Blanc-Betes et al. (2021), resulting in about 28–38% differences in GHG credit of excess electricity. Therefore, the GHG emission factor is highlighted as a key source of uncertainty in the biofuel system.

To investigate the uncertainties associated with GHG emission factors in the biofuel system, we revisited our previous study estimating the GWI of switchgrass-based biofuel (ethanol) in Michigan (Kim et al., 2023a). We then recalculated the GWI and GHG mitigation potential over gasoline, referred to as a "static calculation," using a functional unit of 1 MJ of ethanol fuel based on lower heating value. This process used GHG emission factors from all three LCA databases, with multiple versions of each database. We also conducted Monte Carlo simulations using distribution functions derived from different databases to assess the uncertainties associated with GHG emission factors. Furthermore, we performed both LSA and GSA for GHG emission factors and input parameters to identify those parameters most affecting the GWI.

2 | MATERIALS AND METHODS

2.1 | Switchgrass-based biofuel production system

In our previous study (Kim et al., 2023a), we used switchgrass grown in Michigan as cellulosic feedstock for ethanol production. The potential yield of switchgrass and SOC changes during switchgrass production were estimated using the System Approach to Land Use Sustainability (SALUS) model (Basso & Ritchie, 2015). Agronomic inputs and fuel use were determined based on field experiment data from the GLBRC in Michigan. Only the fertilized biomass production scenario is considered in this analysis. Comprehensive details are available on Dryad (Kim et al., 2023b).

Switchgrass is converted into ethanol in a centralized biorefinery. In the analysis, two different hydrolysis technologies are considered: chemical hydrolysis and enzymatic hydrolysis. The chemical hydrolysis technology (referred to as GVL) involves the utilization of a mixture of γ-valerolactone (GVL), water, and toluene, with the addition of dilute sulfuric acid as a catalyst. This mixture is employed to facilitate the hydrolysis of cellulose and hemicellulose, ultimately converting them into fermentable sugars (Won et al., 2017). In contrast, the enzymatic hydrolysis technology (referred to as ACID) begins with a dilute acid pretreatment stage and proceeds to the enzymatic conversion of cellulose and hemicellulose into fermentable sugars (Tao et al., 2014). The analyses (including static calculations, Monte Carlo simulation, and sensitivity analysis) are carried out for ethanol production using each of these hydrolysis technologies. A comprehensive summary of materials, transportation modes, and fuels used in the switchgrass-based ethanol production system is presented in Table 2. Materials displayed in italics in Table 2 were excluded from the GHG emission calculations due to their small quantities or unavailable GHG emission factors.

2.2 | Scenarios in static calculations

We calculated the GWI and GHG mitigation potential of switchgrass-based ethanol by scrutinizing a total of 29 distinct scenarios (as detailed in Table 3). These scenarios include four variations of GHG emission factors based on the unit processes in the USLCI database, the GREET model, and the Ecoinvent database (GLBRC scenario). Additionally, there are seven versions of the GREET model ranging from 2016 to 2022 (GREET scenario), and six versions of the Ecoinvent database (Ecoinvent scenario). The Ecoinvent scenarios not only include different versions (ranging from 3.2 to 3.7) but also cover three distinct geographical regions: the United States, Europe, and the rest of the world.

It is noteworthy that none of the three LCA databases provided a comprehensive set of GHG emission factors for the switchgrass-based biofuel system, including materials, transportation modes, and fuels. To address this gap, we supplemented the missing emission factors by either incorporating average values from different databases or average values from other regions within the same version of the respective database.

In the GLBRC scenarios, unit process data for energy production, encompassing petroleum, nuclear, natural gas, coal production, and power plants, are sourced from the USLCI database. For materials and transportation modes, the most relevant unit process data from three databases are selected based on their relevance in terms of temporal or geographical boundaries. The fuel mixes for US electricity are derived from the US Energy Information

TABLE 2 Materials, transportation modes, and fuels in the switchgrass-based biofuel production system.

Lifecycle phase	Materials, transportation modes, and fuels
Switchgrass production	Nitrogen fertilizer [ammonia, urea, urea ammonium nitrate (UAN), ammonium nitrate, ammonium sulfate, diammonium phosphate (DAP), monoammonium phosphate (MAP)], phosphorus fertilizer, potassium fertilizer, diesel, herbicides, grid electricity
Transportation and storage	Truck transport, high-density polyethylene (HDPE), diesel
Biorefinery	
GVL	Toluene, calcium hydroxide, yeast, sulfuric acid, grid electricity, *RuSn4/C catalyst, wastewater treatment nutrients*
ACID	Sulfuric acid, calcium hydroxide, sulfur dioxide, ammonia, corn steep liquor, glucose, diammonium phosphate, sodium hydroxide, grid electricity, *sorbitol, host nutrients, polymer, boiler water chemicals, cooling tower chemicals, antifoam agent*
Others	Truck transport, railroad transport, barge transport, air transport

TABLE 3 Scenarios in static calculation.

Scenario	LCA database(s)[a]	GHG emission factor of electricity[b]	Geographical boundary[c]
GLBRC_16	Combination of USLCI, GREET, Ecoinvent based on 2016 US electrical fuel mix	US electricity in 2016	United States
GLBRC_22	Combination of USLCI, GREET, Ecoinvent based on 2022 US electrical fuel mix	US electricity in 2022	United States
GLBRC_eco3.71	Combination of USLCI, GREET, Ecoinvent based on US electrical fuel mix in Ecoinvent 3.71	US electricity in Ecoinvent 3.71	United States
GLBRC_GREET22	Combination of USLCI, GREET, Ecoinvent on US electrical fuel mix in GREET 2022	US electricity in GREET 2022	United States
GREET_22	GREET 2022	US electricity in GREET 2022	United States
GREET_21	GREET 2021	US electricity in GREET 2021	United States
GREET_20	GREET 2020	US electricity in GREET 2020	United States
GREET_19	GREET 2019	US electricity in GREET 2019	United States
GREET_18	GREET 2018	US electricity in GREET 2018	United States
GREET_17	GREET 2017	US electricity in GREET 2017	United States
GREET_16	GREET 2016	US electricity in GREET 2016	United States
Ecoinvent_3.71_US	Ecoinvent 3.71	US electricity in Ecoinvent 3.71	United States
Ecoinvent_3.71_EU	Ecoinvent 3.71	US electricity in Ecoinvent 3.71	Europe
Ecoinvent_3.71_ROW	Ecoinvent 3.71	US electricity in Ecoinvent 3.71	The rest of the world
Ecoinvent_3.6_US	Ecoinvent 36	US electricity in Ecoinvent 3.6	United States
Ecoinvent_3.6_EU	Ecoinvent 3.6	US electricity in Ecoinvent 3.6	Europe
Ecoinvent_3.6_ROW	Ecoinvent 3.6	US electricity in Ecoinvent 3.6	The rest of the world
ECOINVENT_3.5_US	Ecoinvent 3.5	US electricity in Ecoinvent 3.5	United States
ECOINVENT_3.5_EU	Ecoinvent 3.5	US electricity in Ecoinvent 3.5	Europe
ECOINVENT_3.5_ROW	Ecoinvent 3.5	US electricity in Ecoinvent 3.5	The rest of the world
ECOINVENT_3.4_US	Ecoinvent 3.4	US electricity in Ecoinvent 3.4	United States
ECOINVENT_3.4_EU	Ecoinvent 3.4	US electricity in Ecoinvent 3.4	Europe
ECOINVENT_3.4_ROW	Ecoinvent 3.4	US electricity in Ecoinvent 3.4	The rest of the world
ECOINVENT_3.3_US	Ecoinvent 3.3	US electricity in Ecoinvent 3.3	United States
ECOINVENT_3.3_EU	Ecoinvent 3.3	US electricity in Ecoinvent 3.3	Europe
ECOINVENT_3.3_ROW	Ecoinvent 3.3	US electricity in Ecoinvent 3.3	The rest of the world
ECOINVENT_3.2_US	Ecoinvent 3.2	US electricity in Ecoinvent 3.2	United States
ECOINVENT_3.2_EU	Ecoinvent 3.2	US electricity in Ecoinvent 3.2	Europe
ECOINVENT_3.2_ROW	Ecoinvent 3.2	US electricity in Ecoinvent 3.2	The rest of the world

[a]Life cycle assessment (LCA) databases used in each scenario.

[b]GHG emissions of electricity.

[c]Geographical boundaries for major processes.

Administration (2022). GHG emission factors for energy, materials, and transportation modes are then calculated via matrix inversion (Kim et al., 2023a). Each GLBRC scenario is characterized by specific US electricity fuel mixes, allowing us to estimate electricity fuel mix-specific GHG emissions for the materials and fuels used in the biofuel production system under investigation (see Table S1). For example, the GHG emission factors in the GLBRC_16 scenario, which were used in our prior study, are derived from the 2016 US electricity fuel mixes. In contrast, the GLBRC_22 scenario uses the 2022 US electricity fuel mixes as its basis. In the GREET scenarios, indirect GHG emissions from carbon monoxide and volatile organic compounds are not considered as GHG emissions for consistency. On the contrary, in the Ecoinvent scenarios, we considered GHG emission factors from the two regions outside of the United States, but the GHG emission factor of US electricity production from the same version is employed in those scenarios.

The GHG emission factors in each scenario are presented in Figure 1 (also see Table S2). The GHG emission factors in Figure 1 include GHG emissions from cradle to the user's gate, including the transportation of the final product. Additionally, the GHG emissions associated with electricity consider transmission and distribution losses. While certain emission factors across the databases are similar, others exhibit notable differences. These variations in emission factors can be attributed to factors such as temporal and geographical boundaries, technologies employed, or allocation methods. For instance, the life cycle energy consumption for herbicide production in the Ecoinvent database is approximately 60–70% of that in the GREET model. Differences in GHG emission factors for diammonium phosphate (DAP) as a nitrogen source among scenarios primarily result from allocation factors, particularly nutrient ratios. While the GHG emission factors for 1 kg of DAP are comparable across scenarios, the quantities of DAP required for 1 kg of nitrogen nutrient differ significantly between scenarios. In most scenarios, except for the Ecoinvent_3.71 scenarios, 1 kg of nitrogen nutrient requires approximately 1.7–3.0 kg of DAP. In contrast, in the Ecoinvent_3.71 scenarios, these quantities increase to 5.6 kg for DAP.

2.3 | Uncertainty and sensitivity analyses

In this study, we used Oracle Crystal Ball and MATLAB as the primary computational platforms for conducting Monte Carlo simulations and sensitivity analyses. The Monte Carlo simulations involved 10,000 iterations and were executed in two distinct phases, designed to comprehensively address both the uncertainty associated with GHG emission factors and the sensitivity of the model to variations in GHG emission factors and input parameters (summarized in Table S3). In the first Monte Carlo simulation, GHG emission factors are treated as random variables, thereby quantifying the associated uncertainty, while the input parameters are held constant. The second simulation extends this approach by including both GHG emission factors and input parameters as random variables, thereby capturing the sensitivity of the results to each variable.

The distribution functions for GHG emission factors were assumed to follow a lognormal distribution. These distributions were derived from average values and standard deviations obtained from a dataset consisting of 29 different scenarios. To address input parameter

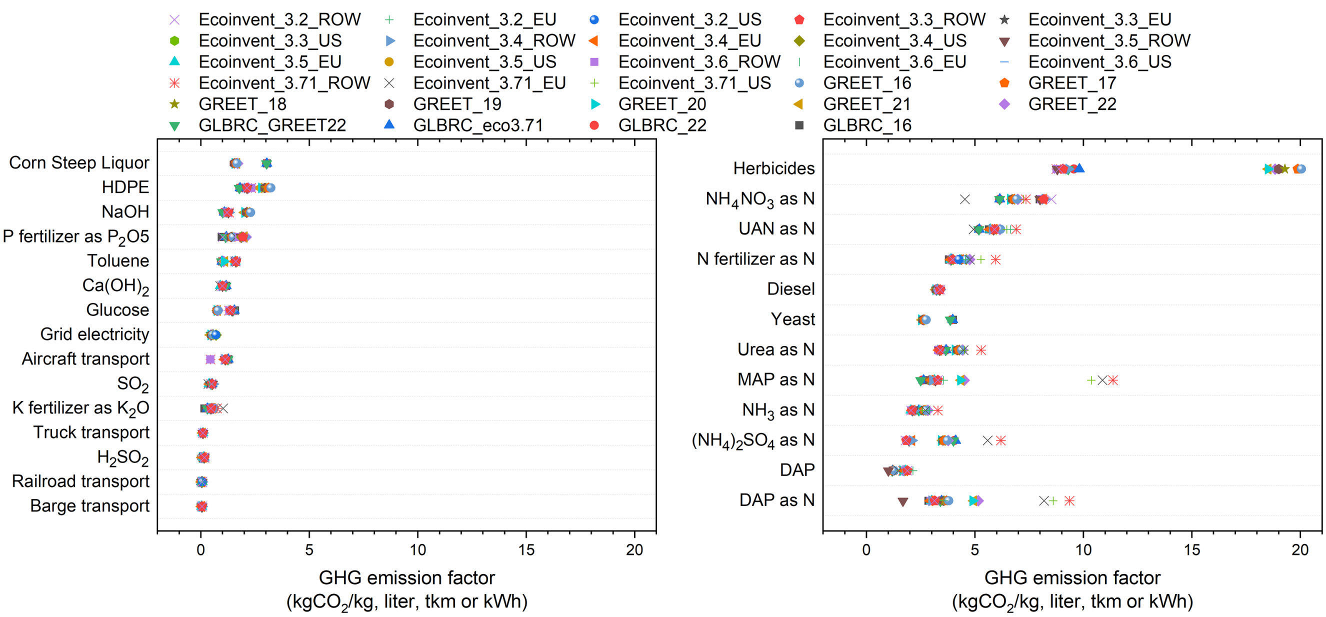

FIGURE 1 Greenhouse gas (GHG) emission factors in different scenarios (note that the physical properties of fuels (e.g., lower heating value, and density) used in the calculations are based on the GREET model.).

uncertainty, we assumed a 10% standard deviation, considering the limited availability of specific data. This 10% standard deviation was used in estimating the lognormal distribution, influencing the shape and spread of the distribution curve.

The major components of switchgrass, including cellulose, hemicellulose and lignin, exhibited differences between the lower/upper values and the median values ranging from 5% to 13% of the median values (Boundy et al., 2011). Additionally, SALUS simulation results indicated that the standard deviations of average switchgrass yields across planting cycles are less than 11% of the average yield. This approach not only incorporates variability and uncertainty in the input parameters but also ensures consistency in scale with the provided values. However, some specific input parameters (e.g., biofuel yield, biomass yield, and SOC change) were modeled using a triangular distribution to prevent unrealistically extreme values that do not align with real-world conditions. Additionally, a uniform distribution was applied to the equivalency factor, which serves as a conversion factor from nitrogen-equivalent mass to kilograms of DAP, due to value choice. The results of random sampling from the Monte Carlo simulations for each GHG emission factor and input parameter are presented in the Figures S1 and S2.

The sensitivity analysis in this study includes both local and global sensitivity analyses. Local sensitivity analysis relies on partial derivatives to assess the impact of small perturbations around default input values on the model's results. In this context, local sensitivity indices are defined as normalized partial derivatives, providing insights into the parameters that have the most direct influence on the model output (Wolf et al., 2017). On the contrary, GSA is based on variance decomposition and employs two types of Sobol sensitivity indices (Groen et al., 2017; Wei et al., 2015): first-order Sobol indices, which evaluate the direct impact of each input parameter on the model output, and total Sobol indices, which provide a broader perspective by measuring the overall effect of each input parameter, including both direct and indirect impacts. Equations for the sensitivity indices are provided in the Supplementary Material.

It is important to note that the sensitivity analyses do not incorporate excess electricity exported from the biorefinery as an input parameter due to the inverse relationship between excess electricity generation and biofuel yield (see Figure S3). Higher yields reduce the availability of carbon for use as fuel in the cogeneration facility, resulting in decreased energy generation. Furthermore, high biofuel yields result in more energy required for biofuel purification than do low yields.

Other possible correlations are seen in switchgrass production: biomass yield versus nitrogen fertilizer rate and biomass yield (or nitrogen fertilizer rate) versus SOC changes. Experimental data from Michigan (Martinez-Feria & Basso, 2020) show a modest increase in biomass yield with higher nitrogen fertilizer application, at a rate of approximately 12 kg of additional dry biomass per hectare for each extra kg of nitrogen applied (see Figure S4). Even though the biomass yield increase attributable to nitrogen fertilizer is relatively small considering the nitrogen fertilizer rate used in this analysis (50 kgN ha^{-1}), this correlation is accounted for in the sensitivity analyses.

In contrast, correlations related to SOC changes are not considered in the sensitivity analyses due to the multitude of factors influencing SOC changes, including initial carbon stock, soil characteristics, the percentage of biomass removal, climatic conditions, nitrogen fertilizer rate, and others. Consequently, SOC changes are treated as independent parameters in the sensitivity analyses.

3 | RESULTS

The mean GWIs of switchgrass-based ethanol across the scenarios in the static calculations are −9.8 (±2.7) gCO_2 MJ^{-1} for the GVL technology and −6.1 (±2.1) gCO_2 MJ^{-1} for the ACID technology, aligning closely with those reported in our previous study (Kim et al., 2023a). Within the scenarios, GWIs for the GVL technology range from −14.2 to −4.8 gCO_2 MJ^{-1}, while for the ACID technology, they vary from −8.2 to −0.6 gCO_2 MJ^{-1}. The GHG mitigation potentials across the scenarios vary from 1.18 to 1.30 $TgCO_2$ $year^{-1}$ for the GVL technology and from 1.17 to 1.27 $TgCO_2$ $year^{-1}$ for the ACID technology at an average of 1.24 $TgCO_2$ $year^{-1}$ for both technologies (see Table S5).

In both the GLBRC and GREET scenarios, the GWI and GHG mitigation potentials associated with the GVL technology exhibit a linear correlation with the GHG emission factor of electricity, as depicted in Figures 2 and 3. This linear relationship results from the role of electricity as the primary contributor to GHG emissions in biofuel production. Moreover, the GHG emission factors of major contributors (see Figure S5) in the GLBRC scenarios also demonstrate a linear correlation with the GHG emission factor of electricity (see Table S7). However, discrepancies in the ACID technology in the GREET scenarios arise from variations in process data for specific major contributors across different model versions, possibly reflecting technological advancements or improvements and leading to observed deviations from linearity.

While electricity remains the dominant source of GHG emissions in the Ecoinvent scenarios, the GHG emission factors of major contributors do not align with the GHG emission factor of electricity. This discrepancy arises from

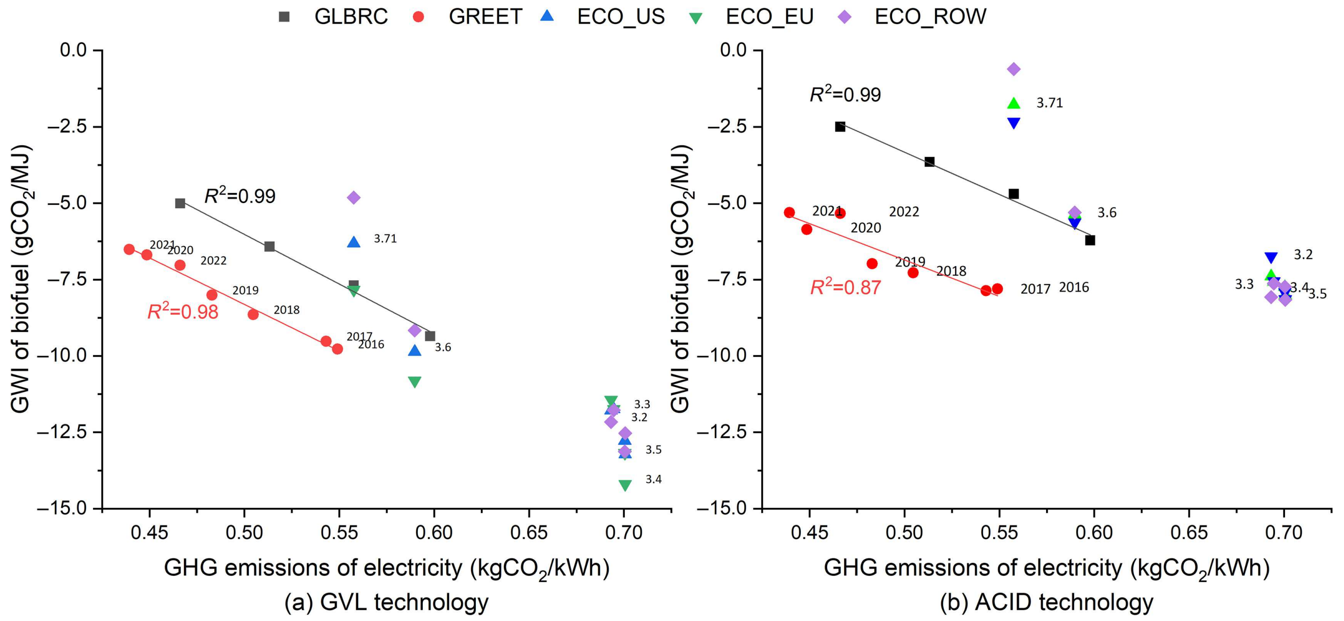

FIGURE 2 Correlation between global warming intensity (GWI) and greenhouse gas (GHG) emissions of electricity ((a) GVL technology; (b) ACID technology).

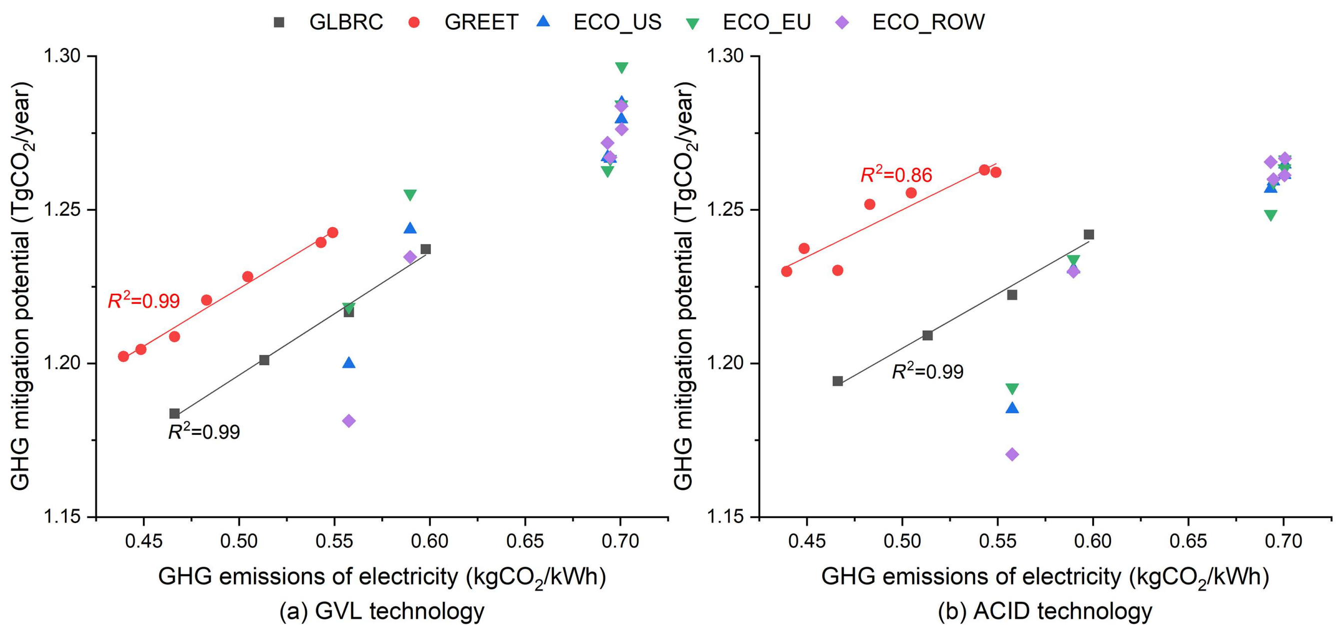

FIGURE 3 Correlation between greenhouse gas (GHG) mitigation potential and GHG emissions of electricity ((a) GVL technology; (b) ACID technology).

the use of US electricity's GHG factors in two regions (Europe and the rest of the world) and the application of average emission factors for some materials and fuels. The electricity mixes for other regions in the Ecoinvent scenario comprise a combination of fuel mixes from different countries, resulting in different GHG emission factors for electricity compared with those from the United States.

The first Monte Carlo simulations yield a mean GWI of −8.8 (±3.4) $gCO_2\ MJ^{-1}$ for the GVL technology and of −6.3 (±3.7) $gCO_2\ MJ^{-1}$ for the ACID technology, as presented in Figure 4 (also see Table S8). The GWI data for the GVL technology exhibit a negatively skewed distribution, with a tail toward lower values, indicating a higher likelihood of values lower than the mean. Conversely, the GWI data for the ACID technology are nearly symmetrical, suggesting a balanced distribution without significant skewness in either direction. The data distributions exhibit a slight kurtosis, indicating a higher concentration

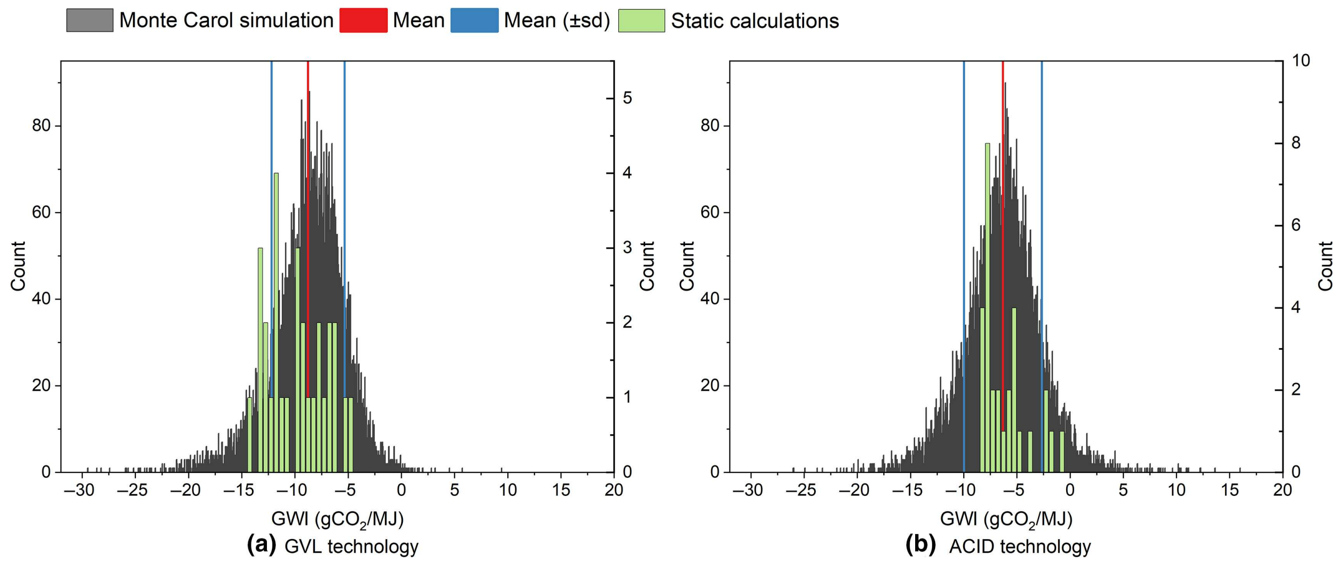

FIGURE 4 Distributions for global warming intensity (GWI) in the first simulation ((a) GVL technology; (b) ACID technology; sd: standard deviation).

of values around the mean compared with the tails of a normal distribution.

Monte Carlo simulations for GHG mitigation potentials reveal a mean of 1.23 (±0.04) $TgCO_2$ $year^{-1}$ with a slightly positively skewed distribution for the GVL technology and 1.24 (±0.05) $TgCO_2$ $year^{-1}$ with a slightly negatively skewed distribution for the ACID technology, as illustrated in Figure 5. The coefficient of variations for both technologies is less than 0.04, indicating moderate variability relative to the mean.

Consistently, Monte Carlo simulations demonstrate means close to those obtained from static calculations, suggesting that static calculations provide reasonable approximations of the central tendency of the data. However, Monte Carlo simulations offer additional insights into the data distribution, including skewness and kurtosis, which are not readily available from static calculations.

In the second simulation, in which input parameters are introduced as random variables, the GWI distribution for both GVL and ACID technologies broadens significantly compared with the first simulation (see Figure S6). This expanded range results from the combined influence of GHG emission factor uncertainties and variations in input parameters. Importantly, despite the broader distributions, the mean GWIs for both technologies exhibit minimal changes compared with the first simulation (see Table S9). This consistency suggests that the central tendency of the GWI data remains similar even with the added complexity of input parameter variations. However, notable increases in standard deviations and variances for both technologies highlight a wider range of potential GWI values, underscoring the profound influence of input parameters on overall GWI uncertainty. These findings emphasize the importance of not only accounting for uncertainties in GHG emission factors but also recognizing the influence of input parameters when assessing the GWI of different ethanol technologies.

The LSA indicates that the most important parameter influencing the GWI of biofuel in both GVL and ACID technologies is biofuel yield, followed by SOC change and GHG emissions of electricity (see Figure 6). Small perturbations in biofuel yield directly affect both the excess electricity produced and the amount of biomass feedstock consumed during biofuel production, consequently impacting the GHG contribution from the feedstock. On the contrary, the GSA results demonstrate that the GWI of biofuel in both technologies is most sensitive to GHG emissions of electricity, followed by biofuel yield, SOC change, and nitrogen fertilizer rate (see Figure 6).

There are no significant differences in the relative importance of parameters between the first-order Sobol and the total Sobol indices (see Figure 6). This suggests that interactions between input parameters are weak or not potent enough to significantly influence overall sensitivity analysis outcomes. The effects of the parameters on the output results appear independent, indicating that the value of any one parameter does not substantially affect the impact of any other parameter. Although each additional kilogram of nitrogen applied can increase yield by approximately 12 kg of dry biomass per hectare, the interaction of nitrogen fertilizer application rate to biomass yield does not alter the relative importance of parameters in both technologies.

Parameters associated with $Ca(OH)_2$ in the GVL technology and parameters associated with corn steep liquor

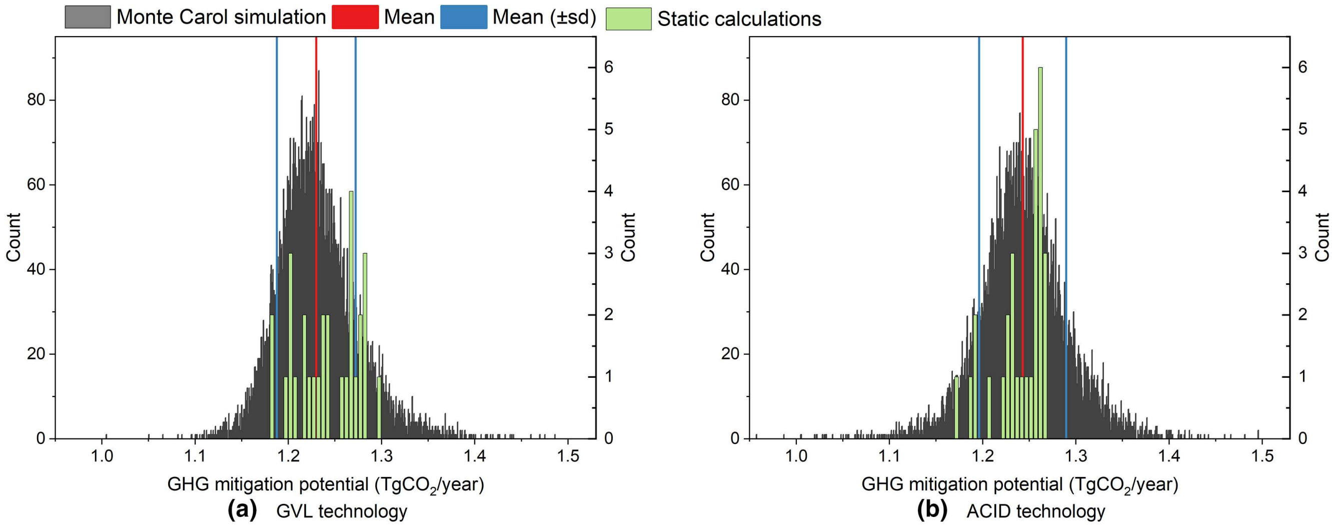

FIGURE 5 Distributions for greenhouse gas (GHG) mitigation potential in the first simulation ((a) GVL technology; (b) ACID technology; sd: standard deviation).

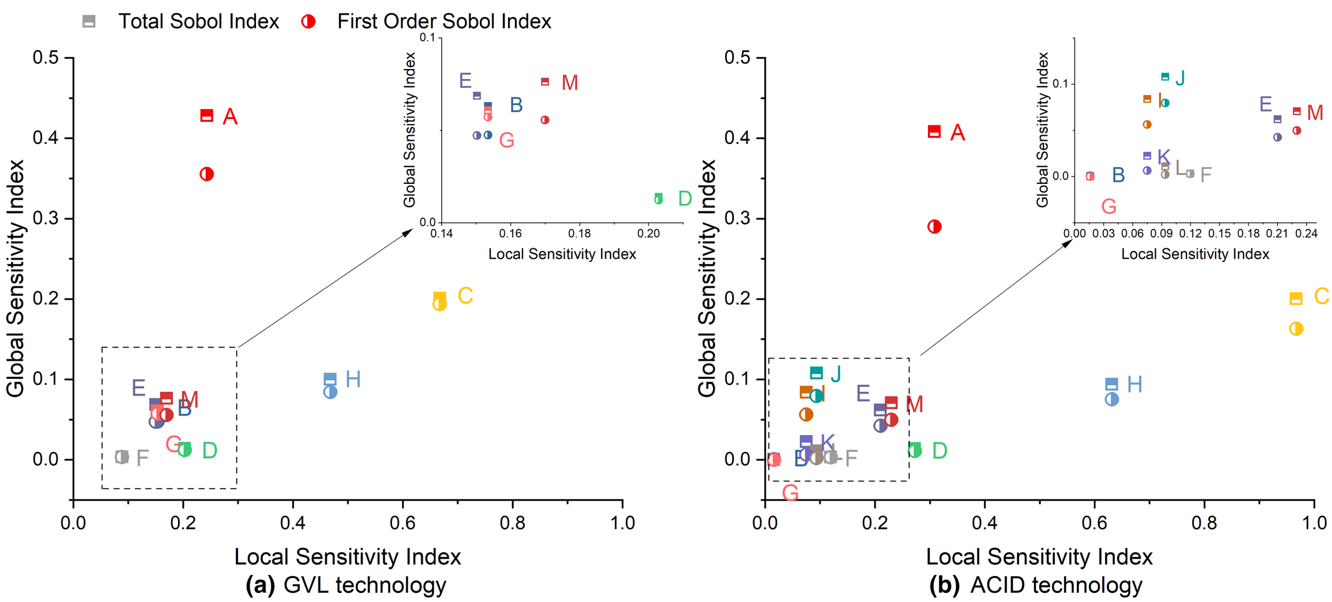

FIGURE 6 Sensitivity indices ((a) GVL technology; (b) ACID technology; (A) greenhouse gas (GHG) of electricity; (B) GHG of $Ca(OH)_2$; (C) biofuel yield; (D) biomass yield; (E) application rate of nitrogen fertilizer with no biomass yield-nitrogen fertilizer rate correlation; (F) N_2O rate from N fertilizer; (G) amount of $Ca(OH)_2$; (H) soil organic carbon (SOC) change; (I) GHG of corn steep liquor; (J) GHG of glucose; (K) amount of corn steep liquor; (L) amount of glucose; (M) application rate of nitrogen fertilizer with biomass yield-nitrogen fertilizer rate correlation).

and glucose in the ACID technology are important in both sensitivity analyses. In the GVL technology, GHG emissions associated with $Ca(OH)_2$ are significant, exceeding 9.7 $gCO_2\ MJ^{-1}$. Despite the relatively narrow distribution for GHG emissions of $Ca(OH)_2$ production (see Figure S1), its large contribution to GWI renders these parameters important.

GHG emissions associated with corn steep liquor and glucose in the ACID technology system are less important. However, the GHG emission factors for corn steep liquor in the GLBRC scenarios are over 1.8-fold larger than those in the GREET scenarios due to GHG emissions from corn grain used as a raw material for corn steep liquor production. The GLBRC scenarios comprehensively consider GHG emissions associated with changes in SOC during corn production, a factor not considered in the GREET scenarios. This comprehensive consideration results in larger GHG emissions for corn-derived materials, such as glucose and corn steep liquor, contributing to the observed wider ranges.

4 | DISCUSSION

This study highlights the importance of a comprehensive approach to assess the GWI of biofuel systems by considering a diverse range of GHG emission factors and databases. Utilizing multiple LCA databases, including USLCI, GREET, and Ecoinvent, each with various versions of the databases, provided a more comprehensive understanding of the potential variations in GHG emission factors and their impact on GWI estimations. The results underscore the sensitivity of GWIs and GHG mitigation potentials to the choice of LCA database and emission factors, leading to substantial variations across scenarios. This variability therefore also highlights the need for cautious interpretation of LCA results and emphasizes the significance of relying on comprehensive and up-to-date data sources.

The convergence of findings between our previous study (Kim et al., 2023a), static calculations, and Monte Carlo simulations adds credibility to the estimated GWI values. The static calculations and Monte Carlo simulations yielded similar mean GWIs, suggesting that static calculations can provide reasonable approximations of the central tendency of the data. This convergence strengthens confidence in the estimated GWI values and provides a basis for further discussions and policy decisions. Despite the convergence observed, the wider range of GWI values obtained from the simulations emphasizes the necessity of incorporating potential variations and uncertainties into real-world applications, reinforcing the practical implications of our findings. The simulations capture the inherent variability associated with input parameters and demonstrate that the actual GWI could deviate from the central tendency due to these variations.

Sensitivity analysis shows that biofuel yield, GHG emissions of electricity, and SOC change have the greatest impact on the GWI of biofuel. Thus, efforts to optimize these factors can substantially reduce the uncertainty associated with GWI. Biofuel yield is crucial as it directly influences the amount of biomass feedstock consumed and the excess electricity produced during biofuel production. GHG emissions of electricity are particularly important due to the substantial contribution of electricity to the overall GWI. Furthermore, the impact of SOC change on the carbon balance of the biofuel system highlights its significance in shaping the overall environmental footprint. Prioritizing these factors in research and policymaking endeavors is crucial for refining GWI assessments and making well-informed decisions regarding sustainable biofuel production strategies.

The accuracy of GHG emissions from electricity production is crucial for assessing the GWI of biofuels, given its substantial contribution to overall GHG emissions. Electricity fuel mixes play an important role in these emissions, influencing the assessment's complexity. Despite the inherently future-oriented nature of cellulosic ethanol systems, most LCA studies have predominantly relied on historical or existing electricity fuel mixes. The uncertainty surrounding future electricity fuel mixes, influenced by factors such as technological advancements, policy changes, and resource availability, introduces additional complexity to this assessment. Considering that cellulosic ethanol systems are not yet widely deployed, accurately projecting future electricity fuel mixes becomes a challenging task, encompassing diverse variables such as technological evolution, policy dynamics, and resource constraints.

Despite the challenges in determining future electricity fuel mixes, it is imperative to acknowledge potential variations and uncertainties when evaluating the GWI of biofuels. Sensitivity analysis emerges as a valuable tool, offering insights into the impact of these uncertainties on GWI, thereby facilitating a more comprehensive understanding of the potential range of GHG emissions linked to biofuel production. Future research should continue to refine methodologies for projecting future electricity fuel mixes and incorporate these projections into LCA studies to enhance the accuracy and robustness of GWI assessments in the context of evolving cellulosic biofuel systems.

This endeavor is not merely academic; it has profound implications for climate change mitigation globally. By accurately quantifying the GWI of biofuels, stakeholders (including researchers, policymakers, and industry leaders) gain crucial insights that can guide sustainable energy policies, investment decisions, and technological innovations. Moreover, contextualizing the impact of these assessments within broader climate action agendas underscores the critical role of sustainable biofuel production in addressing environmental, economic, and social challenges. Through this comprehensive approach, biofuel systems are recognized not only for their potential to mitigate climate change but also for their contributions to job creation, rural development, and energy security, thereby supporting a multifaceted strategy toward a more sustainable and resilient energy future.

AUTHOR CONTRIBUTIONS

Seungdo Kim: Conceptualization; data curation; formal analysis; investigation; methodology; visualization; writing—original draft; writing—review and editing. Bruce E. Dale: Conceptualization; writing—review and editing. Bruno Basso: Data curation; investigation; writing—review and editing.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

DATA AVAILABILITY STATEMENT

The results of the Monte Carlo simulations and the distributions for GHG emission factors are available in Dryad (https://doi.org/10.5061/dryad.rn8pk0pm8).

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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