ACS ES&T Air

pubs.acs.org/estair

Article

Biochemical Conversion of Herbaceous Biomass to Renewable Diesel: Net Greenhouse Gas and Air Pollutant Trade-offs

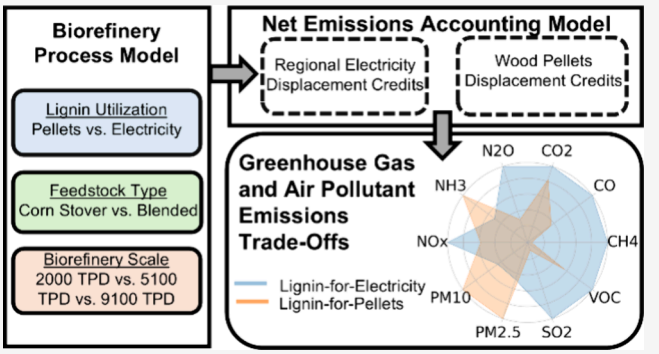
David Quiroz, Vikram Ravi,* Yimin Zhang, Arpit Bhatt, and Garvin Heath

Cite This: *ACS EST Air* 2025, 2, 175−186

Read Online

ACCESS | Metrics & More | Article Recommendations | Supporting Information

ABSTRACT: This study examines greenhouse gas (GHG) and criteria air pollutant (CAP) emissions trade-offs for renewable diesel across 12 scenarios, involving different biochemical conversion designs, biorefinery scales, and feedstocks. A conventional design uses lignin for on-site heat and power, which exports excess power to the grid. An alternative design exports lignin pellets, offsetting other pellet production methods but requiring grid electricity to meet biorefinery power demands. Net emissions were quantified in Iowa and Georgia, selected considering feedstock availability, coproduct displacement, and regional power grids, assuming grid-exported power avoids coal or low-carbon electricity. Results for the conventional design remained consistent across the electricity displacement scenarios. When comparing lignin utilization strategies, pelletizing lignin reduces sulfur dioxide, carbon monoxide, nitrogen oxides, and volatile organic compounds (net emissions −0.66 mg MJ^{-1}, 25 mg MJ^{-1}, 25 mg MJ^{-1}, 7.8 mg MJ^{-1}, respectively). However, lignin pelletization increases net particulate matter (fine and coarse) and ammonia (net emissions of 4.7 mg MJ^{-1}, 13 mg MJ^{-1}, and 0.26 mg MJ^{-1}, respectively), alongside indirect GHG emissions due to grid electricity dependence. Additionally, processing 2000 tonnes corn stover daily minimizes emissions for both designs. Only lignin pelletization with renewable electricity and additional particulate matter and ammonia controls reduces all CAP and GHG emissions simultaneously.



KEYWORDS: *biorefinery, lignin, greenhouse gas protocol, emissions, air quality, renewable fuels*

1. INTRODUCTION

To reduce transportation sector greenhouse gas (GHG) emissions, governments have implemented policies aiming to eliminate fossil fuel consumption through adoption of clean renewable fuels.[1,2] For instance, California's Low Carbon Fuel Standard targets a 20% reduction in the carbon intensity of transportation fuels by 2030 relative to 2010 levels.[3] As a consequence, and given market momentum for electrification of the light duty vehicle fleet, the demand for advanced biofuels for aviation (jet fuel), marine (bunker fuel and diesel), rail (diesel), and heavy-duty vehicles (diesel) is expected to increase in the near future.[4]

Second-generation biofuels derived from agricultural wastes and forest residues can contribute to meeting these environmental targets. Renewable diesel derived from lignocellulosic biomass not only overcomes many limitations of conventional corn ethanol and soybean biodiesel, such as competition with food resources for land and intensive water use, but can also lower GHG emissions compared to conventional (first generation) biofuels and petroleum fuels.[5] Unlike ethanol, which as a gasoline substitute is limited to light-duty vehicles, renewable diesel is seen as a long-term alternative to decarbonize hard-to-electrify transportation modes such as heavy-duty trucking and off-road vehicles.[4] However, economic and technical challenges such as high biomass transportation costs and complex conversion processes required to convert lignocellulosic feedstocks into fuel products still limit the advancement of second-generation biorefineries.[5,6]

Nevertheless, some environmental concerns associated with production and use of lignocellulosic biofuels remain.[7] Environmental impacts of biofuel supply chains are often estimated using life-cycle assessment (LCA). LCA standards and practices provide an appropriate framework for quantifying the life-cycle environmental impacts of technologies and consistently comparing systems producing the same product based on multiple environmental impact categories.[8] Multiple studies have used LCA models to analyze the GHG emissions of biofuel pathways[9−12] with results varying often due to different modeling assumptions. Differences in methods such

Received: July 3, 2024
Revised: January 6, 2025
Accepted: January 7, 2025
Published: January 24, 2025

as system boundary selection, inclusion, or exclusion of land-use change emissions, model structure and input parameters, and coproducts allocation approaches hinder the comparability between LCA studies.[10]

More importantly, the scope of these analyses is often limited to GHG emissions, and the impacts of other critical environmental metrics such as criteria air pollutants (CAPs) are typically overlooked.[7,13] Ground-level ozone, particulate matter (PM), carbon monoxide (CO), sulfur dioxide (SO_2), nitrogen dioxide (NO_2), and lead, the primary CAPs, can cause respiratory issues, exacerbate existing health conditions, and even lead to premature mortality. Unlike GHG emissions, which contribute to long-term climate change, CAPs have more immediate and localized effects. When comparing their monetized impacts, the marginal damage costs of CAPs often exceed those of GHGs by orders of magnitude.[14]

This scarcity of air pollutants analysis in the biofuels LCA literature can be explained because a rigorous investigation of the CAP emissions of biofuel pathways requires detailed process and emission modeling for the biorefineries,[15] as well as data characterizing the emissions of CAPs from upstream processes in the supply chain.[13] Previous assessments have investigated spatial and temporal distributions of air pollutants from different biofuel supply chains across the United States (U.S.).[13,16−19] For example, Tessum et al.[13] constructed life cycle inventories of six air pollutants for corn ethanol and gasoline production. Hill et al.[16] compared the societal costs of GHG emissions and human exposure to fine particulate matter caused by production and use of corn ethanol, soybean biodiesel, and conventional fuels systems. A comprehensive study on the life cycle particulate matter and ozone emissions from various transportation technologies including, gasoline, diesel, corn ethanol, and electric vehicles was provided by Tessum et al.[17] The impact of increased corn ethanol use on the concentration of ozone and air toxics in the U.S. was quantified by Cook et al.[18] through air quality modeling. Similarly, Ravi et al.[19] leveraged life cycle inventory data with air quality modeling to analyze the air quality impacts of aviation fuel production from forest residues in the northwestern U.S.

Thorough, previous assessments tend to focus on corn grain ethanol and corn stover cellulosic ethanol,[13,17,18] which do not support the markets most in need of decarbonization solutions, like diesel for heavy-duty trucking. Also, prior studies that include assessment of air pollutant emissions usually do so without simultaneous assessment of GHG emissions,[13,16−19] thus not allowing for consideration of trade-offs of these two critical environmental dimensions. Additionally, published studies have not provided a detailed enough analysis considering biorefinery design and operations and, therefore, are not able to provide insights into how GHG and CAP emissions are affected by changes to the process design, biorefinery scale, or feedstock type. Understanding the impacts of these parameters is critical to identify process designs that minimize GHG and CAP emissions as well as to size and site biorefineries based on feedstock availability and compliance with potential air regulations.[20]

To fill in gaps in prior research, trade-offs between GHG and CAP emissions for a biorefinery producing a renewable diesel blend-stock (RDB) from two distinct biomass feedstocks (or blends), three process scales, and two lignin utilization strategies (generating a total of 12 design cases) are investigated. This work integrates best-available process-specific emission factors (EFs) (e.g., from Bhatt et al.[20]) with detailed process modeling to quantify GHG and CAP emission as well as credits associated with the displacement of other production pathways for electricity and solid fuels that result from the sale of these as coproducts of the biorefinery. Moreover, regional variability of emissions associated with displaced electricity and the impacts on coproduct credits were studied through two different case study locations. The analysis is not intended to estimate life-cycle emissions of the complete biofuel supply chain but instead aims to identify potential designs that co-optimally minimize GHG and CAP emissions from lignocellulosic biorefineries as well as to construct an emission inventory for future air quality modeling. The CAP emissions analysis explores potential emission credits of five criteria pollutants: nitrogen oxides (NO_x), SO_2, CO, and particulate matter with diameters of 10 μm (PM_{10}) and 2.5 μm or smaller ($PM_{2.5}$). Emissions of two precursors to the atmospheric formation of ozone and PM are also analyzed, namely, volatile organic compounds (VOCs), and ammonia (NH_3). Discussion focuses on comparing trade-offs of the 12 design cases in terms of GHG and CAP emissions.

2. METHODS

2.1. Biorefinery Process Model. The foundation of this analysis is a robust engineering process model that estimates material and energy flows necessary for quantifying the GHG and CAP emissions of the biorefinery. The model described in Davis et al.,[15] along with emission results reported by Bhatt et al.,[20] detailing a biochemical pathway for converting herbaceous biomass into hydrocarbons (RDB), serves as the foundational input to the GHG and CAP accounting model developed in this study. A total of 12 design cases were analyzed herein to assess the net emission outputs based on varying feedstocks, facility scales, and lignin applications. The hypothetical facility processes either corn stover or a uniform format blend (UFB) of cellulosic biomass feedstocks composed of 60% corn stover, 35% switchgrass, and 5% municipal solid waste.[15,20] Consistent with previous modeling work, the plant scales were varied to process 2000, 5100, and 9100 dry metric tons of feedstock/blends per day (dmtd). The 2000 dmtd scale was selected as it is a standard scale used in previous biorefinery design cases in the literature,[6,15] while the 5100 and 9100 dmtd scales correspond to the fifth and 10th percentile of existing petroleum refineries.[20]

One of the waste streams from biochemical conversion of cellulosic biomass is lignin solid cake that results from hydrolysate clarification. Two different uses of this lignin waste were explored, which define two important aspects of the design of the biorefinery. A lignin-for-electricity biorefinery uses lignin as a combustion fuel (in addition to off-gas, biogas, and sludge from wastewater treatment) to produce process heat and power. Based on the amount of lignin solid cake produced, more power can be generated than is demanded on-site, so excess (coproduct) electricity is sold to the electrical grid operator.

Alternatively, a lignin-for-pellets biorefinery was also explored, wherein the lignin solids are pelletized onsite and sold for off-site combustion as a solid fuel. In this design, biorefinery electricity demands are met by purchasing electricity from the grid. Also, the off-gas and biogas waste streams are combusted in a gas boiler, and the wastewater treatment sludge is transported for off-site treatment. This is an example of how the biorefinery is considered as a system,

https://doi.org/10.1021/acsestair.4c00152
ACS EST Air 2025, 2, 175−186

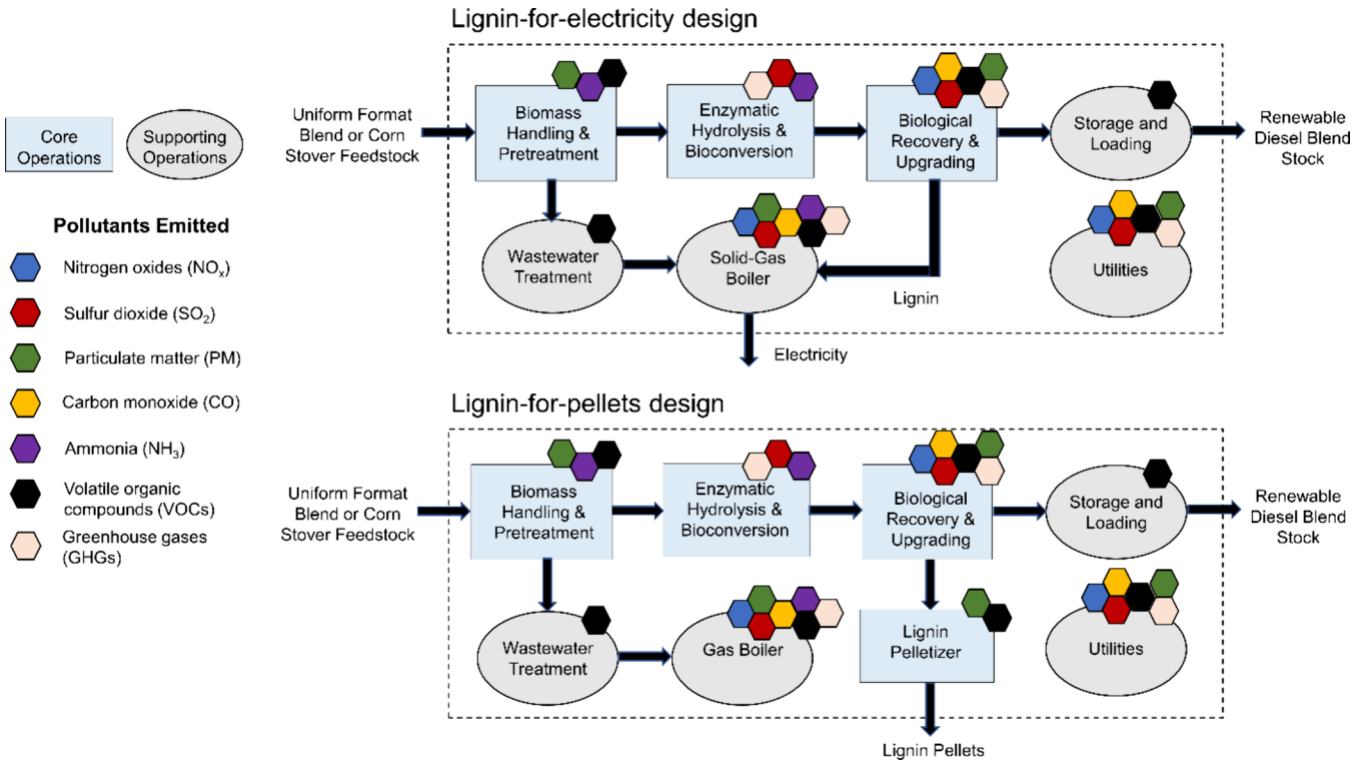


Figure 1. Criteria air pollutants and greenhouse gases (carbon dioxide, methane, and nitrous oxide) emitted by core and supporting operations for the production of a renewable diesel blend-stock from lignocellulosic biomass through aerobic respiration, modeled by Bhatt et al.[19] (modified from Eberle et al.[22]). The principal product of each system is renewable diesel blend stock; coproducts are shown as the other arrows exiting the system boundary. For lignin-for-electricity design, the coproduct is electricity; for the lignin-for-pellets design, the coproduct is pellets.

whose coproducts connect it to other systems that produce the same products.

2.2. GHG and CAP Emissions. In previous analysis, Bhatt et al.[20] leveraged the model developed by Davis et al.[15] and EFs from the U.S. Environmental Protection Agency (EPA) AP-42 database[21] to estimate facility-level potential-to-emit of the 12 design cases configured with different emission control strategies for reducing regulated air pollutants. Potential-to-emit is a metric measuring the maximum capacity of a stationary source to emit an air pollutant under its physical and operational design.[20] In addition to four CAPs and three of their precursors, net emissions from the three main GHGs contributing to global warming were also quantified: carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). The GHGs and air pollutants emitted by core and supporting unit operations within the biorefinery are illustrated in Figure 1 and reported numerically in the Supporting Information (SI).

In all biorefinery designs, certain core operations and boilers are important sources of GHG and CAP emissions. However, as reported by Bhatt et al.,[20] pelletizing the lignin reduces overall emissions from the system due to reduction of the amount of combusted fuel (in terms of heat input) in the boiler.[15] When lignin is used for pellet production, electricity generated on-site is insufficient to meet process demands, and the biorefinery will need to purchase electricity from the grid.[15] Building on the studies by Davis et al.[15] and Bhatt et al.,[20] this work compares the trade-offs between GHG and CAP emissions, by considering coproduct credits.

2.3. Net GHG and CAP Emissions Accounting Method. The life cycle GHG impacts of biofuels have been extensively studied in the literature; however, the body of literature analyzing CAP emissions of advanced biorefineries is limited. Although GHG emissions from stationary sources such as biorefineries are not currently regulated, embodied GHG emissions of the fuels produced are, and biorefineries are expected to be subject to air pollution regulations.[23,24] Thus, understanding the trade-offs between emissions of CAPs and GHGs from the biorefineries, under different design configurations, is important for informed decision-making.

The GHG and CAP accounting approaches used in this analysis considered *net* GHGs and CAPs emissions. Since the objective of this paper is to contrast the direct emission profiles of different biorefinery configurations, upstream and downstream processes for each displaced product such as feedstock processing and combustion of the fuel product were not included. Therefore, the system boundary for each biorefinery scenario included biorefinery operations only, as a first step toward a well-to-pump LCA.

Each biorefinery configuration produced coproducts that displace conventional products in their respective markets. To account for the environmental benefits of coproducts, system boundary expansion was applied. The system boundary expansion approach captures the displacement of conventional market products by biorefinery coproducts by allocating process emissions to the primary product and deducting the environmental impacts of the conventional products displaced by the coproducts. This methodology aligns with LCA methodologies for integrated biorefineries used in other studies,[25−27] avoiding arbitrary allocation factors that can arise from economic and mass-based allocations.[26] Economic allocation, in particular, can lead to distorted results due to market volatility affecting the costs of both electricity and lignin pellets.[26] Similarly, mass allocation was not used, as

https://doi.org/10.1021/acsestair.4c00152
ACS EST Air 2025, 2, 175−186

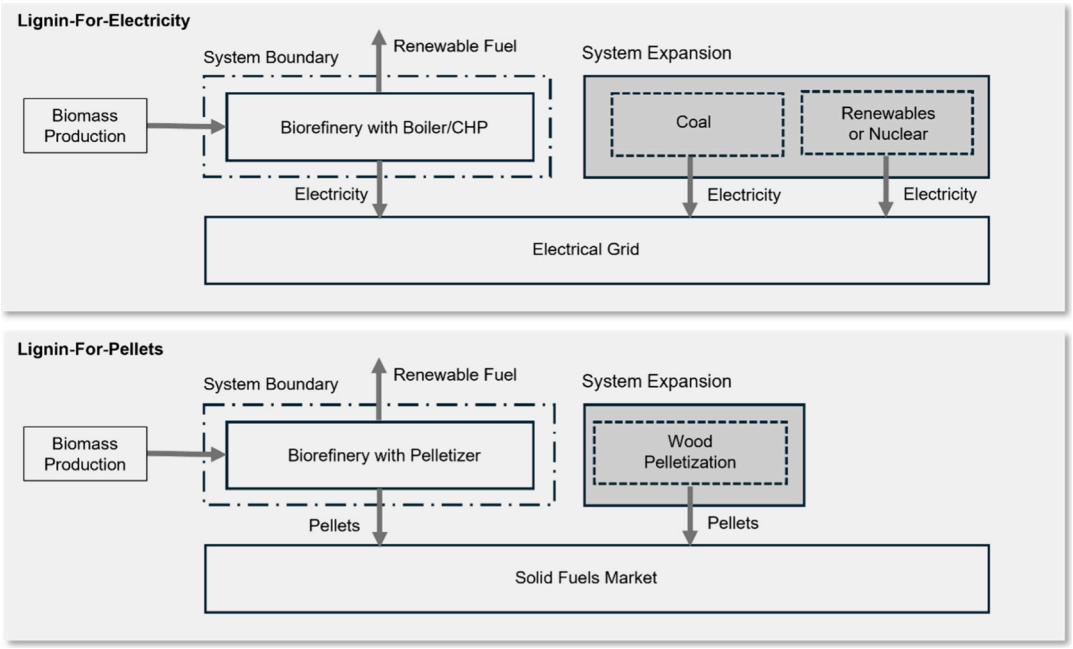


Figure 2. System boundaries and displaced systems for lignin-for-electricity and lignin-for-pellets biorefineries. The lignin-for-electricity biorefinery displaces coal or renewable/nuclear electricity generation, contributing electricity to the grid. The lignin-for-pellets biorefinery displaces conventional wood pelletization.

electricity, one of the coproducts, does not have a direct physical mass relationship with solid products.

The functional unit was set to a megajoule (MJ) of the RDB, reflecting the primary function of the biorefinery in fuel production. Outputs from the process model and the EFs of the products assumed to be displaced by the electricity and pellets coproducts were used to calculate potential emission credits given to the biorefinery. As illustrated in Figure 2, for the biorefinery producing an electricity coproduct, the additional electricity was assumed to displace either coal or zero-carbon electricity, reflecting the curtailment of renewables or nuclear power in the regions examined (elaborated later in the Methods section). These two extremes were chosen to represent the range of outcomes possible given that grid modeling was outside of the scope of this research. Similarly, lignin pellets coproduct were assumed to displace emissions associated with conventional wood pellet production. More details on the methodology used to calculate net emissions are provided in the Supporting Information (section 3).

2.4. Case Study Locations. Considering the availability of the two chosen feedstocks, we selected two case study locations for evaluation. The locations for the two case studies then determined the markets for electricity and pellets coproducts, which provide additional contrast and thus illuminate the potential impacts of different electricity emissions profiles and displacement credits on net emissions of the system.

2.4.1. Feedstock Availability. To select two case study locations, we considered the annual availability of corn stover and switchgrass at a county level as these are the main components in the biomass feedstocks evaluated. Based on published biomass resource assessments,[6,28] we selected Sioux County, Iowa and Brooks County, Georgia, as they are among the counties with the highest feedstock availability at a farmgate price of $60 per dmt. Sioux County, Iowa is projected to produce 592,000 dmt of corn stover and Brooks County, Georgia 37,000 dmt of switchgrass, respectively, in 2025.[6]

2.4.2. Selection of Displaced Products for Each Biorefinery Design and Location. For biorefineries using lignin for electricity generation, exported electricity displaces the generator at the margin. Determining the marginal generator in each of the case study regions requires detailed grid modeling, which is beyond the scope of this work. Therefore, we conducted a bounding analysis where electricity coproduct credits were calculated by assuming that surplus biorefinery electricity displaced either coal or zero-carbon (renewables or nuclear) electricity. Coal was selected as an upper bound as it represents the most emission-intensive technology in the electrical grid of the studied locations.[29] As EPA's 2020 Emissions and Generation Resource Integrated Database[29] (eGRID) indicates, coal generation in the eGRID subregions containing Iowa (Midwest Regional Reliability Organization West [MROW]) and Georgia (Southeastern Electric Reliability Council South [SRSO]) accounted for 38% and 16% of the total generation mix, respectively. Nonemitting, zero-carbon technologies such as solar, wind, hydropower, and nuclear—selected as the lower bound—accounted for 49% and 25% of the total generation in the MROW and SRSO subregions, respectively.[30] Note that our analysis does not consider technologies such as hydrogen-fueled combustion turbines because hydrogen combustion impacts on CAP emissions (especially NOx, but also from hydrogen production approaches) and hydrogen leakage rates are yet to be well-defined.

In a biorefinery using lignin to produce pellets, the displaced emissions were assumed to be those associated with the conventional production of wood pellets. The Southeastern US is a large producer of wood pellets with six facilities in Georgia (cumulative annual production capacity of 1,850,000 dmt).[30,31] Iowa also has a wood pellet facility (10,000 dmt).[30,31] Notably, about 84% of pellets produced in the US are exported,

https://doi.org/10.1021/acsestair.4c00152
ACS EST Air 2025, 2, 175−186

primarily to Europe where they are cofired with coal or used in residential heating.[31] However, domestic markets for pellet use remain underdeveloped, with studies highlighting economic and technical challenges associated with cofiring biomass with coal in the US.[32−34]

In our analysis, we assumed that the demand for pellets remains constant. Therefore, any increase in pellet production from lignin must be met with an equivalent decrease in the level of conventional wood pellet production. This assumption simplifies the displacement calculation by focusing on the direct substitution of lignin-derived pellets for wood-derived pellets without accounting for potential indirect market effects. While in reality increasing the supply of lignin-derived pellets could affect market prices and demand dynamics, our approach assumes a fixed demand scenario, where the additional lignin-based production directly offsets conventional pellet production on a one-to-one energy basis.

Recall that our system boundary is cut off at the production of the displaced fuels and does not include their end use (Figure 2). Correspondingly, embodied emissions of the infrastructure used in the displaced supply chains and other upstream emissions were not accounted for in the analysis; instead, only operational emissions were assumed to be avoided by electricity and lignin pellets coproducts. To capture geographic variations across case study states, location-based and fuel-specific EFs were used in this analysis for displaced electricity generation (section 2.5). Due to a lack of state-level data on solid fuel markets, national-average EFs were used to model the emissions of displaced wood pellets (section 2.6). The EFs used to calculate coproduct emission credits are provided in Table 1.

Table 1. Emission Factors for the Displaced Electricity and Pellets

	lignin-for-electricity		lignin-for-pellets
pollutant	coal electricity generation in Iowa (g kWh^{-1})	coal electricity generation in Georgia (g kWh^{-1})	wood pellets production (g MJ^{-1})
CO_2[a]	1030	863	5.2
CH_4[a]	0.12	0.08	0.011
N_2O[a]	0.017	0.012	0.00029
SO_2[a]	1.1	0.17	0.0045
NO_x[a]	0.77	0.38	0.0089
CO[b]	0.34	0.18	0.0033
$PM_{2.5}$[b]	0.07	0.04	0.00054
PM_{10}[b]	0.09	0.06	0.00069
VOC[b]	0.02	0.02	0.00082
NH_3[b]	0.0069	0.0068	0.000025

[a]Electricity emission factors at an eGRID subregion resolution (Iowa = MROW, Georgia = SRSO). [b]Electricity emission factors at a NERC region resolution (Iowa = MRO, Georgia = SERC). Emission factors for coal electricity correspond to steam turbine coal generation.

2.5. Emission Factors for Electricity Credits. As previously discussed, EFs of coal generation were used to construct an emissions inventory of GHGs and CAPs for calculating electricity credits in lignin-for-electricity biorefineries. Because there is no one database containing EFs for all air pollutants examined in this study, EFs were retrieved from multiple databases, as summarized in Table 1. Coal EFs of CO_2, CH_4, N_2O, SO_2, and NO_x were obtained from EPA's eGRID for year 2020 per eGRID subregion applicable to each case study location.[29] EFs reported by eGRID were derived from plant-level generation and 2020 emissions data per eGRID subregion level applicable to each case study location (MROW and SRSO).[29] Coal EFs for CO, $PM_{2.5}$, PM_{10}, and VOCs at a North American Electric Reliability Corporation (NERC) resolution come from the Greenhouse gases, Regulated Emissions, and Energy Use in Technologies (GREET) model[27,35] (Midwest Reliability Organization (MRO) and Southeastern Electric Reliability Council (SERC) NERC regions were used for Iowa and Georgia, respectively). The data reported by GREET were derived from National Emission Inventory 2017 datasets[35] and includes EFs for different fuel combustion technologies, while eGRID data reports EFs by fuel type and does not distinguish between combustion technologies. EFs for NH_3 were calculated by using data of NH_3 emissions reported to the EPA[36] and electricity generation efficiencies, at a NERC resolution for 2017, obtained from the GREET model.[27] Finally, electricity credits were assumed to be zero in cases where electricity displaced renewable or nuclear sources, given their absence of combustion emissions. It is important to recall that our system boundary considers operational emissions only; therefore, upstream emissions of renewable electricity technologies were not included in the analysis.

2.6. Pelletization Emission Factors. To estimate the emissions reduction from displacing wood pellets with lignin pellets, we used fuel consumption data reported by Morrison and Golden[37] and GHG and CAP EFs from the GREET model.[27] The wood pellets credits were estimated by considering operational emissions only (not life cycle) related to diesel consumption for pelletizing (2.06 kg per tonne of wood pellets)[37] and biomass combustion for drying pellets.[27,37] Given that GREET does not include NH_3 EFs, the NH_3 credit was estimated by using a nonroad diesel EF of 83.3 mg gal^{-1} of diesel and a biomass combustion EF of 0.63 g kg^{-1}, retrieved from EPA.[36]

2.7. Parametric Sensitivity Analysis on Coproduct Credits. A parametric sensitivity analysis was performed to analyze the impact of model parameters used in the calculations of coproduct credits on the resulting net emissions. Our sensitivity analysis focuses on coproduct credits because changing other parameters requires access to the original ASPEN process model, which this author group does not have. Since our objective was to identify high impact parameters across lignin utilization strategies, sensitivity was tested for only one plant location and scale (Iowa, 2000 dmtd), by independently increasing and decreasing five selected parameters: biorefinery emissions, net electricity exports, lignin exports, RDB output, and emission factors of all GHGs and CAPs (Table 1) associated with displaced coal electricity and wood pellets by 20%. These parameters were selected to determine to which of the tested parameters was net emissions most sensitive, as they are the only variables used in the net emissions calculation.

3. RESULTS AND DISCUSSION

Results are organized by accounting methods. Results in section 3.1 are reported in terms of net and gross emission intensities normalized by the established functional unit (MJ). Gross emission intensities were determined using gross emission outputs, while net emission intensities represent the difference between gross emissions and displacement credits. First, results for the 2000 dmtd biorefinery are compared in

https://doi.org/10.1021/acsestair.4c00152
ACS EST Air 2025, 2, 175−186

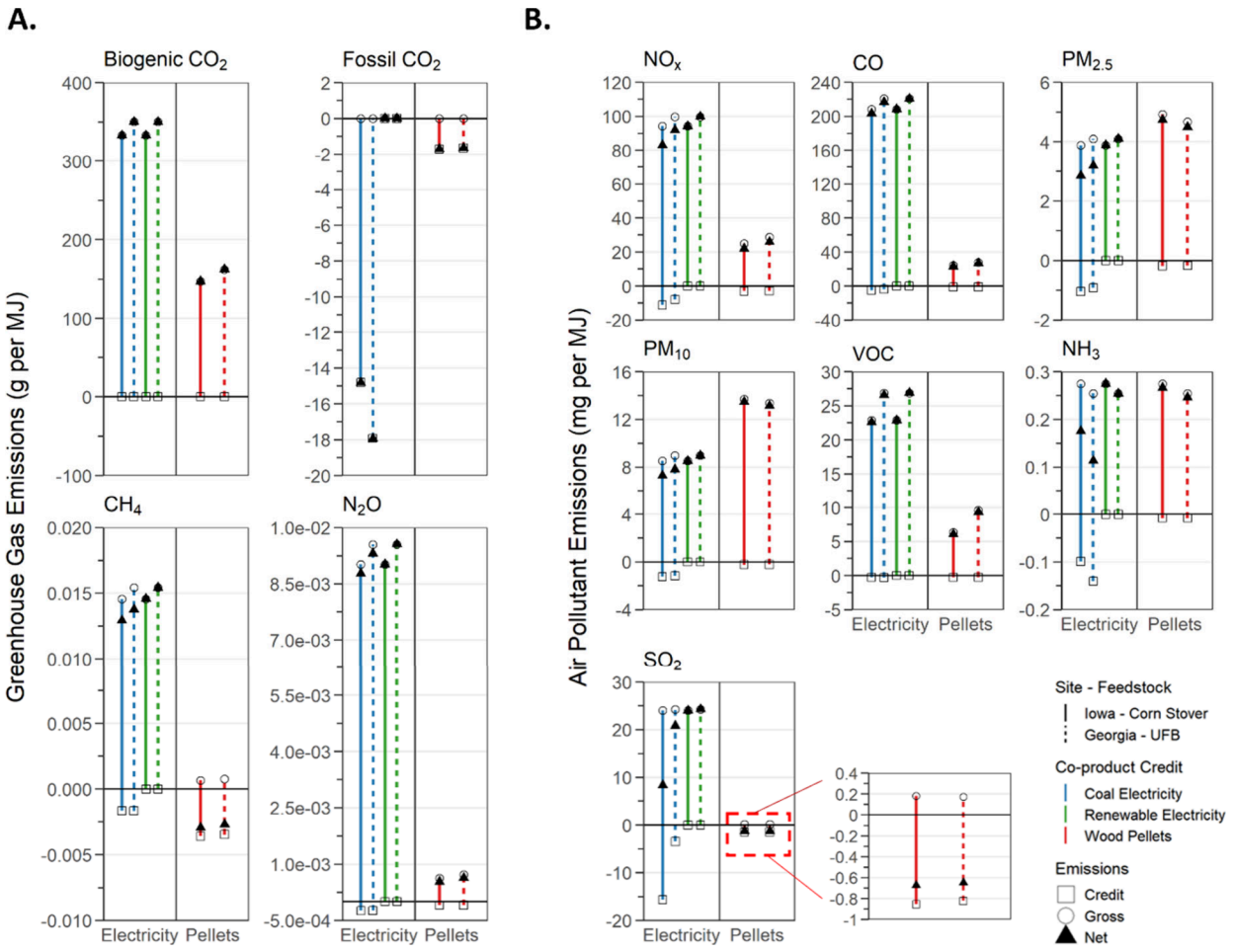


Figure 3. Gross and net: A) greenhouse gas and B) criteria air pollutant emissions for a hypothetical biorefinery processing 2000 dry metric tons per day of two types of herbaceous biomass feedstocks (corn stover and uniform feedstock blend [UFB]) to renewable diesel in Georgia and Iowa. Two different uses of lignin are contrasted: lignin combusted to generate excess electricity exported to the grid and lignin used to produce pellets. The results presented in this figure are reported on a functional unit basis (per MJ of fuel produced), which considers displacement credits from coproducts.

terms of lignin utilization strategies (section 3.1.1), followed by a comparison of electricity displacement credits, location, and feedstock (section 3.1.2) in lignin-for-electricity biorefineries. Next, lignin-for-pellets biorefineries are compared across feedstocks (section 3.1.3). Additionally, a discussion on the impact of the biorefinery scale is presented in section 3.1.4. Results from the sensitivity analysis identifying coproduct-related high impact parameters are interpreted (section 3.2). Finally, the implications of our results on the design of lignocellulosic biorefineries are explored in section 3.3.

3.1. Net Emissions Analysis. *3.1.1. Trade-offs between Lignin Utilization Strategies.* Potential emission trade-offs from different lignin utilization strategies were identified by comparing the outputs of four modeled biorefinery designs at the 2000 dmtd scale, illustrated in Figure 3. Net GHG emissions, categorized by biogenic CO_2, fossil CO_2, CH_4, and N_2O, are presented in Figure 3A. As results indicate, all emitted CO_2 is of biogenic origin and accounted as a positive emission output by the system, with no fossil fuel consumed within the biorefinery under either lignin utilization strategy. In contrast, the avoided CO_2 emissions from displaced coal electricity are of fossil origin, while the majority of CO_2 avoided by displacing wood pellets is also fossil-based, with only a minimal biogenic CO_2 credit from the combustion of biomass (0.4 g of CO_2 MJ^{-1}). When comparing GHG emissions outputs, CO_2 emissions represent the primary GHG emitted by the system, with net biogenic CO_2 emissions of the lignin-for-electricity biorefineries being three and 6 orders of magnitude higher than net CH_4 and N_2O emissions, respectively; a similar trend is observed in lignin-for-pellets results. The results indicate that net biogenic CO_2 emissions from the lignin-for-electricity designs are approximately 117% higher than those of biorefineries in which the lignin is diverted for pellets production. In terms of CH_4 emissions, the lignin-for-pellets design yields net negative CH_4 emissions (due to lower gross biorefinery emissions and larger CH_4 credits from the displacement of wood pellets), different from the net positive CH_4 emissions of the lignin-for-electricity design. Furthermore, the net N_2O emissions of lignin-for-electricity and lignin-for-pellets biorefineries differ by an order of magnitude. Results in Figure 3A demonstrate that lignin-for-electricity biorefineries produce higher net GHG emissions than the lignin-for-pellets biorefinery.

As shown in Figure 3B, net emission intensities of NO_x, CO, and VOCs in lignin-for-electricity biorefineries were generally similar across electricity displacement scenarios. When compared across lignin utilization strategies, net average NO_x emissions (25 mg of NO_x MJ^{-1}) for the lignin-for-pellets

https://doi.org/10.1021/acsestair.4c00152
ACS EST Air 2025, 2, 175−186

biorefinery were determined to be 72% lower relative to the average net emissions of lignin-for-electricity biorefineries (92 mg of NO_x MJ^{-1}). Similarly, the lignin-for-pellets biorefinery yielded an average 87% decrease in net CO emissions compared to the average emissions of lignin-for-electricity biorefineries (212 mg of CO MJ^{-1}). Contrastingly, lignin-for-electricity biorefineries showed lower $PM_{2.5}$ and PM_{10} emissions, with net emissions varying from 14% to 35% and 34% to 43% lower than average net emissions lignin-for-pellets biorefineries, respectively. The lower PM emissions are a consequence of higher gross PM emissions from the pelletization process in lignin-for-pellets biorefineries. Moreover, when lignin is used for pellets production rather than for electricity generation, the average net emissions of VOCs across all lignin-for-electricity scenarios were found to be 69% lower than average emissions from lignin-for-pellets biorefineries. Additionally, although both lignin utilization strategies generate similar gross NH_3 emissions, net NH_3 emissions were found to vary between 113 and 275 μg MJ^{-1} and 253 to 274 μg MJ^{-1} for the lignin-for-electricity and lignin-for-pellets biorefineries, respectively. The broader range of net NH_3 emissions observed in lignin-for-electricity biorefineries is primarily attributed to the displacement of electricity. Regarding net SO_2 emissions, lignin-for-pellets biorefineries yielded net negative results, ranging from −670 to −648 μg of SO_2 MJ^{-1}, whereas the average net emissions of the lignin-for-electricity biorefineries ranged from 8 to 24 mg of SO_2 MJ^{-1}. In summary, the results presented in Figure 3B highlight the reductions in NO_x, CO, VOCs, and SO_2 achieved by lignin-for-pellets biorefineries compared with lignin-for-electricity counterparts.

As our analysis reveals, the lignin-for-pellets biorefinery generates lower net GHG emissions and outperforms the lignin-for-electricity biorefineries in four of the seven studied CAPs and precursors. Despite these reductions, the pellet production process does emit greater $PM_{2.5}$ and PM_{10} relative to lignin-for-electricity biorefineries. Additionally, NH_3 emissions from the pretreatment process and the lack of large displacement credits adversely affect the NH_3 emissions profile of lignin-for-pellets biorefineries. To address these challenges, integrating additional emission controls, such as baghouses and wet scrubbers—which have demonstrated control efficiencies of 99%[38,39] in reducing PM and NH_3 emissions—could be a viable solution. Implementing these retrofits should lead to lower $PM_{2.5}$, PM_{10}, and NH_3 net emissions compared with lignin-for-electricity biorefineries, minimizing both GHG and CAP emissions from lignin biorefineries.

3.1.2. Displacement Credits, Location, and Feedstock Comparison Across Lignin-for-Electricity Biorefineries. Lignin-for-electricity biorefineries were evaluated by considering two displacement scenarios (zero-carbon and coal electricity) for each combination of feedstock and location. As Figure 3A illustrates, displacing either zero-carbon or coal electricity yielded very similar results, both reducing fossil CO_2, N_2O, and CH_4 emissions between 0 and 5%, 0−3%, and 0−11%, respectively. In a comparison of displacement credits from coal electricity across locations, the variation in N_2O and CH_4 credits was less than 1% between locations. However, despite coal electricity in MROW presenting a higher CO_2 EF than SRSO (Table 1), fossil CO_2 credits generated in the Iowa (MROW) biorefinery using corn stover were found to be 17% lower than those of the Georgia (SRSO) biorefinery processing UFB. This discrepancy correlates with the higher electricity output generated by the biorefinery processing UFB, leading to larger displacement credits. Conversely, a higher electricity output correlates with increased boiler emissions and gross emission intensity. In summary, the biorefinery in Iowa utilizing corn stover generates lower net CO_2 emissions (332 g CO_2 MJ^{-1}) compared to the UFB biorefinery located in Georgia (350 g CO_2 MJ^{-1}).

The results shown in Figure 3B illustrate the variability in air pollutant displacement credits across lignin-for-electricity biorefineries. These credits indicate potential reductions in the emission intensities of NO_x (0−12%), CO (0−2%), $PM_{2.5}$ (0−26%), PM_{10} (0−15%), VOCs (0−1%), NH_3 (0−55%), and SO_2 (0−65%), depending on the source of the displaced electricity. Notably, displacing coal electricity can yield considerable SO_2 credits due to the high sulfur content of coal. For instance, coal plants in MRO (Iowa) generate a credit of 15 mg of SO_2 MJ^{-1} compared to those in SERC (Georgia), which generate only 3.5 mg of SO_2 MJ^{-1}. This is attributed to the use of subbituminous coal (lower sulfur content) in MRO and use of bituminous coal (higher sulfur content) in SERC.[40] Similarly, the displacement of coal results in large NH_3 credits relative to those of biorefinery NH_3 emissions. These NH_3 credits likely originate from NH_3 slip in selective catalytic reduction units utilized in coal power plants,[41] while the majority (82%) of biorefinery NH_3 emissions are produced during the enzyme production process.[20] Furthermore, coal displacement leads to moderate NO_x and PM displacement credits, whereas credits for CO and VOC appear to be unaffected by the type of electricity displaced.

Additionally, our findings illustrate the impacts of feedstock and location on the net emission intensities of air pollutants in lignin-for-electricity biorefineries. For example, average net biorefinery emission intensities for NO_x, CO, $PM_{2.5}$, PM_{10}, and VOCs were found to be 9%, 7%, 3%, 2%, and 21% lower in Iowa than those of a biorefinery sited in Georgia. These differences are mainly attributed to the lower boiler heat input required by the corn stover (Iowa) biorefinery compared to that of the UFB (Georgia) biorefinery, resulting in reduced gross emissions. In contrast, net NH_3 emission intensities were found to be lower for the UFB biorefinery in Georgia, with values ranging from 0.11 to 0.25 mg NH_3 MJ^{-1}, compared to 0.18 to 0.27 mg NH_3 MJ^{-1} for the corn stover biorefinery in Iowa. The lower range of net NH_3 emission intensities in Georgia is a result of compositional differences between feedstocks, leading to higher NH_3 emissions for the corn stover biorefinery.[15] To summarize, the results indicate that utilizing a corn stover feedstock reduces the emissions of lignin-for-electricity biorefineries for most of the studied air pollutants, irrespective of the impact of displacement credits.

Results show that displacing coal-generated electricity yields higher GHG and CAP displacement credits compared with displacing zero-carbon electricity. However, as the capacity of low-carbon electricity expands and fossil fuel power plants retire within electrical grids, the potential displacement credits of lignin-for-electricity biorefineries are expected to asymptote toward those for the zero-carbon sources. Moreover, displacing electricity from emerging energy technologies, such as hydrogen-fuel combustion turbines, could present trade-offs in GHG and CAP credits. Hydrogen use can lead to indirect GHG emissions from hydrogen leakage[42] and also produce NO_x emissions from combustion.[43,44] Without substantial coproduct credits, lignin-for-electricity biorefineries will likely require investments in emission controls to treat NO_x, CO,

https://doi.org/10.1021/acsestair.4c00152
ACS EST Air 2025, 2, 175−186

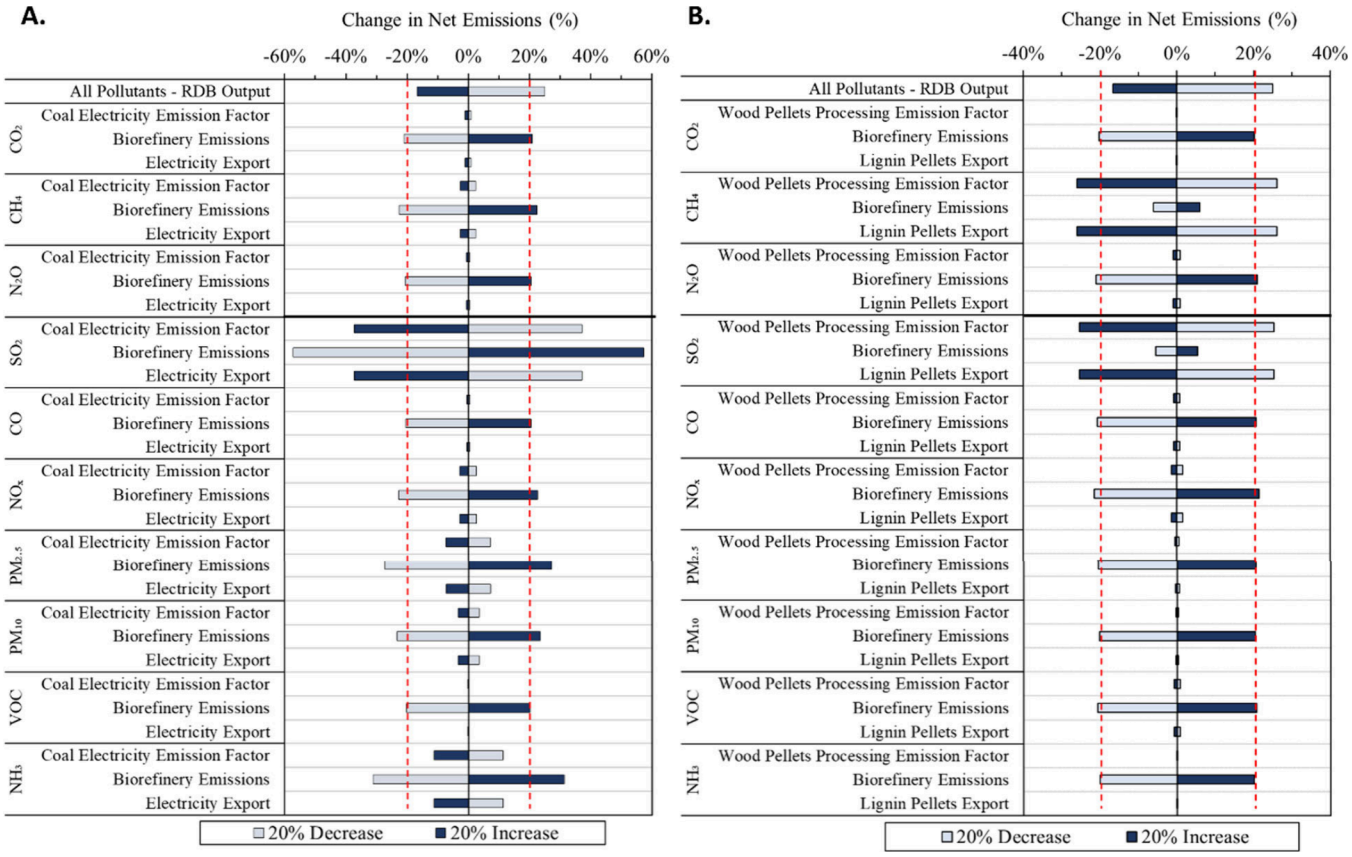


Figure 4. Changes in GHG and CAP pollutant net emissions from the 2000 dry metric ton per day corn stover lignin-for-electricity (A) and lignin-for-pellets (B) biorefinery in Iowa resulting from varying parameters related to the coproduct credits by ±20%: renewable diesel blend-stock (RDB) output, electricity exports, lignin exports, and emission factors.

VOCs, and SO_2 emissions below the levels of lignin-for-pellets biorefineries. An alternative strategy could involve redesigning the biorefinery to produce alternative lignin-based coproducts, especially those that could yield larger GHG and CAP displacement credits, such as chemicals.[45,46]

3.1.3. Feedstock Comparison Across Lignin-for-Pellets Biorefineries. Net emission intensities of the lignin-for-pellets biorefineries in both case study locations were calculated by assuming wood pellets are displaced. However, the analysis provides important insights into the impacts of feedstock and displacement credits across GHGs and CAPs. For instance, the comparison suggests that using corn stover as the feedstock yields net biogenic CO_2 and N_2O emission intensities of 147 g CO_2 MJ^{-1} and 0.6 mg N_2O MJ^{-1}, respectively, while the biorefinery processing UFB showed values of 162 g MJ^{-1} for biogenic CO_2 and 0.674 mg N_2O MJ^{-1} for N_2O. This difference can be attributed to the reduced heat content in the boiler feed from corn stover due its lower noncarbohydrate content, leading to decreased biogas requirements.[15] Additionally, corn stover biorefineries exhibit a lower net negative CH_4 emission intensity (−2.93 mg MJ^{-1}) due to a lower gross CH_4 emission intensity and larger lignin output, in contrast to UFB biorefineries (−2.70 mg MJ^{-1}). Significant CH_4 displacement credits, particularly from the combustion of diesel fuel used in pelletizing wood pellets, contribute to yield net negative CH_4 emissions across both feedstock types. Conversely, displacement credits for CO_2 and N_2O were minimal, suggesting that reducing these emissions requires process improvements or increased lignin production for greater credits. In summary, results underscore the impact of feedstock selection and displacement credits on the net GHG emissions of lignin-for-pellets biorefineries, with wood pellet displacement credits affecting net CH_4 emissions and corn stover usage minimizing net GHG emissions.

In terms of CAP emissions, the net emission intensities of NO_x, CO, VOC, and SO_2 for the corn stover biorefinery were determined to be 14%, 14%, 35%, and 4% lower than those resulting from the UFB biorefinery (Figure 2B). The net negative SO_2 emission intensities for the corn stover (−0.67 mg MJ^{-1}) and UFB (−0.65 mg MJ^{-1}) biorefineries were primarily driven by the large displacement credit associated with avoiding SO_2 emissions from the wood pelletizing process. Moreover, UFB biorefineries presented reductions of 5%, 2%, and 8% in their net $PM_{2.5}$, PM_{10}, and NH_3 emission intensities, respectively, compared with corn stover biorefineries. The higher net emission intensities of $PM_{2.5}$, PM_{10}, and NH_3 observed in the corn stover biorefinery are a result of higher gross emission intensities, as the influence of displacement credits was minimal across all CAPs and their precursors (SO_2 excepted). These results suggest that lignin-for-pellets biorefineries utilizing corn stover could benefit from the implementation of additional PM and NH_3 control technologies discussed earlier. With the incorporation of these control measures, such as baghouses and wet scrubbers, lignin-for-pellets biorefineries with corn stover feedstock could achieve lower net GHG and CAP emission intensities compared to both lignin-for-electricity biorefineries as well as lignin-for-pellets biorefineries with UFB feedstock.

https://doi.org/10.1021/acsestair.4c00152
ACS EST Air 2025, 2, 175−186

3.1.4. Impacts of Scale. Furthermore, the impacts of biorefinery scale on net emissions intensity of the analyzed GHGs and CAPs were compared. Results for all modeled facilities are provided in Figures S1−S2 of the Supporting Information. Facility scale was found to have a negligible impact on the gross emission intensity since emissions and fuel yields tend to scale linearly with feed rate (or scale).[15] Based on the results from all modeled scenarios, the gross emissions intensity of the three modeled facility scales remains constant for a given pollutant regardless of feedstock and lignin utilization strategy. This result provides a practical approach to estimating the gross emission outputs of lignocellulosic biorefineries in this range of scales and can potentially aid in the development of future emission inventories for similar process designs.

3.2. Sensitivity Analysis Results. A sensitivity analysis was performed to analyze the impacts of coproduct-related process parameters, including RBD output, electricity and lignin exports, and EFs used in the estimation of coproduct credits, on the net emission intensity of the studied pollutants, focusing on a single plant location and scale (Iowa, 2000 dmtd). Sensitivity results for the lignin-for-electricity biorefinery are presented in Figure 4A, organized by pollutants. First, the sensitivity of biorefinery RDB output was examined, wherein a 16% decrease across all GHG and CAP emission intensities resulted from a 20% increase in the fuel yields. Conversely a 20% decrease in fuel yields led to a 25% increase in the emission intensities for all studied GHGs and CAPs. As expected, increasing fuel yields led to reduced net emissions when both gross emissions and credits remained constant, while the opposite was true for a reduction in fuel yields. Furthermore, net emissions showed a nonlinear relationship with fuel yields as neither displacement credits nor gross emissions were proportional to changes in yields. This trend is further highlighted in the results of the lignin-for-pellets biorefinery, illustrated in Figure 4B.

The sensitivity analysis for GHG emissions of the lignin-for-electricity biorefinery shows that changes in coal electricity EFs had minimal effects on net CO_2, CH_4, and N_2O emissions (Figure 4A). As displacement credits are directly proportional to EFs, an increase in EFs resulted in larger credits, leading to lower net emissions. Similarly, modifying the biorefinery's electricity export also displayed a similar relationship, with decreases in electricity outputs yielding higher net emissions (lower displacement credits). Notably, changes in biorefinery emissions had a nearly proportional effect on the net emission intensities of GHGs, with a 20% increase leading to 21%, 23%, and 21% increases to the baseline net biogenic CO_2, CH_4, and N_2O emissions, respectively. The greater sensitivity to biorefinery emissions is attributed to the low displacement credits observed in GHG emissions.

The emission intensities of CAPs exhibited a similar trend in response to changes in electricity EFs and exports as observed for GHGs. Notably, SO_2 and NH_3, with the largest displacement credits, showed the greatest change in net emissions, with a 37% and 11% deviation from the baseline, respectively. Conversely, net emissions of CO, NO_x, $PM_{2.5}$, PM_{10}, and VOCs changed by less than 10% relative to the baseline. Additionally, net emission intensities of SO_2 and NH_3 were also most influenced by changes in biorefinery emissions, registering 57% and 31% changes to the baseline, respectively. Sensitivity results further support the conclusion that substantial reductions in the net GHG and CAP emissions from lignin-for-electricity biorefineries require reductions in direct biorefinery emissions.

Results from the lignin-for-pellets biorefinery, illustrated in Figure 4B, indicate that modifications to the EFs of wood pellet production and biorefinery pellet exports have minimal impact on net biogenic CO_2 and N_2O emissions. This can be attributed to the relatively small displacement credits observed for CO_2 and N_2O. However, independently increasing pellet exports and CH_4 EFs results in a 26% decrease in net emissions, as displacement credits outweigh direct CH_4 emissions. Therefore, variations in biorefinery emissions led to a lower change in net CH_4 emissions, while net CO_2 and N_2O emissions changed by 20% and 21%, respectively. In line with the results for the lignin-for-electricity biorefinery, the sensitivity analysis for the lignin-for-pellets biorefinery suggests that minimizing CO_2 and N_2O biorefinery emissions is critical to decrease net GHG emissions.

For CAPs, the EFs for wood pellets and biorefinery pellet exports had a notable influence on the net emission intensity of SO_2, with results indicating a 25% change to the baseline (Figure 4B). Net SO_2 emissions were found to be dominated by displacement credits; therefore, parameters directly impacting displacement credits were found to be highly sensitive. Consequently, for other CAPs, adjustments to the baseline wood pellets EFs and biorefinery pellets exports had a negligible effect on their net emission intensities, with changes lower than 2% relative to the baseline. This is expected, as displacement credits for CO, NO_x, $PM_{2.5}$, PM_{10}, VOCs, and NH_3 were small compared to gross emissions. As a result, biorefinery emissions showed a nearly proportional impact on the net emission intensity of all CAPs except for SO_2, which changed by only 5% relative to the baseline net emission intensity.

3.3. Implications. *3.3.1. Spatial Distribution of Emissions.* Results highlight how lignin pelletization reduces net emissions of most CAPs (SO_2, CO, NO_x, and VOC), while diverting the lignin for electricity export reduces net $PM_{2.5}$, PM_{10}, and NH_3 emissions. However, it is important to consider the spatial distribution of the avoided air pollutants, for which our analysis does not account for. The displacement of electricity and wood pellets can mitigate air pollution originating from the source of the displaced products such as coal power plants or wood pelletizing facilities. For instance, in the case of a biorefinery located in Iowa, air pollutant reductions will occur in North Dakota, where the coal power plant of the MRO market is located. Despite this limitation, our analysis provides the foundations for conducting air quality modeling and evaluating the spatial distribution of air pollutant concentrations, as well as their impact on public health, both in terms of remaining emissions and avoided. Marginal air quality impact from a select renewable diesel blendstock production scenarios analyzed in this work and its comparison to the feedstock production will be included in a future publication.[47]

3.3.2. Electricity-Associated Emissions. As the results indicated, lignin-for-pellets biorefineries were found to reduce biorefinery biogenic CO_2 by over half relative to lignin-for-electricity biorefineries. However, lignin pelletization incurs higher indirect fossil GHG emissions due to the electricity consumption required for biorefinery operations. The extent of electricity-associated emissions in lignin-for-pellets biorefineries depends on the carbon intensity of the local grid (Figure S5). Therefore, to mitigate electricity-associated emissions, it is advisible to locate lignin-for-pellets biorefineries

https://doi.org/10.1021/acsestair.4c00152
ACS EST Air 2025, 2, 175−186

in regions where low-carbon electricity sources are readily available, especially considering the expected decline in average electricity emission factors due to the increasing adoption of renewable energy technologies. Additionally, when factoring in the benefits of reduced CAP emissions, lignin pelletization could offer a cooptimized approach to minimize CAP and GHG emissions. This could be achieved through the implementation of additional control technologies targeting PM and NH_3 emissions, coupled with the utilization of low-carbon electricity to power the biorefinery operations.

An alternative method to mitigate electricity-associated emissions of the lignin-for-pellets biorefinery could involve the combustion of a fraction of the pelletized lignin for on-site electricity generation. This strategy could reduce reliance on grid electricity, thereby reducing indirect electricity emissions. However, this hybrid lignin utilization approach comes with certain trade-offs. The combustion process would likely lead to an increase in emissions of other CAPs, as results from the lignin-for-electricity biorefinery demonstrate. Moreover, the added costs of the solids boiler and the loss of coproduct revenue could make this strategy unfeasible from a techno-economic perspective, a result requiring further investigation beyond the scope of this analysis. Overall, while lignin-for-pellets biorefineries offer a viable solution to reduce both the GHG and CAP emissions of lignocellulosic biorefineries, further analysis is required to fully understand the environmental and economic implications of the strategies discussed here.

3.3.3. Other Potential Lignin Markets. The results presented here provide insights into the combinations of lignin utilization, scale, and feedstock that could effectively reduce both GHG emissions and CAPs in lignocellulosic biorefineries when emissions from electricity and wood pellets are avoided. However, it is important to acknowledge that these results and conclusions must be considered within the context of the displaced products. The environmental benefits of lignin utilization depend on the assumption that lignin-derived products, such as pellets or electricity, are displacing conventional alternatives with higher emissions such as fossil-based electricity. The magnitudes of the GHG and CAP reductions are directly tied to the characteristics of these displaced products and the efficiency with which lignin can substitute for them. Therefore, while the analysis demonstrates potential environmental benefits, the real-world implications are dependent on the specific market dynamics, demand, and availability of both lignin-based and conventional products in each context.

Several studies have demonstrated the potential of lignin as a precursor for carbon fibers, which could displace petroleum-based materials like polyacrylonitrile.[45,48,49] Displacing polyacrylonitrile, which is associated with significant GHG and CAP emissions (Figure S6), would result in different coproduct credits and influence overall net emissions compared to displacing wood pellets. While the coproduct credits from lignin pellets may vary based on the displaced product, lignin-for-pellets facilities still provide greater environmental benefits than lignin-for-electricity facilities, largely due to the significant reduction in boiler emissions. However, further research is needed to fully explore lignin's market potential beyond its role as a fuel, particularly in high-value applications such as carbon fiber production. Future market studies, which were beyond the scope of this analysis, will be crucial to better understand the feasibility of lignin in domestic markets.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestair.4c00152.

Additional details on methods and results (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Vikram Ravi − *Strategic Energy Analysis Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States;* orcid.org/0000-0002-5888-318X; Email: vikram.ravi@nrel.gov

Authors

David Quiroz − *Strategic Energy Analysis Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States; Mechanical Engineering, Colorado State University, Fort Collins, Colorado 80522, United States;* orcid.org/0009-0001-0071-6047

Yimin Zhang − *Strategic Energy Analysis Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States*

Arpit Bhatt − *Strategic Energy Analysis Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States;* orcid.org/0000-0002-3780-1800

Garvin Heath − *Strategic Energy Analysis Center, National Renewable Energy Laboratory, Golden, Colorado 80401, United States*

Complete contact information is available at: https://pubs.acs.org/10.1021/acsestair.4c00152

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was authored by the National Renewable Energy Laboratory, operated by Alliance for Sustainable Energy, LLC, for the U.S. Department of Energy (DOE) under Contract No. DE-AC36-08GO28308. Support for the work was also provided by ExxonMobil under Agreement CRD-18-00765. The views expressed in the article do not necessarily represent the views of the DOE, the U.S. Government or ExxonMobil. The U.S. Government retains and the publisher, by accepting the article for publication, acknowledges that the U.S. Government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this work, or allow others to do so, for U.S. Government purposes.

■ ABBREVIATIONS

CAP:Criteria Air Pollutants
CH_4:Methane
CO:Carbon Monoxide
CO_2:Carbon Dioxide
CO_{2e}:Carbon Dioxide Equivalent
dmtd:Dry Metric Tons Per Day
EF:Emission Factor
EPA:Environmental Protection Agency

eGRID:Emissions & Generation Resource Integrated Database
GHG:Greenhouse Gas
GREET:Greenhouse Gases, Regulated Emissions, and Energy Use in Technologies
IPCC:Intergovernmental Panel on Climate Change
LCA:Life Cycle Assessment
MJ:Megajoule
MROW:Midwest Regional Reliability Organization West
NH_3:Ammonia
NO_2:Nitrogen Dioxide
N_2O:Nitrous Oxide
NERC:North American Electric Reliability Corporation
NREL:National Renewable Energy Laboratory
PM:Particulate Matter
RD:Renewable Diesel
RDB:Renewable Diesel Blend-stock
SI:Supporting Information
SO_2:Sulfur Dioxide
SRSO:Southeastern Electric Reliability Council South
TPD:Tonnes Per Day
UFB:Uniform Format Blend
VOC:Volatile Organic Compounds

REFERENCES

(1) *A Roadmap for Moving to a Competitive Low Carbon Economy in 2050*; European Commission, Brussels. https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=COM:2011:0112:FIN:en:PDF (accessed 2023-03-17).
(2) Sissine, F. *Energy Independence and Security Act of 2007: A Summary of Major Provisions*; Congressional Research Service, 2007. https://apps.dtic.mil/sti/pdfs/ADA475228.pdf (accessed 2023-03-17).
(3) California Air Resources Board. Low Carbon Fuel Standard Regulation, Preprint , 2018. https://ww2.arb.ca.gov/our-work/programs/low-carbon-fuel-standard (accessed 2023-03-17).
(4) Muratori, M., et al. *US National Blueprint for Transportation Decarbonization: A Joint Strategy to Transform Transportation*; No. DOE/EE-2674 ; Department of Energy, Office of Energy Efficiency and Renewable Energy, United States, 2023.
(5) Ale, S.; Femeena, P. V; Mehan, S.; Cibin, R. *Bioenergy with Carbon Capture and Storage*; Academic Press, 2019.
(6) Brandt, C. C.; Davis, M.; Davison, B.; Eaton, L.; Efroymson, R. A.; Hilliard, M. R.; Kline, K. *Billion-Ton Report: Advancing Domestic Resources for a Thriving Bioeconomy, Vol. 1: Economic Availability of Feedstocks*; U.S. Department of Energy Oak Ridge National Laboratory, Oak Ridge, TN, 2016.
(7) McKone, T. E. Grand Challenges for Life-Cycle Assessment of Biofuels. *Environ. Sci. Technol.* **2011**, *45*, 1751−1756.
(8) *International Organization for Standardization, ISO 14001:2015 Environmental Management Systems − Requirements with Guidance for Use*; ISO, 2015.
(9) Han, J.; Canter, C.; Cai, H.; Wang, M.; Qin, J.; Dunn, J.; Gustafson, R.; Kelley, S.; Morales, R.; Oneil, E.; Ou, L. Carbon Dynamics for Biofuels Produced from Woody Feedstocks; Argonne National Laboratory, 2018.
(10) Cai, H.; Dunn, J. B.; Wang, Z.; Han, J.; Wang, M. Q. Life-Cycle Energy Use and Greenhouse Gas Emissions of Production of Bioethanol from Sorghum in the United States. *Biotechnol. Biofuels* **2013**, *6*, 141.
(11) Han, J.; Tao, L.; Wang, M. Well-to-Wake Analysis of Ethanol-to-Jet and Sugar-to-Jet Pathways. *Biotechnol. Biofuels* **2017**, *10*, 21.
(12) Cai, H.; Han, J.; Wang, M.; Davis, R.; Biddy, M.; Tan, E. Life-Cycle Analysis of Integrated Biorefineries with Co-Production of Biofuels and Bio-Based Chemicals: Co-Product Handling Methods and Implications. *Biofuels Bioprod. Biorefining* **2018**, *12*, 815−833.
(13) Tessum, C. W.; Marshall, J. D.; Hill, J. D. A Spatially and Temporally Explicit Life Cycle Inventory of Air Pollutants from Gasoline and Ethanol in the United States. *Environ. Sci. Technol.* **2012**, *46*, 11408−11417.
(14) Galetovic, A.; Muñoz, C. M. Wind, Coal, and the Cost of Environmental Externalities. *Energy Policy* **2013**, *62*, 1385−1391.
(15) Davis, R.; Bhatt, A. H.; Zhang, Y.; Tan, E. C. D.; Ravi, V.; Heath, G. Biorefinery Upgrading of Herbaceous Biomass to Renewable Hydrocarbon Fuels, Part 1: Process Modeling and Mass Balance Analysis. *J. Clean. Prod* **2022**, *362*, No. 132439.
(16) Hill, J.; Polasky, S.; Nelson, E.; Tilman, D.; Huo, H.; Ludwig, L.; Neumann, J.; Zheng, H.; Bonta, D. Climate Change and Health Costs of Air Emissions from Biofuels and Gasoline. *Proc. Natl. Acad. Sci.* **2009**, *106*, 2077−2082.
(17) Tessum, C. W.; Hill, J. D.; Marshall, J. D. Life Cycle Air Quality Impacts of Conventional and Alternative Light-Duty Transportation in the United States. *Proc. Natl. Acad. Sci.* **2014**, *111*, 18490−18495.
(18) Cook, R.; Phillips, S.; Houyoux, M.; Dolwick, P.; Mason, R.; Yanca, C.; Zawacki, M.; Davidson, K.; Michaels, H.; Harvey, C.; Somers, J.; Luecken, D. Air Quality Impacts of Increased Use of Ethanol under the United States' Energy Independence and Security Act. *Atmos. Environ.* **2011**, *45*, 7714−7724.
(19) Ravi, V.; Gao, A. H.; Martinkus, N. B.; Wolcott, M. P.; Lamb, B. K. Air Quality and Health Impacts of an Aviation Biofuel Supply Chain Using Forest Residue in the Northwestern United States. *Environ. Sci. Technol.* **2018**, *52*, 4154−4162.
(20) Bhatt, A. H.; Zhang, Y.; Davis, R.; Heath, G.; Ravi, V. Biorefinery Upgrading of Herbaceous Biomass to Renewable Hydrocarbon Fuels, Part 2: Air Pollutant Emissions and Permitting Implications. *J. Clean. Prod* **2022**, *362*, No. 132409.
(21) U.S. EPA; *Office of Air Quality Planning and Standards. AP-42: Compilation of Air Emissions Factors*. https://www.epa.gov/air-emissions-factors-and-quantification/ap-42-compilation-air-emissions-factors (accessed 2023-03-15)..
(22) Eberle, A.; Bhatt, A.; Zhang, Y.; Heath, G. Potential Air Pollutant Emissions and Permitting Classifications for Two Biorefinery Process Designs in the United States. *Environ. Sci. Technol.* **2017**, *51*, 5879−5888.
(23) U.S. EPA. *New Source Performance Standards and Permitting Requirements | US EPA*. https://www.epa.gov/controlling-air-pollution-oil-and-natural-gas-operations/new-source-performance-standards-and (accessed 2024-02-03)..
(24) U.S.C. Title 42 - the Public Health and Welfare. https://www.govinfo.gov/content/pkg/USCODE-2013-title42/html/USCODE-2013-title42-chap85-subchapI-partA-sec7401.htm (accessed 2024-02-03).
(25) Scown, C. D.; Gokhale, A. A.; Willems, P. A.; Horvath, A.; McKone, T. E. Role of Lignin in Reducing Life-Cycle Carbon Emissions, Water Use, and Cost for United States Cellulosic Biofuels. *Environ. Sci. Technol.* **2014**, *48*, 8446−8455.
(26) Cai, H.; Han, J.; Wang, M.; Davis, R.; Biddy, M.; Tan, E. Life-Cycle Analysis of Integrated Biorefineries with Co-Production of Biofuels and Bio-Based Chemicals: Co-Product Handling Methods and Implications. *Biofuels, Bioproducts and Biorefining* **2018**, *12* (5), 815−833.
(27) Wang, M. *Greenhouse Gases, Regulated Emissions, and Energy Use in Technologies Model ® (2020 Excel)*; Argonne National Laboratory, 2020. DOI: 10.11578/GREET-EXCEL-2020/DC.20200912.1.
(28) Downing, M.; Eaton, L. M.; Graham, R. L.; Langholtz, M. H.; Perlack, R. D.; Turhollow, J. A. F.; Stokes, B.; Brandt, C. C. *U.S. Billion-Ton Update: Biomass Supply for a Bioenergy and Bioproducts Industry*; U.S. Department of Energy, 2011. DOI: 10.2172/1219219. (accessed 2023-02-03)..
(29) US EPA 2020 *Emissions & generation resource integrated database (eGRID)*. www.epa.gov/energy/emissions-generation-resource-integrated-database-egrid (accessed 2023-03-17).
(30) U.S. Energy Information Administration. *Wood and wood waste - U.S. Energy Information Administration*. https://www.eia.gov/

https://doi.org/10.1021/acsestair.4c00152
ACS EST Air 2025, 2, 175−186

energyexplained/biomass/wood-and-wood-waste.php (accessed 2023-03-09).
(31) U.S. Energy Information Administration. *New EIA Survey Collects Data on Production and Sales of Wood Pellets*. https://www.eia.gov/todayinenergy/detail.php?id=29152 (accessed 2023-03-17).
(32) Picciano, P.; Aguilar, F. X.; Burtraw, D.; Mirzaee, A. Environmental and Socio-Economic Implications of Woody Biomass Co-Firing at Coal-Fired Power Plants. *Resour Energy Econ* **2022**, *68*, No. 101296.
(33) Mei, B.; Wetzstein, M. Burning Wood Pellets for US Electricity Generation? A Regime Switching Analysis. *Energy Econ* **2017**, *65*, 434−441.
(34) Xian, H.; Colson, G.; Mei, B.; Wetzstein, M. E. Co-Firing Coal with Wood Pellets for U.S. Electricity Generation: A Real Options Analysis. *Energy Policy* **2015**, *81*, 106−116.
(35) Ou, L.; Cai, H. *Update of Emission Factors of Greenhouse Gases and Criteria Air Pollutants, and Generation Efficiencies of the U.S. Electricity Generation Sector, 1660468*, 162084, ANL-20/41, 2020.
(36) Roe, S. M.; Spivey, M. D.; Lindquist, H. C.; Thesing, K. B.; Strait, R. P.; E.H. Pechan & Associates, Inc. *Estimating Ammonia Emissions From Anthropogenic Nonagricultural Sources - Draft Final Report*; U.S. Environmental Protection Agency, 2004; Vol. *87*. https://www.epa.gov/sites/default/files/2015-08/documents/eiip_areasourcesnh3.pdf (accessed 2023-03-17).
(37) Morrison, B.; Golden, J. S. Life Cycle Assessment of Co-Firing Coal and Wood Pellets in the Southeastern United States. *J. Clean Prod* **2017**, *150*, 188−196.
(38) *EPA Air Pollution Control Cost Manual*, Sixth ed.; United States Environmental Protection Agency, 2002. https://www3.epa.gov/ttncatc1/dir1/c_allchs.pdf (accessed 2024-03-13)..
(39) Phillips, J.; Blaszczak, R. J. *Control and Pollution Prevention Options for Ammonia Emissions*; US EPA, 1995. https://www3.epa.gov/ttncatc1/dir1/ammonia.pdf (accessed 2024-03-13)..
(40) Sherwood, J.; Bickhart, R.; Murawski, E.; Dhanani, Z.; Lytle, B.; Carbajales-Dale, P.; Carbajales-Dale, M. Rolling Coal: The Greenhouse Gas Emissions of Coal Rail Transport for Electricity Generation. *J. Clean. Prod* **2020**, *259*, No. 120770.
(41) NOx Control Technical Division; Institute of Clean Air Companies. *Selective Catalytic Reduction (SCR) Control of NO_x Emissions from Fossil Fuel-Fired Electric Power Plants*; 2009. https://cdn.ymaws.com/icac.site-ym.com/resource/resmgr/Standards_WhitePapers/SCR_WhitePaper_final_2009.pdf (accessed 2024-03-13).
(42) Sand, M.; Skeie, R. B.; Sandstad, M.; Krishnan, S.; Myhre, G.; Bryant, H.; Derwent, R.; Hauglustaine, D.; Paulot, F.; Prather, M.; Stevenson, D. A Multi-Model Assessment of the Global Warming Potential of Hydrogen. *Communications Earth & Environment* **2023**, *4* (1), 1−12 2023 4:1.
(43) Valles, M. A.; Kessler, I.; Windom, B. C. Review and Development of Natural Gas/Hydrogen Fuel Flexible Reduced Chemical Mechanism for High-Order Modeling of Gas Turbines. *Proceedings of the ASME Turbo Expo* 2023, Vol. *86953*, V03AT04A030 . DOI: 10.1115/GT2023-101471.
(44) York, W. D.; Ziminsky, W. S.; Yilmaz, E. Development and Testing of a Low NOx Hydrogen Combustion System for Heavy-Duty Gas Turbines. *J. Eng. Gas Turbine Power* **2013**, *135* (2), No. 022001.
(45) Henry, C.; Nejad, M. Lignin-Based Low-Density Rigid Polyurethane/Polyisocyanurate Foams. *Ind. Eng. Chem. Res.* **2023**, *62* (18), 6865−6873.
(46) Ou, L.; Dou, C.; Yu, J. H.; Kim, H.; Park, Y. C.; Park, S.; Kelley, S.; Lee, E. Y. Techno-Economic Analysis of Sugar Production from Lignocellulosic Biomass with Utilization of Hemicellulose and Lignin for High-Value Co-Products. *Biofuels, Bioproducts and Biorefining* **2021**, *15* (2), 404−415.
(47) Isaias, M.; Ravi, V.; Thind, M.; Heath, G. *Biochemical Conversion of Herbaceous Biomass to Renewable Diesel: Biorefinery Marginal Air Quality Impacts and Comparison to Feedstock Production*; NREL/TP-6A20-89796; National Renewable Energy Lab. (NREL), Golden, CO (United States), 2024.
(48) Das, S.; Warren, J.; West, D.; Schexnayder, S. M. *Global Carbon Fiber Composites Supply Chain Competitiveness Analysis*. No. ORNL/SR-2016/100l NREL/TP-6A50-66071 . 2016.
(49) Souto, F.; Calado, V.; Pereira, N. Lignin-Based Carbon Fiber: A Current Overview. *Mater. Res. Express* **2018**, *5* (7), 072001.

https://doi.org/10.1021/acsestair.4c00152
ACS EST Air 2025, 2, 175−186