



Biorefinery upgrading of herbaceous biomass to renewable hydrocarbon fuels, Part 2: Air pollutant emissions and permitting implications

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ABSTRACT

The development of advanced biofuel production facilities is still at a nascent stage, and the biorefineries could face challenges in obtaining air permits required for their construction and operation because they are novel emission sources. To fill gaps in knowledge regarding potential emissions, we perform a detailed federal regulatory analysis and estimate air pollutant emissions for an advanced biorefinery that produces renewable diesel blendstock (RDB) from lignocellulosic biomass via aerobic respiration documented in Part 1 of the paper. We evaluate 12 design permutations that include two feedstocks, a uniform format blend (UFB) and corn stover; three biorefinery scales, 2,000, 5,200, and 9,100 dry metric tons per day (dmt) of lignocellulosic biomass feed; and two lignin uses, as either a boiler fuel or for pellet production. We also evaluate 6 additional design permutations by incorporating non-emitting renewable power, which could reduce up to 63% carbon monoxide (CO) and nitrogen oxides (NO_x) each, 21% volatile organic compounds (VOC), and 43% hazardous air pollutants (HAP) as opposed to on-site power production, for biorefineries that produce pellets, using either UFB or corn stover. Our results indicate that all 18 design permutations would be classified as a major source under the Clean Air Act's New Source Review program without additional emission controls. Compared to using lignin as a boiler fuel, diverting lignin for pellet production reduces the emissions of CO up to 88%, NO_x up to 73%, sulfur dioxide (SO₂) up to 99%, VOCs up to 72%, and HAPs up to 66%. Additionally, we explore control options that could further reduce emissions and assess whether reductions are enough to achieve minor source classification. Insights from our analysis can help biorefinery developers and regulators develop permitting strategies to mitigate investment risks.

1. Introduction

The United States has the potential to supply one billion dry tons of biomass resources annually, composed primarily of forestry, agricultural residues, urban waste, and algae, reportedly without adversely affecting the environment (U.S. Department of Energy, 2016). Producing and utilizing renewable hydrocarbon fuels from lignocellulosic biomass feedstocks offers a promising option for decarbonizing the transportation sector and reduces fossil energy consumption and greenhouse gas (GHG) emissions. As sustainably produced low emission biomass feedstock is environmentally friendly, utilizing it for production of hydrocarbon fuels and energy provides a cleaner approach towards a green economy while expanding the renewable fuel production capacity.

Because hydrocarbon biofuels from lignocellulosic biomass have not been commercialized, prior research has been focused on exploring the

technical feasibility and economics of various conversion technologies. For example, Brown (2015) reviewed the technical and economic feasibility of various thermochemical cellulosic biofuel pathways. Similarly, Bhatt et al. (2016) evaluated the impact of adopting control technologies on the biofuel production cost. Brown et al. (2014) elaborated economic challenges and strategies pursued to increase the commercial adoption of biochemical and thermochemical biorefineries. Klein-Marcuschamer et al. (2010) developed a model for lignocellulosic ethanol production to help biofuel communities on the energy, environmental, and economic performance metrics. However, evaluation of environmental effects of hydrocarbon fuel production is very limited with the exception of life cycle greenhouse gas (GHG) emissions and fossil energy use. For instance, Han et al. (2013) performed lifecycle analysis of hydrocarbon fuel production from fast pyrolysis process. Similarly, Bartling et al. (2021) performed a combined techno-economic and life cycle analysis for a biorefinery utilizing reductive catalytic

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Abbreviations

RDB	Renewable Diesel Blendstock
UFB	Uniform Format Blend
Dmtd	dry metric tons per day
CO	carbon monoxide
NO _x	nitrogen oxides
VOC	volatile organic compounds
HAP	hazardous air pollutants
GHG	greenhouse gas
NSR	New Source Review
PTE	potential-to-emit
PM	particulate matter
SO _x	sulfur oxides
NSPS	New Source Performance Standards
NH ₃	ammonia
MMBtu	million British thermal unit
H ₂ SO ₄	sulfuric acid
NESHAP	National Emission Standards for HAP

WWT	wastewater treatment
FGD	flue gas desulfurization
SNCR	selective non-catalytic reduction
OFA	over-fire air
EPA	Environmental Protection Agency
SIC	Standard Industrial Classification
CFR	Code of Federal Regulations
PSD	Prevention of Significant Deterioration
NAAQS	National Ambient Air Quality Standards
MON	Miscellaneous Organic Chemical Manufacturing NESHAP
CMAS	Chemical Manufacturing Area Sources
HCl	hydrochloric acid
LDAR	Leak Detection And Repair
BACT	Best Available Control Technology
SER	Significant Emission Rate
RCO	regenerative catalytic oxidizer
SCR	selective catalytic reduction
LAER	Lowest Achievable Emission Rate

fractionation. Zhang et al. (2016) and Bhatt et al. (2017) evaluated criteria and hazardous air emissions and federal air regulatory requirements for biomass converted sugars to hydrocarbons and fast pyrolysis pathways, respectively, but their analyses do not address strategies to mitigate risks associated with permitting and reducing air emissions. While Eberle et al. (2017) assessed the environmental effects of hydrocarbon fuel production via the biochemical and thermochemical biorefinery pathways, the study does not investigate how changes in process design, feedstock and biorefinery scales may affect air emissions and regulatory and permitting requirements. Deploying biorefineries could be risky if additional investment is needed to meet applicable air regulations or if challenges to obtain air permits delay construction.

A biorefinery, considered a chemical production facility, is required to obtain a New Source Review (NSR) permit (EPA, 2022a) under the Clean Air Act (U.S. Code Sec. 7401, 2013; EPA, 2013; 2015a) which protects communities from increases in pollution when a new facility is built or an existing facility is modified. An NSR air permit application requires a biorefinery to estimate potential-to-emit (PTE), which is the maximum capacity of a stationary source to emit a pollutant under its physical and operational design, considering continuous operations. Air pollutants regulated under the NSR program include criteria air pollutants particulate matter (PM); carbon monoxide (CO); sulfur oxides (SO_x); nitrogen oxides (NO_x); ozone, regulated through its precursors, NO_x, and volatile organic compounds (VOCs); and lead, as well as pollutants regulated under the New Source Performance Standards (NSPS) including ammonia (NH₃) and sulfuric acid (H₂SO₄) (EPA, 2016a). Hazardous air pollutants (HAPs) are regulated under the National Emission Standards for Hazardous Air Pollutants (NESHAP) (EPA, 2016b) and are addressed under the Title V program, required under Title V of the Clean Air Act.

Advanced biorefineries are still new, and we lack knowledge about their air pollutant emissions because they employ novel conversion technologies. Therefore, opportunities exist to advance our understanding of how critical factors such as conversion processes, biorefinery size, feedstock type, and energy source may influence the permitting requirements and what strategies may be available to further reduce emissions if needed or desired. The insights gained from this study could also help mitigate investment risks and uncertainties caused by potential permitting delays.

To address such gaps, we perform a detailed air emission analysis for a biorefinery utilizing lignocellulosic feedstock to produce renewable diesel blendstock (RDB) via an aerobic respiration process as described in Davis et al. (2013, 2022). We consider 12 illustrative design

permutations by considering two types of feedstocks (a uniform format blend [UFB] or 100% corn stover), three biorefinery scales (2,000, 5, 200, and 9,100 dry metric tons per day [dmtd] of biomass feedstock), and two uses of lignin (as a boiler fuel or for pellet production). The process variations are detailed in Section 2.1. Although diverting lignin for pellet production reduces air pollutant emissions from the biorefinery when compared to using lignin as a boiler fuel, the boiler likely remains a primary contributor of multiple pollutants. So, we explore six additional design permutations, where electricity demand is met by offsite non-combustion renewable power, to estimate to what extent biorefineries could further reduce emissions after diverting lignin for pellet production.

The objectives of this analysis are to 1) assess the applicability of federal regulations, namely the NSPS and NESHAP, to the 12 process variations employing the aerobic respiration technology to produce RDB, 2) develop preliminary PTE estimates by incorporating federally enforceable limits in the NSPS and NESHAP that potentially apply to the biorefinery, 3) compare the implications of design permutations on air emissions, applicability of federal air regulations, and air permitting requirements, and 4) explore emission reduction technologies and design variations (e.g., adopting renewable electricity to meet power demand), which could further reduce emissions of major source pollutants. Insights from this analysis are expected to help inform process design including emission controls, which may be required to comply with applicable requirements, and to minimize barriers and costs resulting from uncertainties associated with air permit application. However, the results from this analysis should be considered preliminary and applicable to process designs described in Davis et al. (2022). Results for specific projects will vary depending on the actual process configurations and site-specific parameters.

2. Methodology

2.1. Overview of conversion process

Our analysis is based on the process design documented in the Methodology section of Davis et al. (2022) which converts lignocellulosic biomass feedstocks to RDB utilizing a biochemical technology pathway via aerobic respiration. We consider 12 process design variations, as described in Davis et al. (2022) with varying feedstock types, plant scales, and alternative lignin uses. The two feedstock variations considered in the analysis are 1) a UFB feedstock developed by Idaho National Laboratory, and 2) corn stover feedstock (refer to Davis et al.

(2022) for details) (INL, 2014). The three plant scales for the biorefinery consider the base case size of 2,000 dmt of lignocellulosic biomass (UFB or corn stover) in Davis et al. (2022) which is the standard scale for typical biofuel design cases developed by DOE's national laboratories, and then two larger scales at 5,200 dmt and 9,100 dmt (the biorefinery scales refer to biomass feedstock sizes). The latter scales are chosen to correspond to 5th and 10th percentile of petroleum refinery sizes based on inputs to the atmospheric distillation units in the United States from Energy Information Administration (EIA, 2018). For each biorefinery scale, we then consider two alternative lignin uses, 1) lignin utilized as a fuel in the hybrid mixed solid-gas boiler for process steam and electricity production, and 2) lignin diverted for pellet production, as a co-product from the biorefinery. Under the lignin-for-pellet scenario, the off-gases from the hydrotreater and biogas from wastewater treatment (WWT) areas are burned in a gas boiler for process steam and electricity production whereas the wastewater sludge is assumed to be sent off-site for further treatment.

We group the design areas into core, supporting, and miscellaneous operations as shown in Fig. 1 to facilitate illustration of results. Refer to Davis et al. (2022) for a detailed description of each process area. While the core conversion and miscellaneous operations are similar for all process designs, the change in use of lignin considered for the design permutations along with the alternate renewable electricity scenario for the lignin-for-pellet scenario are depicted in Fig. 1.

The current process designs for all design variations include several control technologies, which are 1) one baghouse to reduce PM emissions from feedstock handling and another baghouse for boiler, 2) flue gas desulfurization (FGD) to reduce SO₂ emissions from the boiler, and 3) selective non-catalytic reduction (SNCR) and over-fire air (OFA) system to reduce NO_x emissions from the boiler, as described in Davis et al. (2022).

2.2. Identifying air pollutants and determining applicability of federal regulations

The first step to determine which federal air regulations potentially apply to the biorefinery is to identify the type of regulated air pollutants the biorefinery or specific equipment is likely to emit; some regulations apply to the entire facility while others apply to specific equipment. The design cases, with or without lignin pelletization, and their corresponding Aspen Plus models are used as the basis to analyze each unit operation within the biorefinery (ASPEN, 2010). We utilize U.S. Environmental Protection Agency's (EPA's) Compilation of Air Pollution Emission Factor database (AP-42) (EPA, 1995) as well as available air permits for biorefineries utilizing similar operations to identify the air pollutants the process areas may emit (WLA Consulting Inc, 2011; Burns & McDonnell Engineering Company Inc., 2014). Because the biorefinery is expected to be classified as a "chemical process plant" (EPA, 1998; TCEQ, 2019), which is one of the 28 listed source categories, fugitive emissions must be included in the facility-wide PTE estimation (e.g., Abengoa Bioenergy Biomass of Kansas considered fugitive emissions in their PTE estimation (Burns & McDonnell Engineering Company Inc., 2014)).

Based on the Standard Industrial Classification (SIC) code for the biorefinery and the attributes of unit operation and equipment, we then determine which federal standards under the NSPS and NESHAP would apply to the biorefinery. Eberle et al. (2017) determined that the permits for similar biorefineries utilizing cellulosic sugars to produce hydrocarbon biofuel blendstock would be classified under SIC 286, which corresponds to the industry code for industrial organic chemicals. Because federal standards can apply to the whole facility or to certain unit operations, we review the applicability criteria of each potentially applicable federal standard under the NSPS codified in Title 40, part 60 of the Code of Federal Regulations (CFR) (Federal Register, 2022a) and

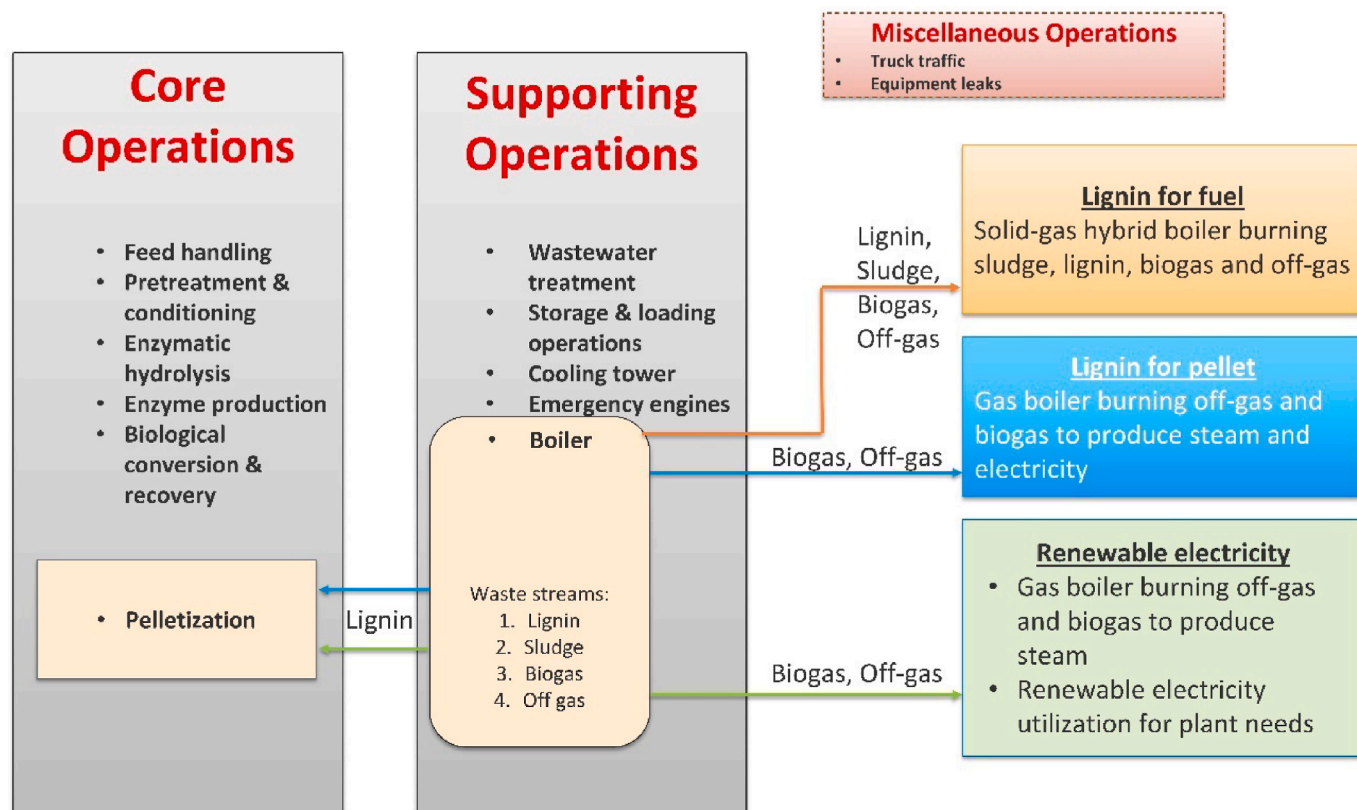


Fig. 1. Schematic diagram of three process variations including a) biorefinery burning lignin for fuel to produce heat and power on-site, b) biorefinery diverting lignin for pellet production that produces heat and power on-site, and c) biorefinery diverting lignin for pellet production that uses 100% renewable electricity.

NESHAP codified in Title 40, Part 61 and 63 of CFR to determine which process units would be subject to a particular regulation (Federal Register, 2022b, 2022c).

2.3. Estimation of potential-to-emit (PTE) and preliminary source classification

In the next step, we estimate PTE for each regulated pollutant likely to be emitted from the biorefinery. PTE is defined as the maximum capacity of a stationary source to emit a pollutant under its physical and operational design (Smith, 1998). In determining the applicability of some federal air emission regulations as well as the construction and operating permitting requirements for a planned source, PTE must always be considered, although other criteria (e.g., whether the biorefinery is to be located in a non-attainment area), may also play a role. To estimate PTE, we utilize several approaches and data sources, including emissions factors from EPA's AP-42 database (EPA, 1995), mass balance based on Aspen Plus model (ASPEN, 2010), source-specific models such as EPA's TANKS 4.09d (EPA, 2006), emission estimates from air permits or air permit applications, and stack test data when available. Table 1 lists the primary approaches and data sources utilized to estimate the PTE for operations within the biorefinery. Refer to Supplementary Information (SI) for a detailed description of emission factors and other data sources utilized for estimating PTE.

As indicated in Table 1, two primary differences between the lignin-for-fuel and lignin-for-pellets scenarios are 1) the type of boiler and 2) the addition of pelletizer(s) when lignin is pelletized. When lignin is used as one of the fuel streams, a hybrid solid-gas boiler is employed, which burns a mixture of off-gas, lignin, biogas, and sludge from WWT.

Table 1

Data sources and approach for estimating the PTE from different emission sources of the biorefinery.

Emission points	Emission source for process variation		PTE estimation approach	Data sources
	Lignin for fuel	Lignin for pellets		
Core operations	Feed handling		Emission factor	Air permits
	Pretreatment and conditioning		Mass balance	Process model, and engineering judgement
	Enzymatic hydrolysis		Mass balance	Process model
	Enzyme production		Mass balance	Process model
	Biological conversion and recovery		Emission factor	EPA's AP-42 database
Supporting operations	Not applicable	Lignin pelletizer	Emission factor	EPA's AP-42 database
	Wastewater treatment		Mass balance	Process model
	Storage and loading operations		Emission factor	EPA's AP-42 database and EPA's TANKS 4.09D software
	Hybrid solid-gas boiler	Gas boiler	Emission factor and mass balance	EPA's AP-42 database and process model
	Cooling tower		Emission factor	Air permits and South Coast Air Quality Management District's Guidelines for Cooling Towers
Equipment leaks	Emergency engines		Emission factor	EPA's AP-42 database
	Equipment leaks		Emission factor	EPA's protocol for leak emission estimates, air permits, and engineering judgement
Truck traffic	Truck traffic		Emission factor	EPA's AP-42 database

On the other hand, when lignin is diverted for pellet production, a gas boiler is used to combust off-gas and biogas to produce steam, which satisfies the facility's process demand for both heat and electricity. The lignin-for-pellet scenario also assumes wastewater sludge will be sent for off-site treatment. When lignin is diverted, pelletizer(s) will be employed on-site to manufacture pellets as a co-product in the biorefinery.

We then develop two sets of PTE estimates, with the first set referred to as baseline PTE, or uncontrolled PTE, and the second set as preliminary PTE. The baseline PTE is the uncontrolled emissions and does not consider any emission reductions from control technologies. The preliminary PTE considers emission reductions resulting from the installed control technologies, along with emission reductions resulting from the compliance requirements (e.g., emission limits and work practice standards) in regulations, to which the biorefinery is expected to be subject. We assume applicable emission limits and emission reduction efficiencies (by control technologies, work practice standards) are used to estimate the preliminary PTE, assuming the emission controls and the compliance requirements are made federally enforceable in a permit.

It should be noted that we do not consider any additional state or local regulations to which the biorefinery may be subject, which may impose additional operating requirements before a permit can be approved.

2.4. NSR permitting and preliminary source determination

PTE is used to determine the construction and operating permitting requirements for a planned source. For a hypothetical biorefinery analyzed in this study, we utilize the preliminary PTE estimate to determine the permitting requirements for the biorefinery under the NSR program.

The biorefinery would need to apply for a construction permit under the NSR program before starting construction. The NSR permit is a legal document by which the facility must abide. There are three types of NSR permitting requirements; the source may have to meet one or more of these permitting requirements. The three types of NSR requirements are (EPA, 2022a).

1. Prevention of Significant Deterioration (PSD) permits, which are required for new sources that are major for at least one pollutant for which the area complies with the National Ambient Air Quality Standard (NAAQS)
2. Nonattainment NSR permits, which are required for new sources that are major for a pollutant for which the area does not comply with the NAAQS
3. Minor source permit.

We compare the preliminary PTE with threshold values to determine the type of permit applicable to the biorefinery. In addition to NSR, all major sources are required to obtain an operating permit, also known as Title V operating permit (EPA, 2015b). Refer to the SI for details.

3. Results

Based on the review of unit operations for each process area in the design cases considered, we find the biorefinery producing RDB via aerobic respiration would emit all the criteria air pollutants or their precursors, which include PM, PM₁₀, PM_{2.5}, NO_x, CO, SO₂, VOCs, and lead plus other NSR-regulated pollutants, ammonia (NH₃) and sulfuric acid (H₂SO₄), in particular. Table 2 lists the pollutants likely to be emitted from each process area or as fugitive emissions, for the lignin-for-fuel and lignin-for-pellet design scenarios. We group the emission points by core operations, supporting operations, and fugitive emissions (truck traffic and equipment leaks) to facilitate illustration and discussion of the results (refer to Fig. 1).

Table 2
Pollutants likely to be emitted from each emission point of the biorefinery.

Emission points	Emission source for process variation		Pollutants likely to be emitted
	Lignin utilized as a fuel	Lignin utilized for pellets	
Core operations	Feed handling		PM, PM ₁₀ , PM _{2.5}
	Pretreatment and conditioning		VOCs, NH ₃ , H ₂ SO ₄
	Enzymatic hydrolysis		None
	Enzyme production		SO ₂ , NH ₃ , CO ₂
	Biological conversion and recovery		PM, PM ₁₀ , PM _{2.5} , NO _x , CO, SO ₂ , VOCs, lead, HAPs, CO ₂
	Not applicable	Lignin pelletizer	PM, PM ₁₀ , PM _{2.5} , VOCs, HAPs
Supporting operations	Wastewater treatment		VOCs, HAPs
	Storage and loading operations		VOCs, HAPs, H ₂ SO ₄
	Hybrid solid-gas boiler	Gas boiler	PM, PM ₁₀ , PM _{2.5} , NO _x , CO, SO ₂ , VOCs, lead, HAPs, NH ₃ , H ₂ SO ₄ , CO ₂
	Cooling tower		PM, PM ₁₀ , PM _{2.5} , VOCs, HAPs
	Emergency engines ^a		PM, PM ₁₀ , PM _{2.5} , NO _x , CO, SO ₂ , VOCs, HAPs, CO ₂
Equipment leaks	Equipment leaks		VOCs HAPs
Truck traffic	Truck traffic		PM, PM ₁₀ , PM _{2.5}

^a Emergency engines include emergency generator and emergency fire pump.

HAPs are also expected to be emitted from the biorefineries. Sufficient information from the design cases, permits, or literature is not readily available to allow us to determine speciated HAPs for emissions from the cooling tower, enzymatic hydrolysis, biological conversion, equipment leaks, and WWT. We tried to identify speciated HAPs for the boiler, loading operation, storage tanks, and emergency engines, based mainly on EPA’s AP-42 and material safety data sheets for similar products (e.g., diesel fuel). The determination of speciated HAPs should be considered preliminary in this study because of the lack of testing data needed to verify the existence of any species.

3.1. Applicability of federal regulations

We first identify NSPS and NESHAP (Code of Federal Regulations,

1987, 1993, 2003, 2006, 2007a, 2007b, 2007c, 2009, 2011a, 2011b, 2004) that could be relevant to the equipment and operations associated with the biorefinery (refer to SI for details). We find that the feedstock types and facility sizes do not affect which NSPSs and NESHAPs would be applicable to a specific design permutation. However, whether the lignin is used as a boiler fuel or for pellet production may affect the applicability of specific regulatory requirements. Based on the specific process parameters and unit equipment and operations described in the design cases, we determine the NSPS and NESHAPs that the biorefinery is anticipated to be subject to. These are summarized in Table 3. Because we lack detailed descriptions of attributes or parameters for some equipment (e.g., emergency pumps) and lack emission testing data for some processes, our evaluation of NSPS and NESHAP applicability should not be considered comprehensive or conclusive. Biorefinery developers and owners will need to review all NSPS and NESHAP themselves to determine applicability when applying for air permits. Despite this limitation, the proposed approach provides an important first step for biorefinery developers to accelerate the permitting process.

Because the biorefinery producing RDB is considered a chemical processing facility, one of the two chemical manufacturing NESHAPs is expected to apply to the biorefinery. These two NESHAPs are: Miscellaneous Organic Chemical Manufacturing (40 CFR 63, Subpart FFFF), commonly referred to as the “MON”, and Chemical Manufacturing Area Sources (40 CFR 63, Subpart VVVVVV), commonly referred to as “CMAS”. Subpart FFFF and Subpart VVVVVV are mutually exclusive because a facility will be classified either as a major or an area source of HAPs based on the PTE of HAP emissions. Refer to SI for details on applicability criteria for MON and CMAS. When information is unavailable, we make conservative assumptions to ensure the PTE estimates take into account worst-case scenarios.

3.2. Estimated PTE and permitting classifications

3.2.1. Baseline PTE

The baseline PTE is estimated without taking into consideration any emission controls employed in the current process designs or emission limits imposed by potentially applicable federal regulations. Because the biorefinery examined is a chemical production facility, which is one of the 28 listed source categories, fugitive emissions (e.g., from equipment leaks) are included in the facility-wide PTE estimation. Fig. 2 illustrates

Table 3
NSPS and NESHAP that apply to the unit operations for different process variations at the biorefinery.

Lignin-for-fuel scenario				Lignin-for-pellet scenario			
Emission point	Potential federal regulation	Applicability criteria	Target pollutants	Emission point	Potential federal regulation	Applicability criteria	Target pollutants
Boiler	NSPS Subpart Db	Heat input capacity > 100 MMBtu/hr.	SO ₂ , PM, and NO _x	Same as lignin-for-fuel scenario			
	NESHAP Subpart DDDDD Or NESHAP Subpart JJJJJJ	Facility is a major source of HAPs or minor source of HAPs	CO, PM, HCl, and HF				
Emergency fire pump and generator	NSPS Subpart IIII	Applies to all new internal combustion engines	PM, Non-methane hydrocarbons + NO _x	Same as lignin-for-fuel scenario			
	NESHAP Subpart ZZZZ	Applies to reciprocating ICES at all major or area source of HAPs	HAPs				
Cooling tower	NESHAP Subpart FFFF or NESHAP Subpart VVVVVV	Facility is a major source of HAPs or Minor source of HAPs ^a	HAPs	Same as lignin-for-fuel scenario			
Equipment leaks	NESHAP Subpart FFFF or NESHAP Subpart VVVVVV	Facility is a major source of HAPs or minor source of HAPs	HAPs	Same as lignin-for-fuel scenario			
Not applicable				Pelletizer	NESHAP Subpart FFFF or NESHAP Subpart VVVVVV	Facility is a major source of HAPs or minor source of HAPs	HAPs

^a In addition to major or minor source of HAPs applicability, the cooling tower is also assumed to have at least 5% (by weight) of total HAP concentration for the NESHAP to be applicable.

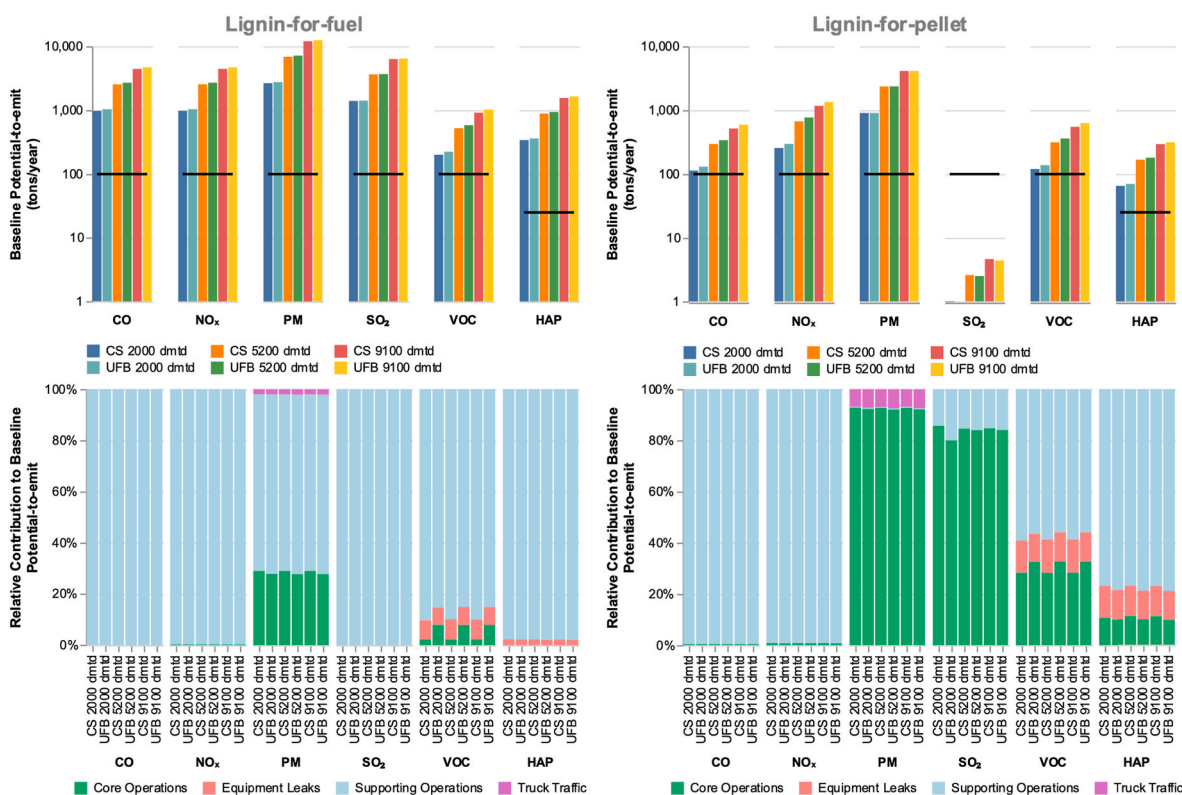


Fig. 2. Baseline PTE for two lignin utilization variations processing either UFB or corn stover feedstock at a plant scale of 2,000 dmt, 5,200 dmt, and 9,100 dmt. The top panels show the baseline PTE of several regulated air pollutants for six design permutations for each lignin utilization variation that includes lignin utilized as a fuel and lignin utilized for pellet production. The black solid line represents the major source threshold value (100 tpy) for criteria air pollutants and HAPs (25 tpy). The bottom panels show the relative contribution of each emission point to the overall PTE. Note that the y-axes in the top panels are plotted on a log-scale.

the uncontrolled emissions and relative contribution of grouped emission points.

While not explicitly shown here, the baseline PTE of PM₁₀ and PM_{2.5} (refer to PTE excel spreadsheet in SI for details) are estimated to be 92% and 60% of the baseline PTE for PM, respectively, for the lignin-for-fuel scenario. In the lignin-to-pellet scenario, the PTE of PM₁₀ and PM_{2.5} are 93% and 19% of that for PM, respectively. Also, the PTE of lead is less than 1 ton per year for both lignin utilization variations (see SI for details). Moreover, the required input of SO₂ in the enzyme production area is higher in corn stover than UFB feedstock in the lignin-for-pellet scenario for a 2,000 dmt biorefinery size while the value changes for large biorefineries where UFB has a higher SO₂ input rate than corn stover.

As currently estimated, the baseline PTE of all regulated air pollutants are above their respective major source thresholds (Fig. 2) for a source in an attainment area when lignin is used as a boiler fuel. The boiler, a supporting operation, is a major contributor of all regulated air pollutants in the lignin-for-fuel scenario except for VOCs, for which the cooling tower contributes to the majority of emissions (refer to SI for details on emissions from each emission point).

In the lignin-for-pellet scenario, the baseline PTE of all regulated air pollutants except for SO₂ is above their respective major source threshold (Fig. 2). The boiler contributes to the majority of CO, NO_x, VOC, and HAP emissions while the cooling tower is the major contributor of both VOC and HAP emissions. For SO₂, which is below the major source threshold, enzyme production results in the highest emissions compared to other emission sources. Feed handling, a core operation, contributes to the majority of PM emissions.

As shown in Fig. 2, the overall emissions from a same-size biorefinery utilizing lignin as boiler fuel are higher as compared to the one diverting lignin for pellet production. That is because lignin accounts for more than 40% of heat input to the hybrid solid-gas boiler burning lignin

along with other waste streams. The low SO₂ emissions from the lignin-for-pellet scenario is because of the low sulfur content in the fuel burned in the boiler. The variations in feedstock composition do not significantly affect emissions, although overall emissions for corn stover are slightly lower than for UFB feedstock.

3.2.2. Preliminary PTE

We then estimate the preliminary PTE, taking into consideration emission reductions resulting from the emission controls and additional emission limits required by applicable NSPS and NESHAP sections. Because the baseline PTE suggests the biorefinery is likely a major source for HAPs, the preliminary PTE also accounts for emission reductions required by NESHAP that apply to a major source of HAPs (e.g., Subpart FFFF). Table 4 shows the regulatory requirements and limits to which the biorefinery is expected to be subject, as well as whether the current design cases already comply with a specific requirement or limit when we account for the emission controls in the design cases, shown in the last column of Table 4. Unlike the baseline PTE, for the preliminary PTE, we consider all the emission reductions from implementing the emission limits and practices shown.

As shown in Table 4, regulatory requirements and limits imposed by applicable NSPS and NESHAP vary by pollutant and lignin utilization variation:

- 1) The boiler is subject to boiler NSPS (40 CFR 60, Subpart Db) and NESHAP (40 CFR 63, Subpart DDDDD), which regulate filterable PM emissions to 0.1 lb/MMBtu and 0.03 lb/MMBtu of heat input to the boiler, respectively, when lignin is utilized as a fuel. In the lignin-for-pellet scenario, the boiler is subject to boiler NSPS (40 CFR 60, Subpart Db), which regulates filterable PM emissions to 0.1 lb/MMBtu of heat input to the boiler. However, the boiler NESHAP Subpart DDDDD does not have a specific emission limit for PM for a

Table 4

Summary of federal regulations that apply to the two lignin utilization variations at the biorefinery.

Process variation	Pollutant	Applicable federal regulations	Affected equipment	Emission limits	Base design already in compliance?
Lignin utilized as a fuel	PM (filterable)	NSPS Subpart Db	Boiler	0.1 lb/MMBtu	✓
		NESHAP Subpart DDDDD		0.03 lb/MMBtu	
	SO ₂	NSPS Subpart Db	Boiler	0.2 lb/MMBtu	✓
	HCl, HF	NESHAP Subpart DDDDD	Boiler	0.022 lb/MMBtu	✓
	CO	NESHAP Subpart DDDDD	Boiler	0.46 lb of CO/MMBtu	✓
	VOCs, HAPs	NESHAP Subpart FFFF	Cooling tower	Work standard for cooling tower water monitoring	✗
Lignin utilized for pellets	PM (filterable)	NSPS Subpart Db	Equipment leaks Boiler	Leak detection and monitoring, monthly 0.1 lb/MMBtu	✗ ✓
		SO ₂	Boiler	0.2 lb/MMBtu	✓
	None	NESHAP Subpart DDDDD	Boiler	Work standard for annual tune-up of the boiler	✗
	VOCs, HAPs	NESHAP Subpart FFFF	Pelletizer	98% reduction from continuous vents	✗
			Cooling tower	Work standard for cooling tower water monitoring	
			Equipment leaks	Leak detection and monitoring, monthly	

lb/MMBtu = pounds per million British thermal unit.

gas boiler in the lignin-for-pellet scenario. The baghouse employed in the boiler area is anticipated to reduce PM emissions by at least 99% and therefore comply with the limits for both lignin use scenarios. Refer to SI for details.

- The boiler NSPS section requires SO₂ to be below 0.2 lb/MMBtu, which would be achieved through FGD for both lignin use scenarios.
- The boiler in the lignin-for-fuel scenario is subject to major source NESHAP (40 CFR 63, Subpart DDDDD), which limits hydrogen chloride (HCl) to 0.022 lb/MMBtu of heat input. The HCl limit is meant to be a surrogate for all acid gas emissions from the boiler. HCl and hydrogen fluoride (HF) are the only two acid gases expected to be emitted from the boiler. The acid gas emissions reduction can be achieved by the FGD system, which can effectively mitigate HCl and HF emissions. Assuming a 98% reduction in HCl emissions and 88% reduction in HF emissions (Staudt, 2011), the emission can be reduced to 0.0015 and 0.00013 lb/MMBtu, respectively, for HCl and HF (i.e., below the NESHAP limit of 0.022 lb/MMBtu). In the lignin-for-pellet scenario, only the off-gases from the biorefinery and biogas from WWT is burned in the boiler, which requires an annual tune-up as a work practice standard.
- In the lignin-for-fuel scenario, the boiler NESHAP Subpart DDDDD regulates CO as a surrogate for HAP control, limiting it to 0.46 lb/MMBtu. The design case does not specify the type of boiler. We assume it is a stoker-type of boiler based on Humbird et al. (2011) For this type of boiler, the CO emissions are limited to 0.58 lb/MMBtu of steam generated. Based on a boiler efficiency of 80% (Davis et al., 2013), this would equate to 0.46 lb of CO/MMBtu of heat input. This limit is expected to be met by a new boiler that is properly designed and tuned. In the lignin-for-pellet scenario, no specific limit is imposed by regulations for CO when the gas boiler is used. However, an annual boiler tune-up is required as a standard work practice.

Because the design cases are intended to be feasibility-level studies, they do not explicitly specify or consider work practices in detail. As shown in Table 4, the biorefineries are subject to certain work practice standards required to control emissions from cooling tower and equipment leaks to comply with Subpart FFFF for both the lignin-for-fuel and lignin-for-pellet scenarios. For the cooling tower, a source testing of water and repairing any leaks indicated by higher concentrations of HAPs in the water are required to comply with the Subpart FFFF requirement. Through minimizing leaks into the cooling tower, the biorefinery is expected to reduce VOCs by about 88%, from 6 to 0.7 lb VOCs/million gallons of cooling water (South Coast Air Quality Management District, 2006) (see SI for details). The leak monitoring practice could reduce the PTE of HAPs from the cooling tower by 50% assuming

50% of VOCs at the biorefinery are HAPs (Zhang et al., 2016; Eberle et al., 2017). Similarly, for equipment leaks, a monthly leak detection and repair (LDAR) program for monitoring and repairing leaking equipment (e.g., valves, connectors, pumps) will be needed to meet the Subpart FFFF compliance requirement. EPA's LDAR guide details the elements and practices that can be used to increase the effectiveness of such a program (EPA, 2007).

When lignin is diverted for pellet production, the lignin pelletizer has a continuous vent stream that meets Subpart FFFF applicability criteria and will be subject to emission control. To comply with Subpart FFFF 98% VOC and HAP reduction requirement, the biorefinery can consider adopting a thermal or catalytic incinerator, which could reduce 98% of VOCs and HAPs from the dryer vent. The emission controls and work practices for compliance with the federal regulatory requirements are summarized in the SI.

Fig. 3 shows the estimated preliminary PTE for 12 design permutations after incorporating federally enforceable emission controls as well as regulatory requirements applicable to the biorefinery.

Unlike baseline PTE, the preliminary PTE of PM₁₀ and PM_{2.5} (refer to PTE excel spreadsheet in SI for details) are estimated to be 49% and 22%, respectively, of that for PM in the lignin-for-fuel scenario. For the lignin-for-pellet scenario, the PTE of PM₁₀ and PM_{2.5} are 43% and 15%, respectively, of that for PM. For lead, the preliminary PTE is less than 1 ton per year for both lignin utilization variations (see SI for details), similar to baseline PTE.

As shown in Fig. 3, the preliminary PTE of at least four regulated air pollutants is above the major source threshold for both lignin utilization variations if the biorefinery is to be in an attainment area. For a 2,000 dmt/d biorefinery using lignin as a fuel, the preliminary PTE of CO, NO_x, SO₂, VOCs, and HAPs exceeds their respective major source threshold. For a 2,000 dmt/d biorefinery producing pellets, the preliminary PTE of CO, NO_x, PM, and HAPs exceeds their respective major source threshold. For larger biorefineries (i.e., 5,200 and 9,100 dmt/d), the PTE of PM, CO, NO_x, and HAPs is estimated to exceed the major source threshold value, regardless of how lignin is utilized. The PTE of SO₂ is below the major source threshold when lignin is diverted for pellets whereas the PTE of SO₂ exceeds the threshold when lignin is used as fuel. Estimated VOC emissions exceed the major source threshold for larger biorefineries (i.e., 5,200 and 9,100 dmt/d) burning or diverting lignin with one exception being the 5,200 dmt/d biorefinery, which utilizes corn stover as a feedstock and diverts lignin. The PTE of NH₃ and H₂SO₄ is below their respective major source threshold for all permutations with one exception where the PTE is estimated at 100 tpy when the biorefinery processes 9,100 dmt/d of UFB feedstock and uses lignin as a fuel to the boiler. Refer to PTE excel spreadsheet in the SI for details on other regulated

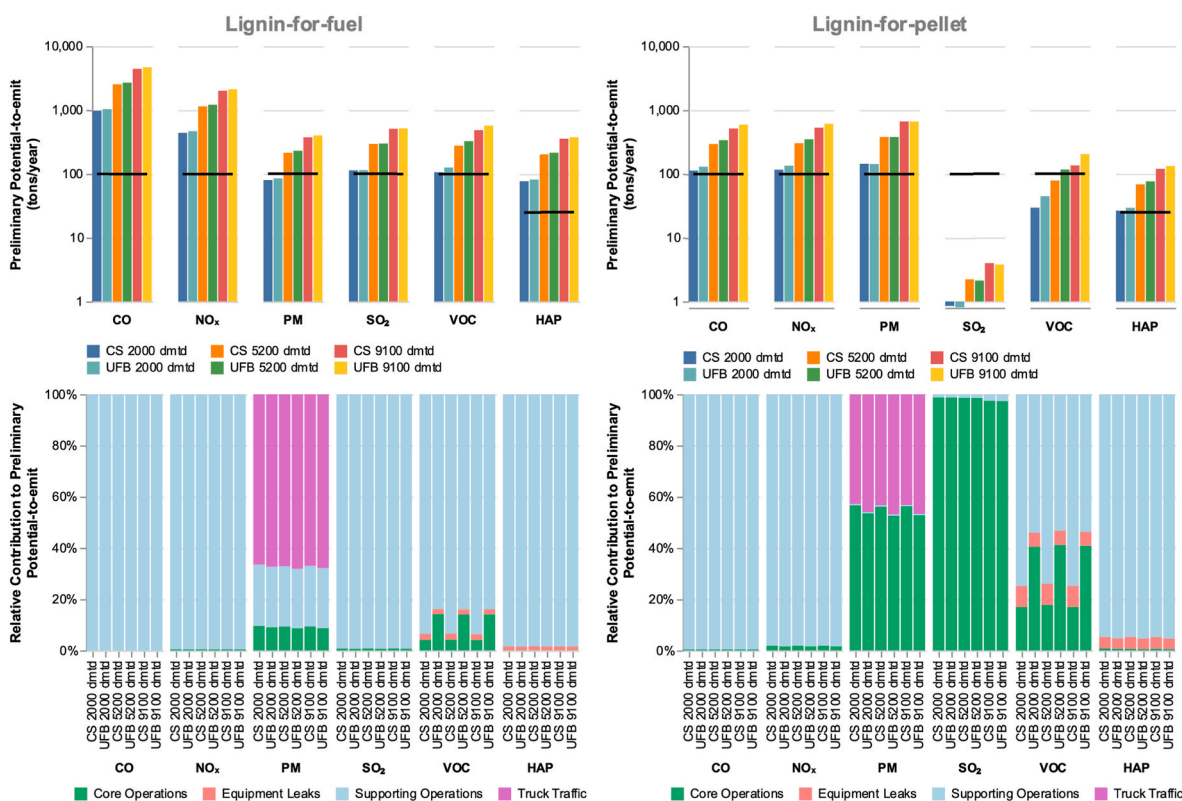


Fig. 3. Preliminary PTE for two lignin utilization variations processing either UFB or corn stover feedstock at a plant scale of 2,000 dmt/d, 5,200 dmt/d, and 9,100 dmt/d. The top panels show the preliminary PTE of several regulated air pollutants for six design permutations for each lignin utilization variation that includes lignin utilized as a fuel and lignin utilized for pellet production. The black solid line represents the major source threshold value (100 tpy) for criteria air pollutants and HAPs (25 tpy). The bottom panels show the relative contribution of each emission point to the overall preliminary PTE. Note that the y-axes in the top panels are plotted on a log-scale.

pollutants (e.g., PM_{10} , $PM_{2.5}$, and lead).

Incorporating applicable federal regulatory requirements and limits significantly reduces potential emissions. In the lignin-for-fuel scenario, the preliminary PTE of PM, SO_2 , and HAPs are 97%, 92%, and 77% lower, respectively, as compared to the baseline PTE. For NO_x and VOCs, the reductions from baseline PTE are estimated at 55% and 44%, respectively. The baseline and preliminary PTE for CO remains unchanged because it is assumed the new boilers will meet the emission limit with proper design and tuning.

For the lignin-for-pellet scenario, the emissions of PM, VOCs, and HAPs are reduced by 84%, 68%, and 58%, respectively, when compared to the baseline PTE. The reductions in VOC and HAP PTE are greater in the lignin-for-pellet scenario than the lignin-for-fuel scenario because of the Subpart FFFF requirement, which reduces 98% of additional VOCs and HAPs from the dryer vent in the pelletization area. For NO_x , the preliminary PTE is 55% lower than the baseline because of including SNCR and OFA control. The preliminary PTE of SO_2 is reduced by about 13% from its baseline because of including FGD control. Similar to the lignin-for-fuel scenario, the preliminary PTE for CO is identical to its baseline PTE.

The boiler is the largest contributor to CO, NO_x , VOC, and HAP emissions in both lignin use variations. In the lignin-for-fuel scenario, truck traffic is the single largest PM emission source whereas the pelletizer, a core operation, is the largest contributor of PM emissions when lignin is utilized for pellet production. For SO_2 emissions, the boiler is the largest contributor in the lignin-for-fuel scenario while enzyme production makes up most of the SO_2 emissions in the lignin-for-pellet scenario. Similar to the baseline PTE, the feedstock composition does not significantly affect emissions with overall emissions being slightly lower using corn stover than UFB except for SO_2 emissions. In the lignin-for-pellet scenario, SO_2 requirement in the enzyme

production area is lower using UFB as compared to corn stover. Table 5 summarizes the percentage reduction in preliminary PTE when compared to the baseline PTE for each pollutant.

Our results suggest diverting lignin for pellet production is an effective option for reducing air emissions and could lower the PTE of several regulated pollutants including CO, NO_x , SO_2 , VOCs, HAPs, and H_2SO_4 mainly because of two factors: 1) the lower heat input to the boiler, with the boiler burning gases in the lignin-for-pellet scenario vs. burning a combination of biomass waste in the lignin-for-fuel scenario and 2) being compliant with additional Subpart FFFF requirements applicable to the dryer vent from the pelletizers. The PTE of CO, NO_x , and SO_2 in the lignin-for-pellet scenario is 87–88%, 71–73%, and 99% lower, respectively, relative to the lignin-for-fuel scenario across feedstock types and biorefinery scales. Similarly, the reductions in VOC and HAP emissions in the lignin-for-pellet scenario range from 64–72% and 64–66%, respectively, when compared to the lignin-for-fuel scenario.

4. Discussion

4.1. Permitting strategy

Our estimates of preliminary PTE are developed based on the best available information, including Aspen Plus design of the process, emission factors from EPA's AP-42 and guidelines such as LDAR (EPA, 2007), air permits and applications of similar unit operations. However, uncertainties are inherent in our estimates because of the lack of specific design parameters for certain equipment such as storage tanks, species of VOC and HAP pollutants, and testing data, all of which are required for more accurate quantification. In the absence of this data, we make conservative assumptions based on our best engineering judgement. As such, the PTE is considered preliminary and should be refined when

Table 5
Summary of major findings from preliminary PTE estimation.

Pollutants	Lignin utilized as a fuel				Lignin utilized for pellet production			
	Corn Stover		UFB		Corn stover		UFB	
	Percent below baseline PTE	Above major source for three scales?	Percent below baseline PTE	Above major source for three scales?	Percent below baseline PTE	Above major source for three scales?	Percent below baseline PTE	Above major source for three scales?
CO	0%	Yes	0%	Yes	0%	Yes	0%	Yes
NO _x	55%	Yes	55%	Yes	54%	Yes	54%	Yes
PM	97%	Yes, except for 2,000 dmt/d	97%	Yes, except for 2,000 dmt/d	84%	Yes	84%	Yes
SO ₂	92%	Yes	92%	Yes	13%	No	14%	No
VOCs	47%	Yes	44%	Yes	75%	Yes, except for 2,000 and 5,200 dmt/d	68%	Yes, except for 2,000 dmt/d
HAPs	77%	Yes	77%	Yes	59%	Yes	58%	Yes

facility- and equipment-specific information is available.

Our preliminary PTE estimates account for federally enforceable requirements and limitations potentially applicable to the biorefinery as well as the emission controls in the design case and indicate the biorefinery would be considered a major source under the NSR program because at least one NSR-regulated non-GHG pollutant (e.g., NO_x) exceeds the major source threshold even for a 2,000 dmt/d facility, which diverts lignin for pellet production (Fig. 3). Assuming the biorefinery is to be located in an attainment area, the NSR program would require biorefinery to install the best available control technology (BACT) and conduct a PSD review for each pollutant, whose PTE exceeds the significant emission rate (SER). Refer to the SI for additional requirements under PSD and nonattainment NSR permit.

If a biorefinery prefers to avoid being classified as a major source, two options are generally available to reduce the PTE of the pollutants for which the facility is a major source; the biorefinery could consider 1) adopting additional emission controls or opting for more advanced and efficient control technologies than those in the current design case and/ or 2) requesting a federally enforceable limit for the pollutants.

4.1.1. Additional control options available for major source pollutants

For the 12 design permutations evaluated, we explore additional potential emission controls for each pollutant whose PTE is estimated to exceed the major source threshold. We discuss whether adopting additional emission controls may reduce the PTE of the pollutants to below their corresponding major source thresholds. We estimate potential emission reduction that could be achieved by adopting certain emission controls, to illustrate whether it may be feasible to reduce the PTE of the pollutant to below its major source threshold. If it is not possible to avoid being a major source, the biorefinery might prefer to reduce the PTE below the SER limit for as many pollutants as possible. The facility can avoid conducting a PSD review for a regulated pollutant if the PTE of that pollutant is lower than the SER. A PSD review could be time- and effort-consuming because it requires 1) installation of BACT, 2) an air quality analysis, 3) additional impacts analysis (e.g., the impacts of air pollution on soils, vegetation, and visibility), and 4) public involvement (EPA, 2022b). Note that the emission reduction efficiencies of control technologies vary by operational conditions. The examples provided here are for illustration purposes only. We are transparent regarding data sources and specific assumptions used to estimate the emission reductions.

1) CO

The boiler is the dominating CO contributor, accounting for more than 95% of facility-wide PTE. A precious metal-based regenerative catalytic oxidizer (RCO) could destroy up to 98% of CO (EPA, 2003a). Assuming a 98% CO reduction efficiency, an RCO has the potential to reduce the PTE of CO below the major source threshold and the SER, 100

tpy for CO, for all 12 design permutations with the highest PTE estimated at about 99 tpy for the biorefinery, which processes 9,100 dmt/d and uses lignin as a fuel. It should be noted that the feed stream to the RCO will need to be evaluated thoroughly because certain compounds (e.g., PM and its fractions) present in the stream may cause catalyst deactivation (EPA, 2003a), resulting in much lower CO reduction efficiency by the RCO.

2) NO_x

The boiler is the main NO_x emission source in the biorefinery (Fig. 3). The SNCR and OFA currently planned in the design case are estimated to reduce NO_x emissions from the boiler by 55%. A selective catalytic reaction (SCR) system has higher NO_x reduction efficiency and is capable of reducing NO_x in the range of 70–90% (EPA, 2003b), depending on the NO_x level in the stream and other factors (e.g., the compounds present in the stream). Using an SCR in lieu of a SNCR has the potential to reduce the PTE of NO_x to below major source threshold for the biorefineries, which process 2,000 and 5,200 dmt/d of either feedstock and divert lignin for pellet production, assuming a 90% NO_x reduction efficiency from using a combination of a SCR and OFA. However, replacing a SNCR with a SCR will not help biorefineries avoid being subject to a major source if lignin is used as a fuel, regardless of the scale and feedstock used.

If the biorefinery is already a major source for another NSR-regulated non-GHG pollutant, adopting a combination of a SCR and OFA could reduce the PTE of NO_x below the SER only for a biorefinery processing 2,000 dmt/d of feedstock and diverts lignin for pellet production. A larger biorefinery processing 5,200 or 9,100 dmt/d will be subject to a PSD review for NO_x even with both a SCR and OFA.

It is important to note that if the new boilers from the manufacturers are equipped with low NO_x burners capable of reducing 80% of NO_x emissions (EPA, 1999), the PTE of NO_x for a 2,000 dmt/d biorefinery processing corn stover or UFB can be reduced to below major source threshold. However, for larger biorefinery (5,200 or 9,100 dmt/d), a low NO_x burner alone does not provide sufficient NO_x reduction to make the biorefinery a minor source for NO_x.

3) PM, PM₁₀, PM_{2.5}

As shown in Fig. 3, truck traffic is responsible for more than 66% of facility-wide PM emissions for biorefineries using lignin as a boiler fuel. When lignin is diverted, lignin pelletization emits more than 48% of facility-wide PM across design permutations followed by truck traffic.

In estimating PM emissions from truck traffic, we assume half the roads within biorefinery boundaries are paved with the other half unpaved. Paving all roads could be an option to reduce PM emissions; however, this option alone is not sufficient to lower PM emissions to below major source threshold for the 5,200 and 9,100 dmt/d

biorefineries using lignin as a boiler fuel (note that the 2,000 dmtD biorefinery does not exceed the PM, PM₁₀, and PM_{2.5} major source threshold). For the 5,200 dmtD biorefinery using lignin as a fuel, paving all the roads would reduce the emissions of PM₁₀ below major source threshold while the 9,100 dmtD biorefinery size cannot achieve reductions to below 100 tpy with this control alone. The emissions of PM_{2.5} for this process design is already below major source for all biorefinery sizes.

For a 2,000 dmtD biorefinery that diverts lignin for pellet production, PM could be further reduced by more than 99% by installing a baghouse or similar technology (e.g., electrostatic precipitator) in the pelletization area to capture emissions from both the drying and pelletizing processes (EPA, 2003c; 2003d). Assuming a 99% PM reduction efficiency using a baghouse, the emissions of PM could be reduced to below major source threshold. If a 5,200 or 9,100 dmtD biorefinery adopts a baghouse for pelletization and paves all roads to reduce truck traffic emissions, it is not expected to lower PM emissions enough to be below major source threshold, making it difficult to achieve minor source classification. Adopting these emission controls (baghouse for pelletization and paved road for truck traffic) can reduce PM_{2.5} to below major source threshold for both biorefinery sizes. However, the PTE of PM₁₀ can be reduced below 100 tpy for only the 5,200 dmtD biorefinery size while the 9,100 dmtD biorefinery still exceeds the major source threshold for PM₁₀.

If the biorefinery is a major source for another NSR-regulated non-GHG pollutant, adopting these controls would reduce the PTE of PM, PM₁₀, and PM_{2.5} below the SER only for a biorefinery processing 2,000 dmtD of either feedstock that also diverts lignin for pellet production. For a large biorefinery, 5,200 or 9,100 dmtD, that diverts lignin and for all biorefineries (all three sizes) using lignin as a fuel, the biorefineries would be subject to a PSD review for PM, PM₁₀ and PM_{2.5}, despite implementing the additional emission controls.

4) SO₂

When biorefineries divert lignin for pellet production, enzyme production is the primary SO₂ emission source, accounting for 97% of facility-wide SO₂ emissions. Without burning lignin in the boiler, biorefineries using either corn stover or UFB at all three sizes will not exceed the major source threshold for SO₂. The estimated PTE is also below the SER for SO₂; therefore, the biorefinery will not be subject to PSD review for SO₂ when lignin is pelletized.

On the other hand, biorefineries burning lignin as a fuel will need additional SO₂ mitigation strategies to reduce PTE to below major source threshold. The current design case incorporates a FGD system with a 92% emission control efficiency. SO₂ reduction efficiencies up to 98% are achievable using the most efficient wet scrubber systems (EPA, 2003e). If the biorefinery opts for a wet scrubber with 98% SO₂ reduction efficiency, the facility can reduce SO₂ to below the major source threshold for the two smaller sizes (i.e., 2,000 and 5,200 dmtD). However, even the most efficient wet scrubbers are not expected to lower SO₂ emission to below the major source threshold for the largest biorefinery, 9,100 dmtD, based on our preliminary PTE estimates. In addition, the most efficient wet scrubber has the potential to bring the PTE below the SER for SO₂ for a 2,000 dmtD facility to avoid PSD review, though a 5,200 dmtD biorefinery would be subject to PSD review for SO₂ even with a high-efficiency wet scrubber.

5) VOCs

The boiler is the largest emission source for VOCs in the biorefineries utilizing UFB feedstock, accounting for 68% when lignin is used as a boiler feed and 32% when lignin is diverted for pellets. In biorefineries using corn stover, the boiler contributes an estimated 76% of facility-wide VOC emissions when lignin is utilized as a fuel and accounts for 25% of total VOCs when lignin is diverted for pellet production. Wet scrubbers such as spray towers or packed towers are often used to

remove VOCs from flue gas and are capable of removing 50–95% of VOCs (EPA, 2003e). Routing emissions from the boiler to wet scrubbers could be a feasible option to reduce VOCs. The cooling tower, equipment leaks, and the WWT plant also contribute to considerable VOC emissions when lignin is diverted for pellet production.

If a 2,000 dmtD biorefinery burning lignin adopts a wet scrubber with a removal efficiency of 90%, its PTE of VOCs can be lowered to below the major source threshold, regardless of which feedstock is used. Although the PTE for VOCs would be below the SER for a 2,000 dmtD biorefinery utilizing corn stover, the PTE will exceed the SER for a same-size biorefinery utilizing UFB feedstock. When the biorefinery size increases to 5,200 dmtD, adding a wet scrubber with 90% VOC removal to a biorefinery using corn stover could bring the PTE of VOCs below its major source threshold, but the PTE will exceed the major source threshold if UFB feedstock is used. For the largest biorefinery at 9,100 dmtD, installing a wet scrubber is not expected to reduce the PTE of VOCs to below major source threshold.

For a 2,000 dmtD biorefinery that diverts lignin for pellet production, the VOCs will not make the facility a major source because the estimated PTE is below the major source threshold. For the same size biorefinery using corn stover, its PTE is below the SER, and therefore the facility will not be subject to PSD review for VOCs even if it is a major source for another NSR-regulated non-GHG pollutant. On the other hand, a 2,000 dmtD biorefinery using UFB feedstock will be subject to PSD review for VOCs if no additional emission control technology or practice is adopted. Installing a wet scrubber to the boiler and monitoring cooling tower and equipment leaks more frequently could potentially bring its PTE to below the SER. For a 5,200 dmtD biorefinery that diverts lignin for pellet production, the PTE for VOCs does not exceed the major source threshold when corn stover is used as the feedstock. However, the PTE of a same size biorefinery would exceed the threshold when UFB is utilized, although the PTE could potentially be further reduced to below the threshold if the biorefinery opts for a wet scrubber (e.g., with 90% efficiency) or further reduces emissions from cooling tower and equipment by increasing monitoring frequency. When the size of the biorefinery increases to 9,100 dmtD, the biorefinery will exceed the major source threshold unless additional emission control technologies are incorporated to reduce VOCs from boiler, pretreatment, cooling tower and equipment leaks.

6) HAPs

The boiler is the largest emission source for HAPs in the biorefineries utilizing UFB feedstock, accounting for 83% of facility-wide HAP emissions when lignin is used as a boiler fuel and 59% when lignin is diverted for pellets. When corn stover is used as a feedstock, the boiler contributes to 83% of total HAP emissions when lignin is utilized as a fuel while it accounts for about 57% of total HAP emissions when lignin is diverted for pellet production. An RCO, which is utilized for CO reduction, is capable of reducing 85–90% of formaldehyde and benzene emissions from the boiler (Roy S., 1999). Routing the vent stream to an RCO could be considered a feasible option to reduce certain HAPs. The WWT plant and cooling tower are also significant HAP emission sources for both lignin process variations. Adopting a steam-stripping or air-stripping control can reduce HAP emissions from the WWT plant by 76–99% and 58–99%, respectively (EPA, 1994).

If a 2,000 dmtD biorefinery that diverts lignin for pellet production adopts an RCO with a HAP (formaldehyde and benzene) removal efficiency of 90% in the boiler area and adopts a steam- or air-stripping control HAP reduction efficiency of 99% from the WWT plant, its PTE of HAPs can be lowered to below major source threshold (25 tpy overall and 10 tpy for individual HAPs), with either feedstock. For large biorefinery sizes processing 5,200 and 9,100 dmtD of feedstock, adding an RCO and a steam- or air-stripping control is not expected to reduce the PTE of HAPs to below major source threshold.

Even for a 2,000 dmtD biorefinery that burns lignin as a fuel,

adopting an RCO control in the boiler process area along with a steam-or air-stripping control at a WWT plant would not be enough to reduce the emissions of HAPs to below major source thresholds. In addition to these controls, the biorefinery would need to implement more frequent monitoring of cooling tower and equipment leaks to potentially bring down its PTE below 25 tpy. As the speciation of HAPs from the cooling tower and equipment leaks are not well understood, testing would be needed to determine the HAP species and assess whether a minor source classification can be achieved. For large-scale biorefineries (5,200 and 9,100 dmt/d), the PTE of HAPs will exceed major source thresholds even if several additional emission controls (e.g., RCO and steam/air stripping) are incorporated into the process areas to reduce emissions.

7) NH₃, H₂SO₄, and lead

In addition to air pollutants discussed above, there are several NSR-regulated pollutants including NH₃, H₂SO₄, and lead whose PTE also needs to be considered when determining the permitting applicability. As shown in the SI, the PTE of NH₃, H₂SO₄, and lead for all the design permutations are below the major source thresholds under NSR permitting.

Based on our findings, the two large biorefinery sizes (5,200 and 9,100 dmt/d) would not likely be able to reduce the PTE of all NSR-regulated pollutant to below their respective major source thresholds even after additional emission control technologies are considered and lignin is diverted. Because the lignin-for-pellet scenario has lower emissions than the lignin-for-fuel scenario, we evaluate an alternative process scenario for the lignin-for-pellet scenario by considering utilizing electricity from a renewable source, discussed in detail in Section 4.2.

4.2. Renewable electricity scenario

While our results suggest that diverting lignin is an effective option to reduce emissions of multiple regulated pollutants, the emission reductions are still not sufficient to help biorefineries avoid being subject to major source permitting requirements especially for larger refineries, 5,200 and 9,100 dmt/d). Because the boiler is still a major contributor of most of the NSR-regulated pollutants in the lignin-for-pellet biorefineries, we explore an alternative scenario that could further reduce emissions by assuming the biorefineries only produce process heat from the boilers. The biorefineries in the alternative scenario will meet their power demand with non-combustion renewable electricity such as solar or wind (Fig. 1).

This alternative scenario is evaluated for all three biorefinery sizes

and two feedstock types where lignin is utilized for pellet production with a total of six design permutations to understand to what extent the PTE of regulated pollutants could be further reduced and whether using 100% non-emitting renewable electricity could help the biorefinery avoid being a major source. Just like for the other designs, we conducted a regulatory analysis and conclude that, compared to the lignin-for-pellet scenario, this alternative scenario does not trigger additional regulatory requirements beyond the ones already listed in Section 3.2.

Fig. 4 shows the preliminary PTE along with the contribution of grouped emission points to the facility-wide PTE for this alternative scenario using 100% renewable electricity. The left panel shows the magnitude of emissions for NSR-regulated air pollutants considering all federally enforceable emission controls and the regulatory requirements applicable to the process for six design permutations (three sizes and two feedstocks). The installed emission controls along with regulatory limitations required through the NSPS and NESHAP sections applicable to the process reduces the preliminary PTE of NO_x, PM, VOCs, and HAPs by at least 54%, 84%, 67%, and 64%, respectively, as compared to the baseline PTE (refer to the SI for details). The preliminary PTE for CO is identical to the baseline PTE. Because sulfur is insignificant in the boiler fuel, the change in preliminary PTE of SO₂ is insignificant relative to baseline PTE.

The right panel of Fig. 4 shows the contribution of emission points to overall PTE for each pollutant. The boiler is the dominant source of CO and NO_x emissions, contributing 97% and 94% of facility-wide emissions, respectively, and enzyme production is the major contributor, 98%, to SO₂ emissions. WWT is the single largest VOC emission source, contributing 29% of facility-wide VOC emissions when corn stover is utilized as a feedstock while pretreatment operation is the major VOC contributor accounting for 48% of overall VOC emissions when UFB is used as a feedstock. This is because of the composition difference in acetate between the two feedstocks. For PM, lignin pelletization and truck traffic combined contribute 94% of facility-wide PM emissions. For HAP emissions, wastewater contributes the majority, ~41%, of facility-wide HAP emissions for both feedstocks because of the reduction in emissions from the boiler under the applicable NSPS and NESHAP. Refer to PTE excel spreadsheets in the SI for details on major contributor for each pollutant in the renewable electricity scenario.

The PTE of the biorefinery utilizing 100% renewable electricity is estimated to be lower by 57–63% for CO, 56–63% for NO_x, 18–21% for VOCs, and 38–43% for HAPs across different feedstocks and biorefinery scales when compared to the biorefineries in the lignin-for-pellet scenario, which produces both heat and power on-site from waste gases to meet process energy demand. This is mainly because of the reduction in emissions from the boiler, whose heat input capacity decreases by

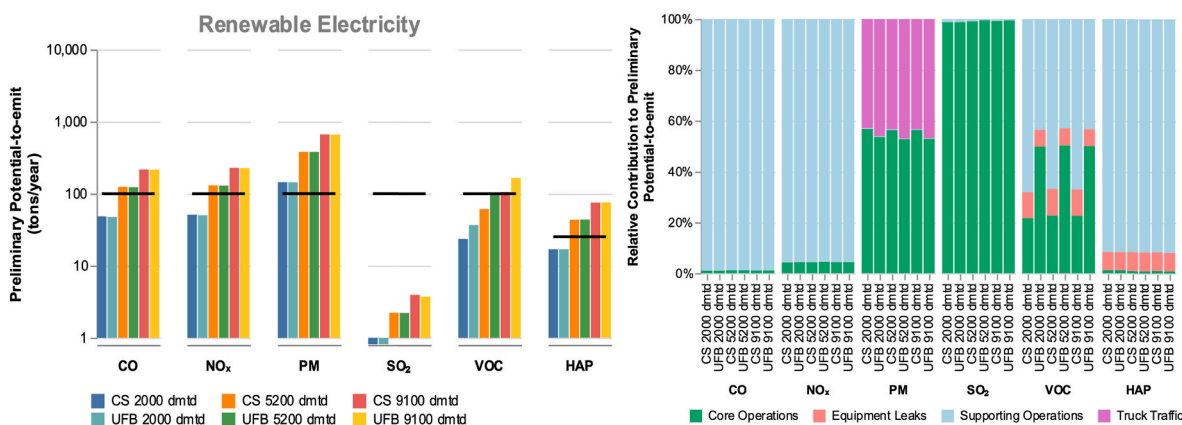


Fig. 4. Preliminary PTE for the process utilizing lignin for pellet production where renewable electricity is used for meeting electricity needs for a biorefinery processing 2000 dmt/d, 5200 dmt/d, and 9100 dmt/d of either UFB or corn stover as a feedstock. The left panel shows the preliminary PTE of several regulated air pollutants for six design permutations, and the right panel shows the relative contribution of each emission point to the overall PTE. The black solid line represents the major source threshold value (100 tpy) for criteria air pollutants and HAPs (25 tpy). Note that the y-axis in the left panel is plotted on a log-scale.

58–64% depending on the feedstock type as compared to a biorefinery producing electricity on-site. For PM and SO₂, the change in PTE is insignificant because there is no significant change in their primary emission sources, lignin pelletizer, truck traffic, and enzyme production, because of using renewable electricity.

As a result of our preliminary PTE analysis, we find that the emissions of all NSR-regulated pollutants and HAPs are below their respective major source threshold for a biorefinery processing 2,000 dmttd except for PM, whose PTE needs to be reduced by more than 44 tpy to be below major source threshold; this is despite the fact that the PTE of both PM₁₀ and PM_{2.5} are below the major source threshold. For larger biorefineries (5,200 and 9,100 dmttd), the PTE of CO, NO_x, PM, VOCs, and HAPs are above major source thresholds except for VOCs at the 5200 dmttd biorefinery size using either feedstock.

As discussed in Section 4.1, several control options and technologies are available to further reduce the emissions of the regulated pollutants. Whether the biorefinery would be able to achieve minor source classification would depend on the magnitude of emissions from each biorefinery scale. For example, an RCO and SCR can be utilized for larger biorefineries, 5,200 and 9,100 dmttd, to reduce 98% and 90% of CO and NO_x emissions, respectively, and these reductions could result in emissions below major source thresholds for CO and NO_x for both feedstocks. Similarly, a combination of paved roads and a bargehouse on the pelletizer can reduce the PTE of PM to below major source threshold for the 2,000 dmttd biorefinery size using either feedstock and 5,200 dmttd biorefinery size using corn stover as a feedstock; however, it would not quite reduce emissions enough for the 5,200 dmttd (using UFB) and 9,100 dmttd (using corn stover or UFB) biorefinery scales to be below major source levels. Refer to SI for examples on VOCs and HAPs.

4.3. Limitations

The estimates of PTE are based on information available from the literature, permit documents for facilities employing similar unit operations, EPA models, and engineering judgement. Because the purpose of a design case of a biorefinery is to demonstrate the technical feasibility of the process, the results from our analysis should be considered preliminary as further refinement to the process (e.g., to reduce cost or increase product output) may result in changes in process parameters, which may have significant implications on the applicability of air regulations, including NSPS and NESHAP and emission estimates. Also, we only consider U.S. federal regulations, and additional requirements by state and local regulations may affect the results. Moreover, some processes (e.g., the solid-gas boiler) do not have well-established emission factors, and the emissions from certain unit operations (e.g., WWT plant) are based on engineering judgement because specific design parameters are lacking. These estimates will need to be refined based on test data from similar unit operations, design parameters, and data provided by equipment suppliers (e.g., boiler manufacturer).

Although emission control technologies and alternative designs could further reduce the overall PTE of regulated air pollutants to help avoid being a major source (e.g., diverting lignin for pellet production, smaller scales, using renewable electricity), the cost of incorporating additional emission controls and varying design configurations will need to be evaluated because these changes to the biorefinery design could significantly impact the economics of biofuel production. While we discuss potential emission controls biorefineries may elect to further reduce emissions, some of the control technologies (e.g., RCO for CO pollutant discussed in Section 4.1) are only demonstrated for certain applications such as on gas boilers. As a result, the same destruction efficiency may not be practically achievable for the novel processes such as the solid-gas boiler. A refinement of emission control efficiencies would be needed based on data obtained from the boiler and control technology manufacturers.

Our analysis assumes the biorefinery is to be in an attainment area that meets the NAAQS. Siting a biorefinery in a nonattainment area

for a pollutant whose PTE from the facility would trigger the Nonattainment NSR review could be challenging and lead to additional federal, state, and local requirements to meet the Lowest Achievable Emission Rate (LAER). Because the Nonattainment NSR review is location-specific, evaluating the Nonattainment NSR permitting requirements is beyond the scope of this analysis.

5. Concluding remarks

Utilizing abundant lignocellulosic biomass for biofuel production offers opportunities for the transportation sector to mitigate GHG emissions from fossil fuel consumption. Few analyses of biofuel production pathways have examined regulatory impediments to commercialization such as air permitting implications for advanced biorefinery designs. In this paper, we fill this gap by analyzing how multiple process design variations including feedstock variability, plant scale, and lignin use may influence air pollutant emissions, applicability of federal regulations, and air permitting requirements. Because advanced biorefineries are still nascent, this type of analysis could provide insights into strategies to minimize adverse environmental impacts as well as investment risks associated with air permitting requirements.

By leveraging process designs developed in Aspen Plus and documented in Davis et al. (2022), we carry out a thorough regulatory and air emissions assessment for an advanced biorefinery utilizing lignocellulosic feedstock to produce RDB via an aerobic respiration process. The results suggest the design cases as they currently stand may not meet all federal air regulatory requirements, to which the biorefinery is subject; depending on the design configuration, additional emission controls or work standards will need to be incorporated to comply with all applicable federal air regulatory requirements. Our analysis indicates the biorefinery scales and the type of feedstocks do not affect which federal regulations (i.e., NSPS and NESHAPs) would potentially apply to the facility or unit operations. However, how the lignin is used by the biorefinery affects the process design (e.g., boiler type, inclusion of lignin dryer and pelletizer) and consequentially influences the applicability of certain regulatory requirement because of the varying equipment (e.g., Subpart FFFF would apply to the continuous dryer vent). Regardless of how lignin is used, all three biorefinery scales using corn stover or UFB as feedstock for the 12 design permutations—not including six additional renewable electricity scenarios—are estimated to be major sources under the NSR permitting program. The pollutants that exceed their major source thresholds include PM, CO, NO_x, SO₂, VOCs, and HAPs. A biorefinery burning lignin as a fuel emits more CO, NO_x, SO₂, VOCs, and HAPs compared to a same size biorefinery that diverts lignin to produce pellets because of the larger boiler size and the higher emission factors for the boiler employed to combust both solid and gaseous fuel streams. Control options are available to reduce the PTE to below major source threshold for all pollutants for the smallest-scale biorefinery (2,000 dmttd) utilizing lignin-for-pellet production. However, the technical feasibility and cost-effectiveness of these emission controls need to be assessed prior to making the final investment decision, and all the considered controls could not reduce emissions below major source for the 2,000 dmttd biorefinery utilizing lignin-for-fuel and two larger biorefinery sizes.

We also explored the effect of a popular strategy for companies to reduce air emissions and its potential to achieve minor source status for biorefineries. This scenario uses the lignin-for-pellet design as its base case and assumes the biorefinery opts to use 100% renewable electricity to fulfill process power demand, thus reducing the use of biogas and off-gases in the boiler to fulfill only the facility steam requirements. This reduces the heat input capacity of the boiler and emissions from it. Our estimate shows that adopting renewable electricity could reduce the PTE of CO, NO_x, VOCs, and HAPs by 18–63% compared to the lignin-for-pellet scenario. We find that for the smallest biorefinery, adopting this strategy could reduce the PTE of all regulated pollutants to below major source thresholds, except for PM. Such a biorefinery could elect to pave

all roads within the biorefinery and adopt a baghouse in the pelletization area to lower its PTE of PM to below major source threshold and therefore avoid being subject to major source permitting requirements. However, for larger biorefineries, 5,200 and 9,100 dmt/d, the PTE of multiple regulated pollutants still exceeds the respective major source thresholds. It is possible that utilizing several emission controls (e.g., baghouse, electrostatic precipitator, wet scrubber) can reduce the NSR-regulated pollutants to below major source thresholds. However, the larger biorefineries likely will trigger Title V permitting requirements because of exceeding the major source threshold for HAPs. Detailed information on HAP species is needed to evaluate whether and how the larger biorefineries could further reduce the HAP emissions, a subject for future research.

Our findings can help the air permitting process for novel biofuel production facilities and inform developers and stakeholders of the risks associated with timely obtaining required air permits, which are often overlooked while deploying the technologies. Our results can also help develop strategies to expedite the permitting process, shed light on sizing and siting biorefineries, and show the impact of feedstock and alternative uses of lignin on air permitting requirements.

CRedit authorship contribution statement

Arpit H. Bhatt: Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Funding acquisition. **Yimin Zhang:** Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Funding acquisition. **Ryan Davis:** Methodology, Writing – review & editing. **Garvin Heath:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition. **Vikram Ravi:** Visualization.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2022.132409>.

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