

Life Cycle Assessment of a PHA Production

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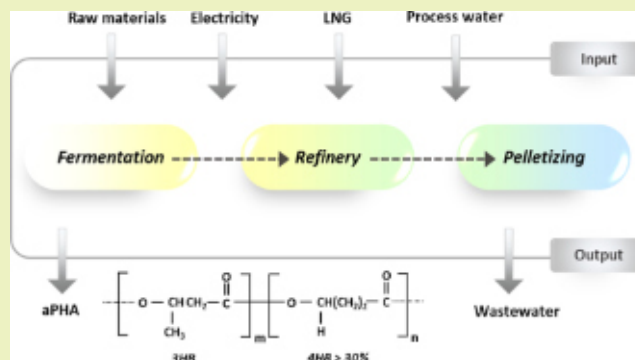
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ABSTRACT: The global production of plastic has been increasing steadily in recent years, with plastic packaging accounting for approximately 39% of the total. Plastics used for packaging are fossil-fuel-based and thus have severe environmental impacts. Polyhydroxyalkanoates (PHAs) are considered alternatives to fossil fuel-based plastics. Amorphous PHAs (aPHAs), are used in a wide range of applications owing to their rubber-like properties and remarkable fresh-water and ocean biodegradability. However, few studies have performed environmental impact assessments on aPHAs. Therefore, this study performed a life cycle assessment (LCA) based on site-specific and actual aPHA production data. The results of key impact categories and a single score for the three endpoints of aPHA production provided comprehensive insights into the environmental sustainability of aPHAs. In addition, parametric analysis (PA) was performed to analyze the possibility of increasing the sustainability of the aPHA production process. The global warming potential (GWP) of aPHA production using sugarcane ranged from 0.86 to 1.59 kg CO₂ equiv/kg aPHA, while that of corn grains ranged from 0.01 to 1.96 kg CO₂ equiv/kg aPHA. Thus, in terms of its environmental impact, the findings suggest that aPHA, as a biobased and biodegradable plastic, shows improved performance compared to fossil fuel-based plastics.

KEYWORDS: aPHA, life cycle assessment (LCA), global warming potential (GWP), first-generation PHA, parametric analysis



INTRODUCTION

The global production of plastics reached 390.7 Mt in 2021, which was approximately 10% greater than that in 2018.¹ Plastics are primarily applied for packaging, which accounts for 39% of plastic demand.² Plastics used for packaging are fossil fuel-based plastics, such as polyethylene (PE), polypropylene (PP), and polyethylene terephthalate (PET). In 2021, 90.2% of plastics produced were fossil fuel-based, 8.3% were recycled, and only 1.5% were biobased. The massive production of fossil fuel-based plastics causes severe environmental problems. First, fossil fuels used to produce plastics account for approximately 8–9% of the global oil and gas consumption, contributing to resource depletion.³ Additionally, approximately 66% of the produced plastics end up in landfills or leak into the environment,⁴ thereby negatively impacting marine⁵ and terrestrial⁶ ecosystems as well as human health.^{7,8}

Bioplastics are recognized as alternatives to fossil fuel-based plastics and can be broadly categorized as biodegradable or nonbiodegradable (Figure 1). The bioplastic market is expected to expand approximately 2.9 times from 2022 (2.2 Mt) to 2027 (6.3 Mt).⁹ Moreover, the market for biodegradable plastics is larger than that for nonbiodegradable plastics.

Among various biobased and biodegradable plastics, polyhydroxyalkanoate (PHA) is gaining increasing attention owing to its fresh water and ocean biodegradability. Hence, the

environmental impact of PHA production has been continuously analyzed through life cycle assessments (LCAs), and many studies have focused on its global warming potential (GWP). The first generation of PHA used sugarcane or corn grains as feedstock. Harding et al. analyzed the GWP of PHA using sugarcane as a feedstock and reported the GWP value of PHA to be 1.96–2.60 kg CO₂ equiv/kg PHA.¹⁰ The LCA method adopts a cradle-to-gate scope and uses experimental and simulation data. Lopez-Arenas et al. also analyzed the GWP of PHA using simulation data and reported that the GWP value of PHA was 1.70–1.90 kg of CO₂ equiv/kg PHA.¹¹

In comparison, corn as a feedstock showed a GWP range wider than that of sugarcane. Kim and Dale conducted an LCA of PHA using corn grains as feedstock.¹² They used data from various references to create their LCA input data and proposed an integrated system that used corn grains and corn stover to produce PHA. As a result, they reported that the GWP value of PHA was –1.93 to –0.28 kg of CO₂ equiv/kg of PHA for the

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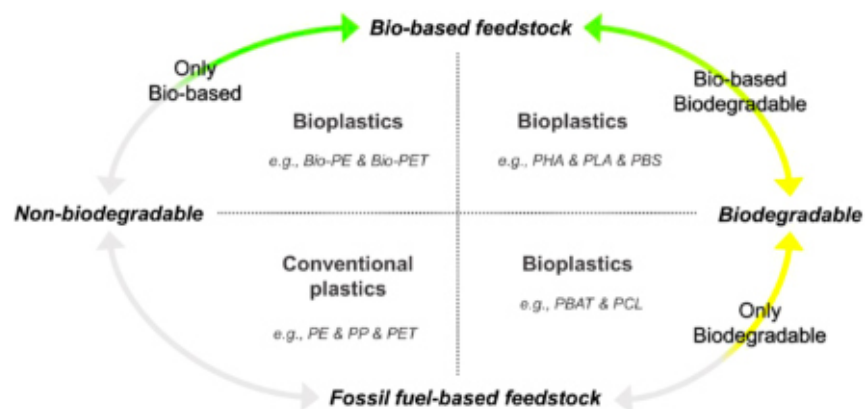


Figure 1. Varioustypesofplastics.

proposed system. However, the GWP value of PHA production using corn grain only was 1.60–4.10 kg of CO₂ equiv/kg of PHA. In a follow-up study, Kim and Dale conducted a cradle-to-gate LCA based on site-specific data from Telles. Without considering system expansion and the pelletizing stage, the GWP value of PHA production from corn grain was −0.45 kg CO₂ equiv/kg PHA.¹³

These second generation of PHA used non food materials and waste as feedstock. GWP credits can be derived from recycling waste when it is used as a feedstock for PHA production. Da Costa et al. analyzed the LCA of PHA production using wastewater and concluded that the GWP value was 1.97–2.60 kg CO₂ equiv/kg PHA.¹⁴ Kendall reported that the GWP value for PHA production using organic waste as feedstock was 3.10–5.10 kg CO₂ equiv/kg PHA.¹⁵ These GWP values only include the cradle-to-gate LCA results and exclude the results of the PHA end-of-life scenario. López-Abelairas et al. reported that the GWP value for PHA production using the organic acid gluconate as a feedstock was 4.08–29.46 kg CO₂ equiv/kg PHA.¹⁶ These studies used a cradle-to-gate scope, experimental and simulation data, or data obtained from various references. Recently, third-generation PHA was developed using carbon dioxide (CO₂) as an energy source. Koch et al. found that the production of PHA using CO₂ as a feedstock had a GWP value of 8.68–23.58 kg CO₂ equiv/kg PHA.¹⁷ This is the result of a cradle-to-gate LCA based on experimental results from various studies.

As mentioned above, three types of PHA production methods have been developed using various feedstocks, and many studies have attempted to analyze the environmental impact of these products. However, most previous studies were based on data from experiments, simulations, or pilot facilities. To provide a more reliable environmental impact of PHA production, an LCA using site-specific and real plant data is required. Although Kim and Dale used site-specific data, they were not from real plant data. In addition, no previous studies have analyzed the environmental impact of amorphous polyhydroxyalkanoates (aPHAs).

In this study, to provide reliable results on the environmental impact of aPHAs, an LCA was conducted using actual production and site-specific data from CJ CheilJedang, the exclusive provider of aPHA. aPHA production LCA using sugarcane and corn grains were conducted, and parametric analyses were performed by considering raw material usage, transportation, and proportion of renewable energy use depending on the plant location to address the uncertainty.

Finally, the environmental impact of aPHA production was compared to that of other biobased plastics and conventional fossil fuel-based plastic to analyze the sustainability of aPHA production.

MATERIALS AND METHODS

aPHA Details. CJ CheilJedang, the exclusive producer of aPHA, constructed a PHA plant in Pasuruan, Indonesia (Figure 2). aPHA



Figure 2. Panoramic view of CJ CheilJedang's aPHA plant in Pasuruan, Indonesia.

is a copolymer of 3-hydroxybutyrate (3HB) and 4-hydroxybutyrate (4HB) (Figure 3) that is formed when the proportion of 4HB exceeds

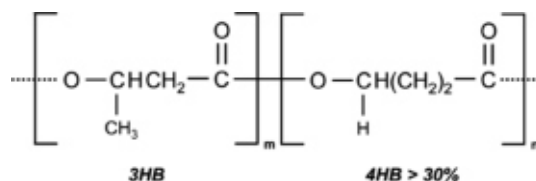


Figure 3. aPHA structure.

30%, and it has rubber-like soft properties compared with crystalline and semicrystalline PHAs (cPHA and scPHA, respectively). The properties of each PHA type are summarized in Table S1.

As shown in Figure 4, aPHA has a high degree of fresh-water and ocean biodegradability. According to the experimental results by Korea Conformity Laboratories (KCL), aPHA showed a significant weight loss of 57.1% after 11 weeks in the marine environment on the coast of Daebu Island, South Korea. This is because the degradation rate increases as the crystallinity decreases.¹⁹ Thus, aPHA, which has the lowest crystallinity among PHAs, exhibited the fastest degradation rate.

	0 week	5 week	11 week	Weight loss(%)
aPHA				57.1 %
scPHA				27.7%

Figure 4. Biodegradability of aPHA and scPHA in the marine environment¹⁸

Because of their characteristics, aPHA can replace fossil fuel-based plastics in food packaging and plastic bags. In addition, owing to its rubber-like properties, aPHA can be used as a physical modifier to enhance processability by adding flexibility. For example, the mechanical properties of brittle polymers, such as polylactic acid (PLA), can be improved by adding aPHA.²⁰ aPHA can also be blended with other biodegradable materials in addition to PLA, and its applications are summarized in Table 1.

Table 1. Applications of aPHA

processing type	examples
extrusion	film, straw
thermoforming	sheet, tray
injection molding	containers, case
fibers	nonwoven, monofilament, multifilament
coating	extrusion coating, emulsion coating
ingredient	personal care, medical care

Life Cycle Assessment. This study performed an LCA to evaluate the prospective environmental consequences associated with the production of aPHA derived from sugarcane or corn grains. The LCA was modeled using the commercial software SimaPro (version 9.5.0.0) and conducted according to ISO 14040/44 standards.²¹

According to ISO 14040, the LCA procedure is divided into four stages: goal and scope, inventory analysis, impact assessment, and interpretation (Figure 5). Details of each stage are discussed in the following sections.

Goal and Scope. This LCA study aimed to quantify the environmental impacts associated with the production of 1 kg of aPHA. The LCA in this study was conducted using a cradle-to-gate approach, and the system boundaries are denoted by the dotted line

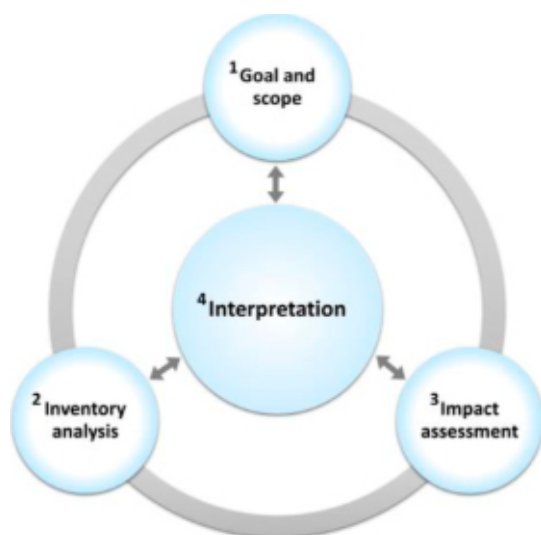


Figure 5. LCA methodology by ISO 14040.

in Figure 6. Transportation of chemicals and utilities is also included in the system boundaries. As previously mentioned, aPHA can be produced using either sugarcane or corn grains. Sugar, a disaccharide produced from sugarcane, and dextrose (i.e., D-glucose), a monosaccharide produced from the hydrolysis of starch in corn grain, are the two feedstocks for aPHA production. Therefore, this study evaluated the environmental impact of aPHA production in both cases.

Sugar, the raw material for aPHA production, was derived from sugarcane. Sugarcane is composed of approximately 12% sugar, 10% fiber, and other components, including water.²² Sugar is extracted from sugarcane through milling, which produces sugar and byproducts, such as bagasse, molasses, and filter cake.²³ Bagasse can be burned to generate electricity for sugarcane milling, and filter cake can be utilized as a soil conditioner for sugarcane production.²⁴ Finally, molasses can be used as a transportation fuel.²⁵ The use of these byproducts was considered in this study.

Dextrose, which is the raw material for aPHA, was derived from corn. Corn is composed of approximately 74% carbohydrates (mainly starch), 9% protein, 7% total dietary fiber, and 10% water, and it can be divided into corn grains, corn kernels, and corn stover. Starch was extracted from corn grain through a wet mill process and then enzymatically hydrolyzed to produce dextrose.²⁶ Therefore, among the three corn parts, only the corn grain required for dextrose production was included within the system boundaries.

A comparison of corn and sugarcane shows that corn has a starch content of approximately 74%,²⁶ with 100% of the starch assumed to be converted to dextrose,²⁷ while sugarcane contains approximately 12% sugar,²² with an approximately 74.6% sugar extraction efficiency through sugarcane milling.²⁸ Consequently, aPHA production requires a larger quantity of sugarcane than corn. Starting with the fermentation process, dextrose from corn and sugar from sugarcane were subjected to similar procedures to produce aPHA.

Dextrose and sugar served as nutrient sources for microorganisms during fermentation. Microorganisms consume carbohydrates and fatty acids as energy sources. However, in the presence of excess carbon, these organisms store energy in the form of aPHA, leading to their production. Consequently, aPHA is formed during fermentation and extracted from microorganisms through a refining process. The accumulated aPHA in microorganisms is recovered by physicochemical fracturing after separating other impurities (such as carbohydrates, proteins, fatty acids, and organic acids) from the fermentation broth. In addition, high-purity aPHA can be obtained by removing impurities such as lipid bilayers, membrane proteins, and nucleic acid through chemical and enzymatic purification, dehydration processes, and drying. PHA was then pelletized to produce aPHA.

Inventory Analysis. Primary and secondary databases were used for the LCA. For the primary database, the material and energy balances for sugarcane within the system boundaries were based on CJ Cheil Jedang's aPHA production plant in Pasuruan, Indonesia. However, the material and energy balances for corn grain within the system boundaries were based on key experimental and simulation data provided by CJ Cheil Jedang. Based on these data, CJ Cheil Jedang plans to build aPHA plants in Iowa in the Central United States. Therefore, the primary data from CJ Cheil Jedang provide the quantity of raw materials used in aPHA production. Furthermore, the consumption of utilities, such as electricity and liquefied natural gas (LNG), is also based on these data. The same applies to the amount of wastewater generated from the fermentation to pelletizing processes. The methods for collecting and validating these primary databases are provided in Figure S1.

These secondary databases included Ecoinvent 3.9.1 and the United States Life Cycle Inventory (USLCI), and it was included in SimaPro 9.5.0.0. The secondary database contains production data for the raw materials used in aPHA production. In the case of corn grains, which serve as raw materials for dextrose, the United States contributes 37% of the total corn production.²⁶ Therefore, this study analyzed the environmental impact of corn grain production using the USLCI, an inventory dataset provided by the United States. Dextrose production from corn grains was investigated using Ecoinvent 3.9.1. Dextrose was

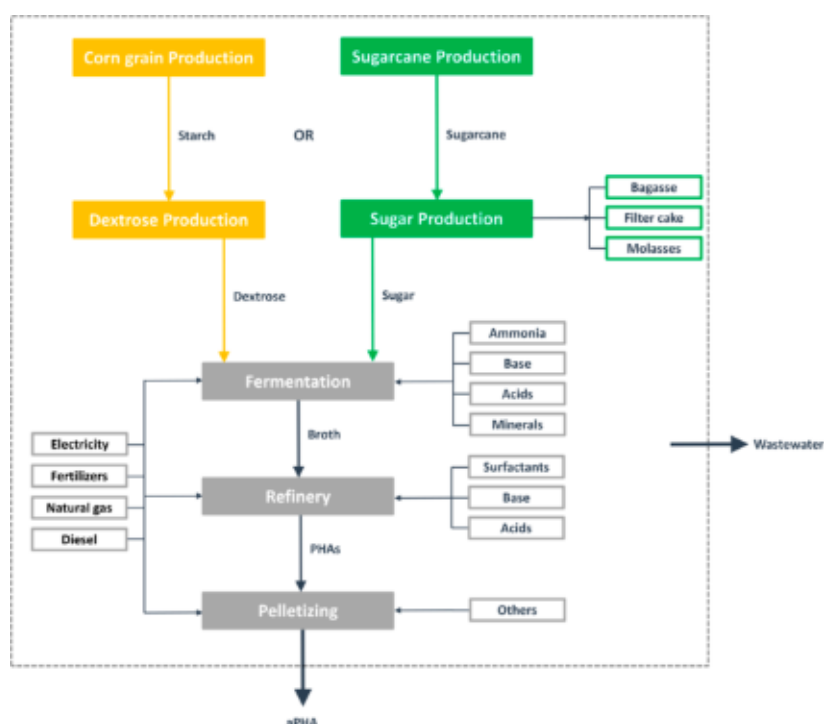


Figure 6. Simplified flow diagram for the production of aPHA. Dotted lines represent system boundaries.

assumed to be produced in the United States, and the rest of the world (ROW) production data fromecoinvent were used, because U.S.-based production data were unavailable. As Brazil is the largest sugarcane producer,²⁹ sugarcane and sugar production data were obtained fromecoinvent's Brazilian production data. Transportation from raw material production sites to the aPHA production plants was also considered usingecoinvent data. Table 2 summarizes the details of the primary and secondary databases used in this study.

Table 2. Inventory Database for aPHA Production

process	database
sugarcane production	ecoinvent 3.9.1
sugar production	ecoinvent 3.9.1
corn grain production	USLCI
dextrose production	ecoinvent 3.9.1
fermentation	CJCheilJedang production data
refinery	CJCheilJedang production data
pelletizing	CJCheilJedang production data

Impact Assessment. The environmental impact of aPHA manufacturing was calculated by using the ReCiPe 2016 Midpoint (H) methodology. Eighteen impact categories were considered in the methodology. Among these categories, the following impact categories were analyzed: GWP, fine particulate matter, land use, and water consumption. The impact categories that are not considered in the Results section are aggregated into endpoint based on the ReCiPe 2016 Endpoint (H) methodology. In this methodology, 18 midpoint impact categories are comprehensively considered based on three endpoints: Human Health, Ecosystems, and Resources. Additionally, each of the three endpoints are weighted to calculate a single score that provides an integrated view of the environmental impact. Additional details are provided in Figure S2 and Tables S2 and S3. In addition, nonrenewable (fossil) and renewable energy uses were analyzed using the cumulative energy demand methodology.

Case Study. Case 1: Production of aPHA Using Sugarcane. Case 1 evaluated the environmental impact of aPHA production using

sugarcane as the feedstock. As previously mentioned, the production of sugarcane and sugar was based on theecoinvent Brazil database. CO₂ absorbed during photosynthesis³⁰ was considered during sugarcane growth. While some CO₂ was emitted back into the air, the remainder was stored in aPHA. The amount of CO₂ uptake by aPHA was calculated as follows¹⁷

$$\text{carbon mass in aPHA monomer (C}_4\text{H}_6\text{O}_2\text{)} = \frac{\text{mass of C}_4}{M_w \text{ of C}_4\text{H}_6\text{O}_2}$$

$$= \frac{0.558 \text{ kg-C}}{1 \text{ kg-aPHA}} \quad (1)$$

$$\text{CO}_2 \text{ uptake mass in aPHA} = \frac{0.558 \text{ kg-C}}{1 \text{ kg-aPHA}} \times \frac{M_w \text{ of CO}_2}{M_w \text{ of C}}$$

$$= \frac{2.046 \text{ kg-CO}_2}{1 \text{ kg-aPHA}} \quad (2)$$

As a result, aPHA made from sugarcane stored 2.046 kg of CO₂. Considering the end of life of aPHA, we release CO₂ back into the air. However, in this study, a cradle-to-gate analysis was conducted; therefore, the release of CO₂ was not considered.³¹

As shown in Figure 6, sugar is converted to aPHA through a process that starts with fermentation and proceeds to pelletizing. The electricity used in the plant is produced from geothermal power, which is a renewable energy source. As renewable energy certificates (RECs) can be purchased in Indonesia, the use of 100% renewable energy for electricity was assumed.³² Wastewater produced from fermentation to pelletizing processes contains organics that can be anaerobically digested to produce natural gas. Natural gas produced from wastewater is known as renewable natural gas (RNG). Carbon in wastewater is biogenic, and CO₂ emissions are not produced during the combustion of RNG.³³ Based on the experimental results (see Table S4), 0.00105 Nm³ RNG was produced from 1 kg of wastewater from the aPHA production process. Therefore, it was assumed that some of the LNG used to produce aPHA was substituted with the RNG.

Case 2: Production of aPHA Using Corn Grains. Compared with case 1, case 2 evaluated the environmental impact of aPHA production using corn grains as feedstock. The production of corn

grains and dextrose was based on the US LCI andecoinvent databases, and the CO₂ uptake was also considered. The amount of CO₂ uptake depends on the type of crop,³⁴ but it was assumed that the amount of CO₂ uptake in a PHA remains constant.¹⁷

Similar to case 1, a PHA production using corn grains was based on primary data. Accordingly, RNG was partially substituted for LNG and all electricity was produced from renewable sources. However, unlike case 1, which is based in Indonesia, case 2 assumes that a PHA was produced in lowland in the Central United States because most corn grain is produced in the Central United States. Therefore, electricity generation was assumed to be from wind energy, which is most commonly employed in the Central United States.³⁵ Similar to case 1, all electricity was produced using 100% renewable energy. This is because CJ CheilJedang's U.S. plant will receive electricity from the Mid American Energy Company (MEC), which has committed to 100% renewable energy by 2025.³⁶ The comparison of case 1 and case 2 are summarized in Table 3.

Table 3. Comparison of Case 1 and Case 2

description	case 1	case 2
plant location	Pasuruan, Indonesia	Central United States
feedstock	sugarcane	corn grain
region of origin of feedstock	Brazil	Central United States
CO ₂ uptake	2.046 kg	2.046 kg
electricity	geothermal power	wind power
byproducts	bagasse, filter cake, molasses	N/A
RNG from wastewater	partial LNG substitution	partial LNG substitution

Parametric Analysis for Case 1 and Case 2. Parametric analysis (PA) was conducted for cases 1 and 2. As previously mentioned, wastewater is used to produce RNG, which partially substitutes for LNG. Compared with RNG, which has no negative environmental impact during combustion, LNG has an environmental impact. Therefore, in cases 1 and 2, the environmental impact of substituting all LNG with RNG was analyzed.

For case 1, changes in the sugar production areas were analyzed. In this study, a PHA was produced in Indonesia using sugar produced from Brazil, and the transportation distance from Brazil to Indonesia was assumed to be approximately 17,800 km. Sugarcane is also produced in Thailand and Colombia, with assumed transportation distances of 2330 and 19,000 km, respectively. Based on PA, the regions most suitable for sugar production were identified. The GWP and transportation distance for each production region are listed in Table 4.

Table 4. GWP Values by Database for 1 kg of Sugar Production

corn grain	database	GWP
sugar (base)	ecoinvent 3.9.1 (Brazil)	0.095 kg CO ₂ equiv
sugar 1	ecoinvent 3.9.1 (ROW)	0.98 kg CO ₂ equiv
sugar 2	ecoinvent 3.9.1 (Columbia)	0.29 kg CO ₂ equiv

Additionally, case 2 considered changing the database for corn grain and dextrose production. According to Tables 5 and 6,

Table 5. GWP Values by Database for 1 kg of Corn Grain Production

corn grain	database	GWP
corn grain (base)	US LCI	0.29 kg CO ₂ equiv
corn grain 1	US LCI	0.076 kg CO ₂ equiv
corn grain 2	NatureWorks LLC ²⁷	0.16 kg CO ₂ equiv

Table 6. GWP Values by Database for 1 kg of Dextrose Production

dextrose	database	GWP
dextrose (base)	ecoinvent 3.9.1 (ROW)	0.50 kg CO ₂ equiv
dextrose 1	ecoinvent 3.9.1 (^a RER)	0.35 kg CO ₂ equiv
dextrose 2	NatureWorks LLC ²⁷	0.18 kg CO ₂ equiv

^aRER: Rest of the Europe.

variations occurred in the GWP values for corn grain and dextrose production depending on the database. The reference is based on production data from NatureWorks LLC,²⁷ which uses corn grain to produce PLA in the Central United States. Therefore, these data are suitable for a PHA produced in the Central United States. The PA for each case was based on GWP and is summarized in Figure 7.

RESULTS

Results of Environmental Impact Assessment for Case 1 and Case 2. This section describes the results of the impact assessment of the production of 1 kg of a PHA in cases 1 and 2. As previously mentioned, the impact assessment was conducted based on the ReCiPe 2016 Midpoint (H), ReCiPe 2016 endpoint (H), and cumulative energy demand. Key impact assessment categories for each case, including GWP, fine particulate matter, land use, water consumption, and energy usage, are summarized in Table 7. Further details of the LCI and impact assessment categories are provided in Tables S5 and S6.

Contributions of each input to the GWP for cases 1 and 2 are shown in Figure 8. Inputs for a PHA production include raw materials, process water, raw material transportation, LNG, and electricity. In addition, as previously mentioned, the CO₂ uptake was considered in cases 1 and 2. Consequently, the GWP of case 1 was 1.59, which is lower than that of case 2 (1.96). The main reason for this is that case 2 had a higher proportion of raw materials than case 1. In case 2, the proportion of corn grains and dextrose in the raw material was approximately 3.7 times the proportion of sugarcane and sugar in case 1. This implies that the proportions of corn grains and dextrose in case 2 were substantially higher. In the case of sugarcane, the GWP was reduced using byproducts from the sugar production process (e.g., electricity production via bagasse). In comparison, byproducts were not used in the production of dextrose. Therefore, it can be concluded that a difference in GWP has occurred.

Cases 1 and 2 also exhibited differences in transportation. In case 1, sugarcane and sugar were reproduced in Brazil and a PHA was produced in an Indonesian plant. Therefore, the distance between Brazil and Indonesia was assumed to be approximately 17,800 km. By transporting these raw materials, the GWP in case 1 increased by 0.53. However, in case 2, there was no increase owing to transportation because corn grains, dextrose, and a PHA were all produced in the Central United States.

The share of GWP in electricity used to produce a PHA differed. Cases 1 and 2 produced electricity using 100% renewable energy sources. As previously mentioned, case 1 generated electricity from geothermal power in Indonesia, and case 2 generated electricity from wind power in the United States. However, the GWP of geothermal electricity generation in Indonesia³⁷ is higher than that in the Central United States.³⁸ In addition, as shown in Table S5, the electricity consumption for case 2 was 38% lower than that for case 1.

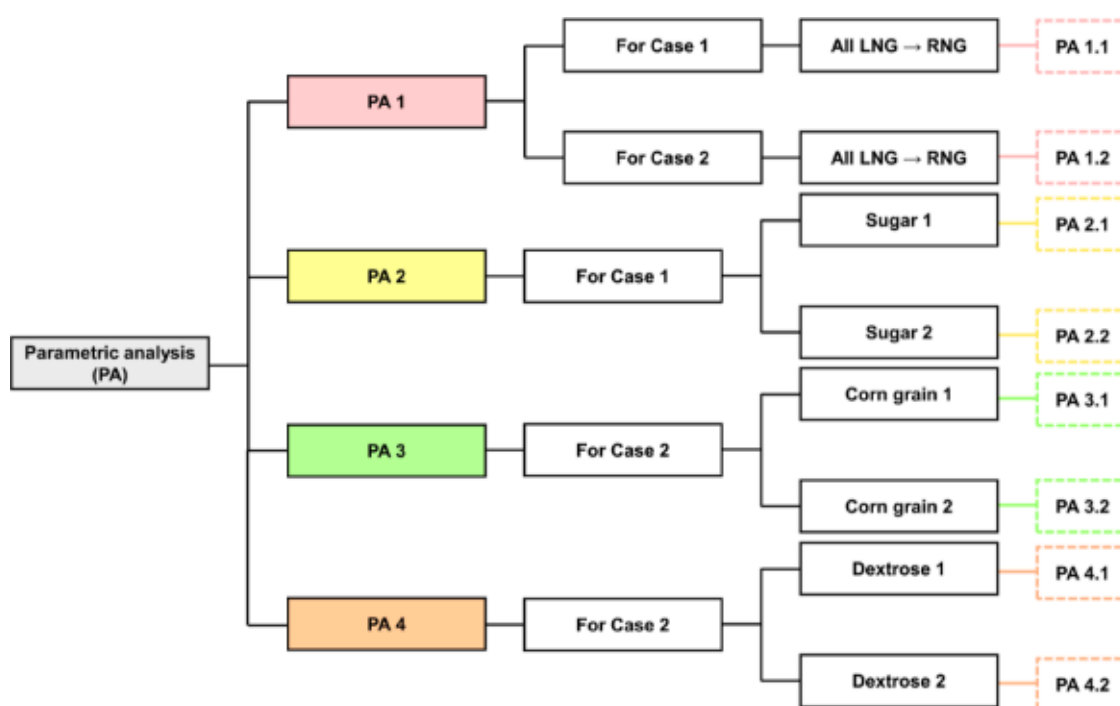


Figure 7. Summary of the parametric analysis.

Table 7. Results of the Key Impact Categories for Each Case

impact category	unit	case 1	case 2
global warming potential	kgCO ₂ equiv	1.59	1.96
fine particulate matter	kgPM _{2.5} equiv	0.015	8.24 × 10 ⁻³
land use	m ² acroequiv	4.09	3.29
water consumption	m ³	0.24	0.27
nonrenewable, fossil	MJ	55.21	47.97
renewable energy	MJ	53.84	18.55

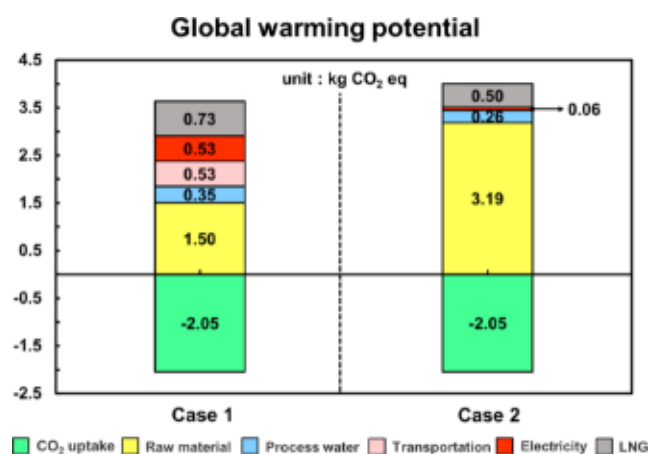


Figure 8. Contribution of each input to GWP for each case.

Therefore, although both cases used renewable energy to generate electricity, the GWP share of the electricity differed.

The contribution of each input to nonrenewable, fossil, and renewable energy for cases 1 and 2 is shown in Figure 9. As shown in Figure 9, case 1 required approximately 1.2 times more nonrenewable and fossil and approximately 2.9 times more renewable energy than case 2. However, as shown in Figure 8, the GWP of case 2 was greater than that of case 1 because case 1 had a GWP credit for the byproduct. The

combustion of bagasse produced electricity that can be used for a PHA production. For 1 kg of sugar production, approximately 2.2 kg of bagasse was generated.^{39,40} Additionally, 1 kg of bagasse contains 0.787 kg of dry matter, and 1 kg of dry matter produces 15.4 MJ of energy.²⁸ Therefore, approximately 26.7 MJ of electricity can be generated, leading to an equivalent reduction in GWP. The same applies to another byproduct, filter cake. Approximately 0.3 kg of filter cake is produced per 1 kg of sugar production.⁴⁰ Using this as a soil conditioner in sugarcane production, a reduction in GWP can be obtained.²⁵ However, despite the lower energy consumption, case 2 had a higher GWP value because no byproduct utilization was considered.

Contributions of each input to fine particulate matter formation, land use, and water consumption for cases 1 and 2 are shown in Figure 10. As shown in Figure 10, the fine particulate matter formation value was approximately 1.9 times higher in case 1 than in case 2 because the nonrenewable fossil fuel energy consumption in case 1 was higher than that in case 2. In case 1, the transportation of sugarcane and sugar was included, which increased the fine particulate matter emissions.

In addition, land use was approximately 1.2 times higher in case 1 than in case 2. Land use is affected by factors such as land area, crop productivity, and fertilizer usage. Sugarcane productivity was higher than that of corn grains.⁴¹ However, approximately 11 kg of sugarcane is consumed to produce 1 kg of sugar, which is greater than the amount of corn grains used to produce dextrose.²⁸ Therefore, land use in case 1 tended to be higher because of the greater consumption of sugarcane than corn grains.

Finally, water consumption was approximately 1.1 times higher in case 2 than in case 1. Process water usage in case 2 was lower than that in case 1. Process water represents the water used in the manufacturing of a PHA, during the fermentation to pelletizing processes. However, the amount of water used in the raw material was approximately 1.7 times

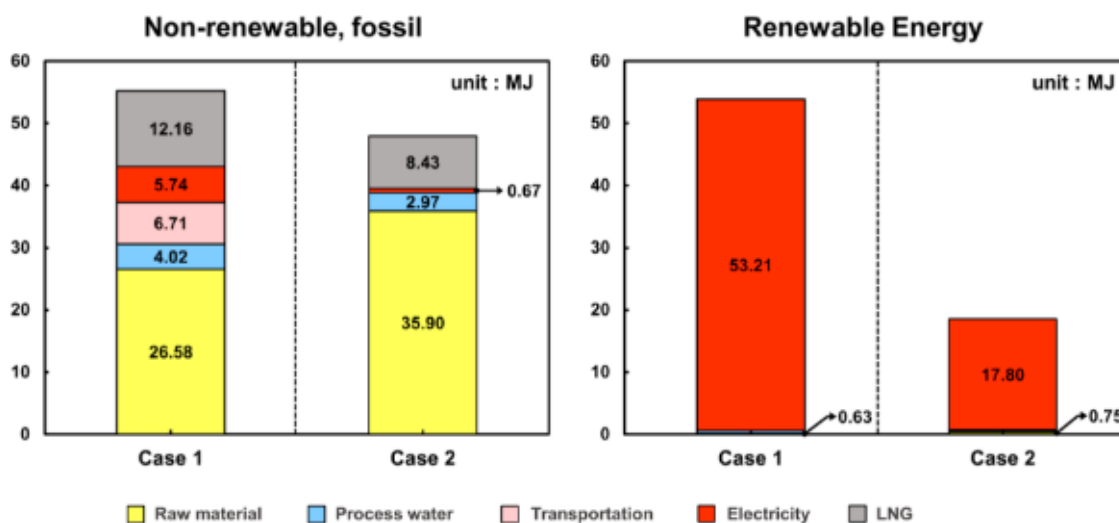


Figure 9. Contribution of each input to nonrenewable, fossil, and renewable energy for each case.

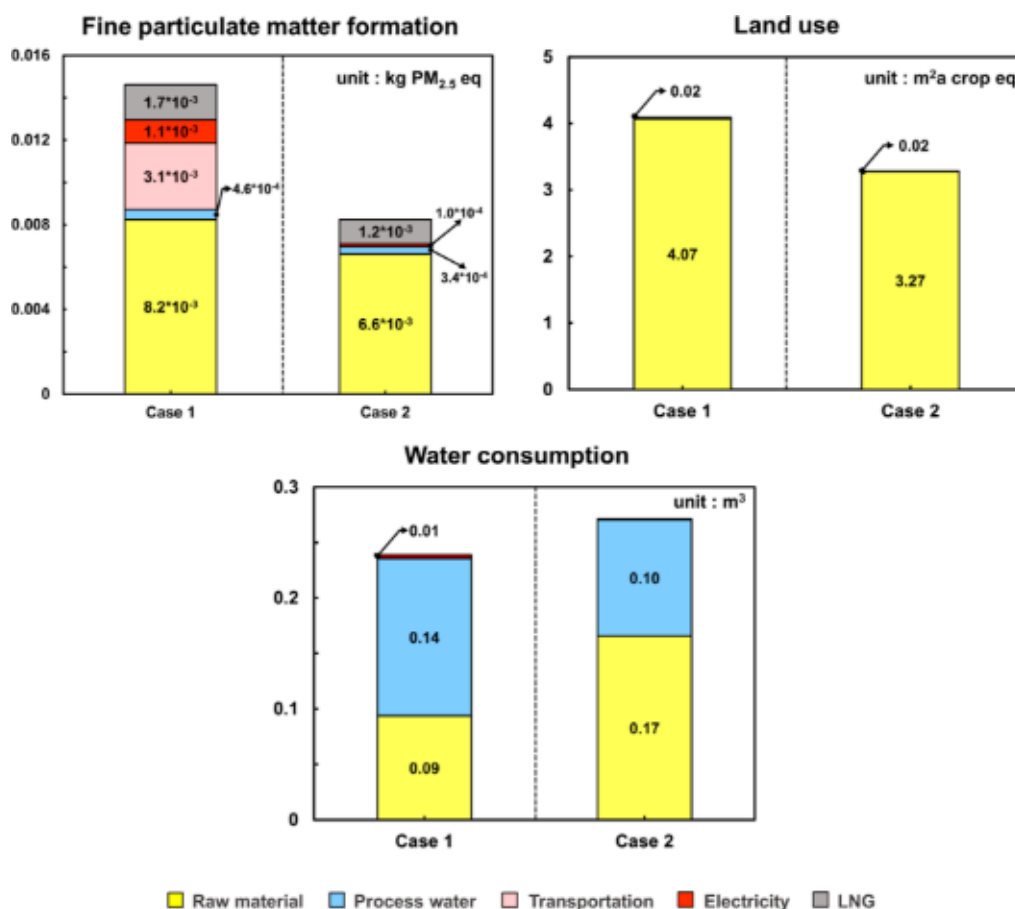


Figure 10. Contribution of each input to fine particulate matter formation, land use, and water consumption for each case.

higher in case 2 than in case 1. First, the water consumption of corn grain production is approximately 12 times higher than that for sugar cane production.⁴² In addition, dextrose is produced by the enzymatic hydrolysis of starch in corn grain.⁴³ This process consumes approximately 1.2 times more water than the sugar milling process.²⁸ Therefore, case 2 had higher water consumption than case 1.

Additionally, single score results for cases 1 and 2, providing an integrated view of the environmental impact, are shown in

Figure 11. In Figure 11, the single score of the impact category, representing a cumulative percentage of approximately 99%, is shown. The unit mPt indicates one-millionth of the one-year environmental burden of the average European population.⁴⁴

For Human Health, the total value for case 1 was 222.87 mPt, which is approximately 1.38 times higher than the 161.46 mPt of case 2. In Figure 11, both cases 1 and 2 have the largest single score for fine particulate matter formation. This indicates that fine particulate matter formation has a significant

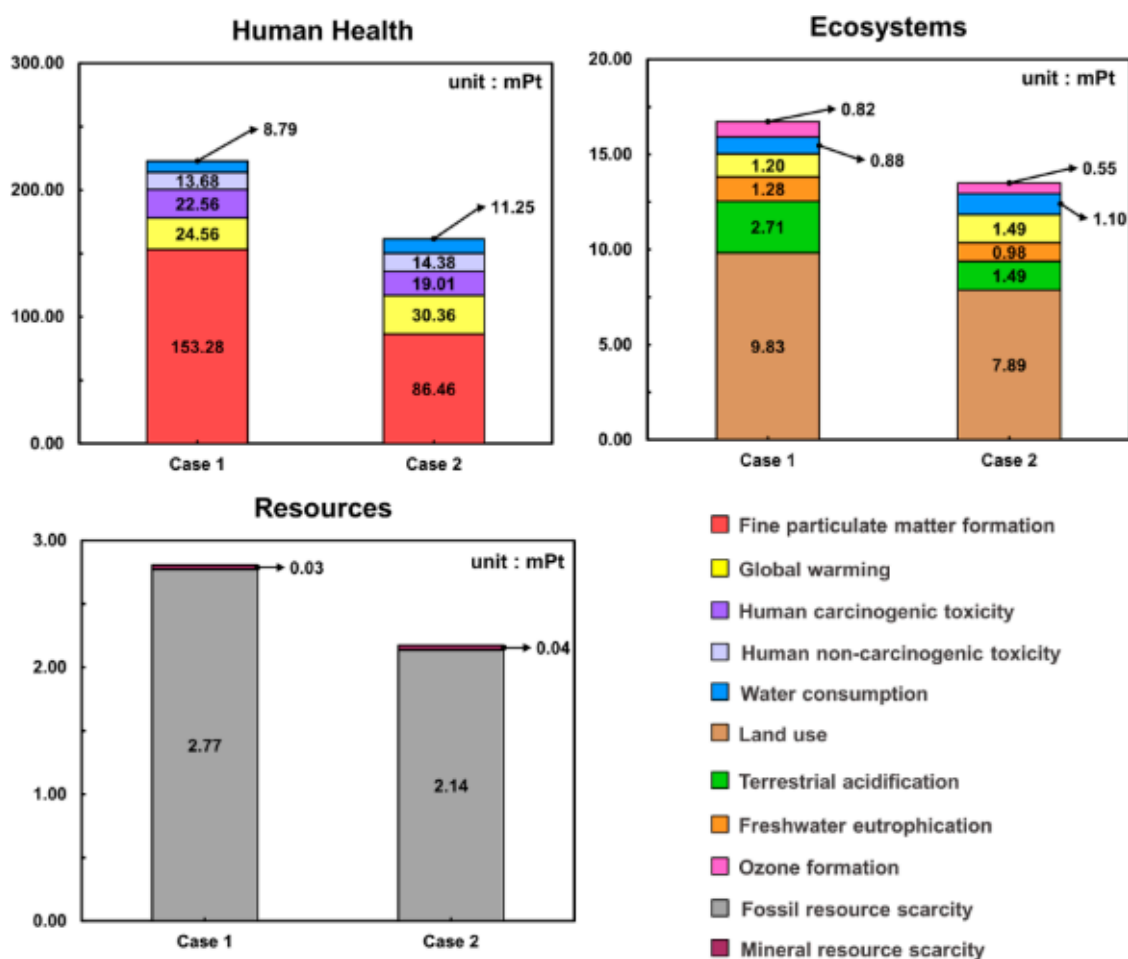


Figure 11. Single scores of impact categories for the three endpoints (Human Health, Ecosystems, and Resources).

impact on human health. As previously mentioned, case 1 had high values for fine particulate matter than case 2 due to the high nonrenewable energy consumption and inclusion of long-distance transportation of sugar to the PHA production site.

For Ecosystems, the single score of case 1 was 16.72 mPt, which is approximately 1.24 times higher than the 13.50 mPt of case 2. Ecosystems were revealed to be significantly impacted by land use. Because land use can modify land surface and species composition, energy flows and biogeochemical cycles are altered in terrestrial ecosystems.⁴⁵ As previously mentioned, case 1 had higher land use because it consumes more sugarcane compared to corn grain in case 2. Consequently, land use increased to produce more sugarcane, resulting in a higher single score.

For Resources, the total score for case 1 was 2.80 mPt, which is approximately 1.29 times higher than the 2.18 mPt of case 2. As shown in Figure 11, resources are highly affected by fossil resource scarcity. Fossil fuel resources such as crude oil and natural gas are used as nonrenewable fossil energy. The higher usage of these energy sources leads to a scarcity of fossil resources. Therefore, as shown in Figure 9, case 1, with higher values for nonrenewable and fossil resources, had a higher single score for fossil resource scarcity than case 2.

In the midpoint assessment, some impact categories were higher for case 1 while others were higher for case 2. Additionally, in the comprehensive environmental impact assessment based on the single score of the three endpoints,

case 2 exhibited a lower single score, indicating a smaller environmental impact compared to that observed in case 1.

Results of the Parametric Analysis for Case 1 and Case 2. As summarized in Figure 7, PA was performed for cases 1 and 2. The GWP results are listed in Figure 12. All environmental impact assessment results for PA1 and PA2 are provided in Table S7. For PA3 and PA4, only GWP results are provided due to data limitations from the literature.²⁷ First, PA1 indicates that substituting all LNG with RNG reduces the GWP. The GWP for cases 1 and 2 decreased by approximately 46 and 26% to 0.86 and 1.46, respectively. The larger RNG substitution effect in case 1 was due to its higher LNG usage than that in case 2. PA2 represents the GWP results for sugar production in each region. These results suggest that sugar production is the most environmentally friendly option for Brazil. First, Brazil replaced nearly 40% of its petrol consumption with renewable ethanol.⁴⁶ Sugarcane, the raw material for sugar, is burned to facilitate its harvest, resulting in negative environmental impacts. Brazil has enacted laws prohibiting sugarcane burning. As a result, it was assumed that all sugarcane will be harvested without burning by 2025.⁴⁷ Therefore, the production of sugarcane and sugar in Brazil is more environmentally friendly than that in other regions. In addition, at PA3, the effects of changing the database on corn grain production were observed. Consequently, the GWP in case 2 decreased to a minimum of 1.37, which was a larger effect than that of the RNG replacement in PA1. In the case of PA3.1, a significant reduction in the GWP occurred when

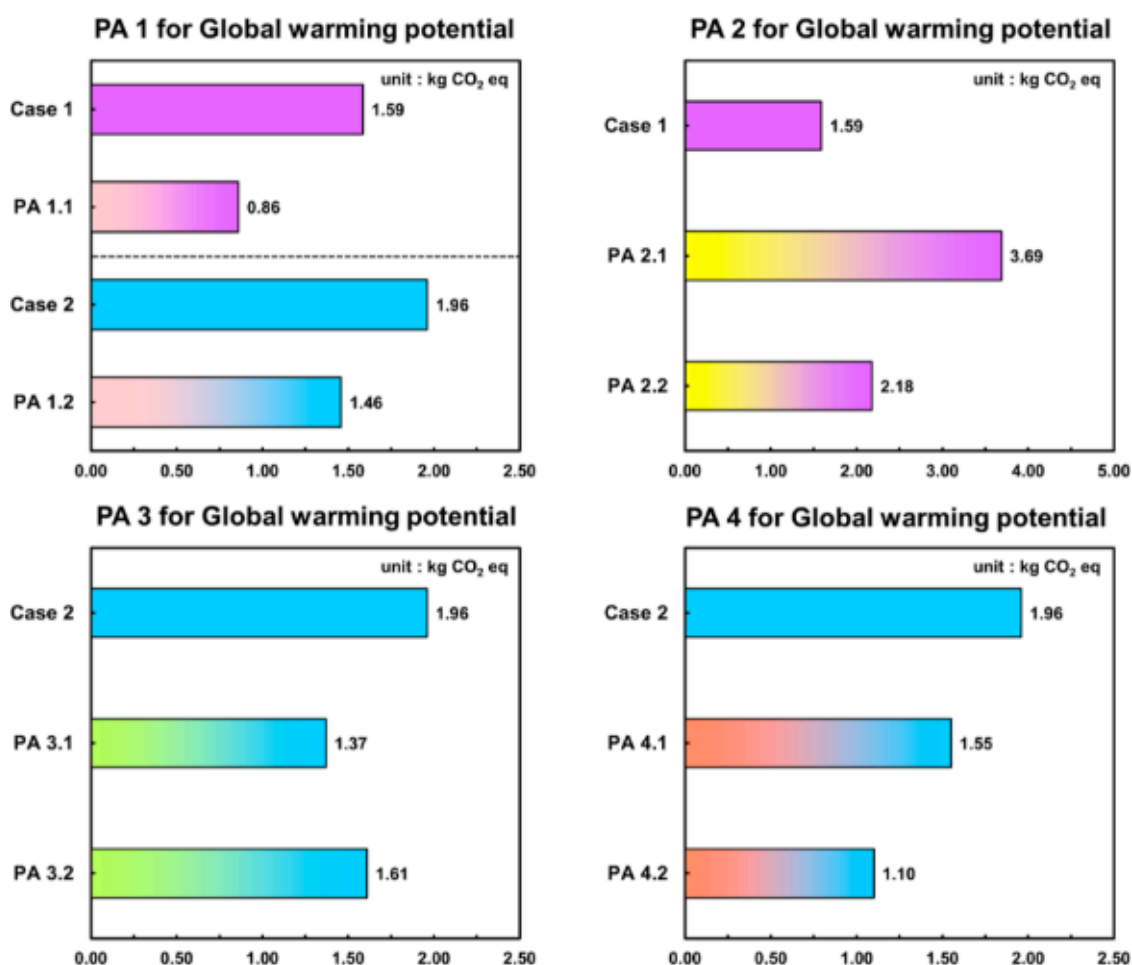


Figure 12. Parametric analysis (PA) results focusing on GWP values.

biomass was used as the energy source for corn grain production.²⁸ Finally, at PA4, the dextrose production database in case 2 changed, resulting in the lowest value of 1.10 for PA4.2. This was based on NatureWorks LLC data,²⁷ which yielded a low GWP owing to 100% renewable resources usage.

The lowest GWP values were obtained from the PA results. In case 1, substituting all LNG with RNG (PA 1.1) yielded a minimum value of 0.86. Case 2 had a minimum value of 0.01 based on PA 1.2, 3.1, and 4.2. The GWP results for the best case (cases 1 and 2) are shown in Figure 13. In Figure 13, the sugarcane best substitutes all LNG in case 1 with RNG, thereby removing the share of LNG. The same applies for corn grain best, which substituted all LNG in case 2 with RNG. Additionally, in the case of corn grain best, corn grain and dextrose production appeared to be more sustainable compared to those in case 2. Therefore, the share of raw materials was reduced by approximately 45% compared with that in case 2.

DISCUSSION

In the Results section, the LCA results of a PHA production using sugarcane and corn grains were analyzed. Table 7 provides the results for six impact categories ranging from GWP to renewable energy. The analysis of contributions of different inputs to these impact categories is shown in Figures 8 and 10. These results show that case 1 (sugarcane) had a

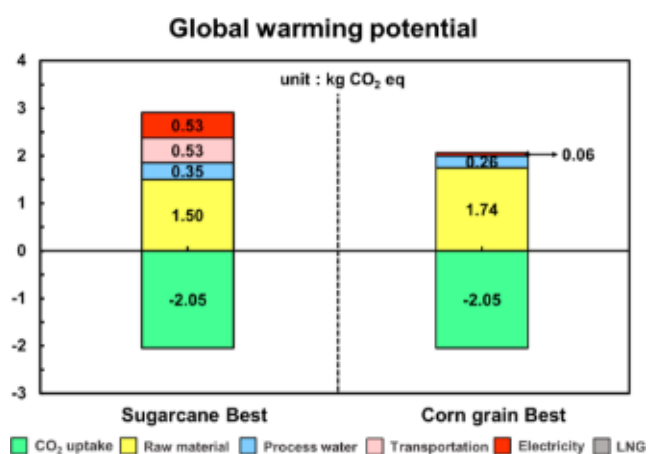


Figure 13. Contribution of each input to the best GWP values for a PHA production using sugarcane (left) and corn grains (right).

lower GWP value than case 2 (corn grain), indicating a smaller contribution to global warming. However, fine particulate matter formation, land use, and other factors had high values in case 1. Therefore, for the comprehensive evaluation of the environmental impact of case 1 and case 2, a single score for the three endpoints (Human Health, Ecosystems, and Resources) was analyzed, as shown in Figure 11. For all three endpoints, case 2 has lower values than case 1.

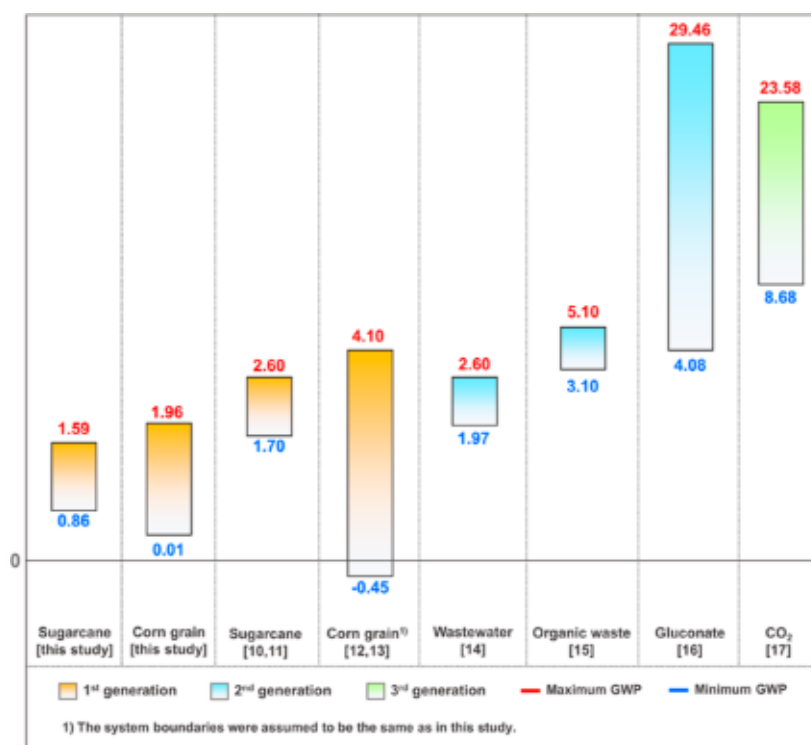


Figure 14. Comparison of GWP values: cradle-to-gate LCA for PHA production^{10–17}

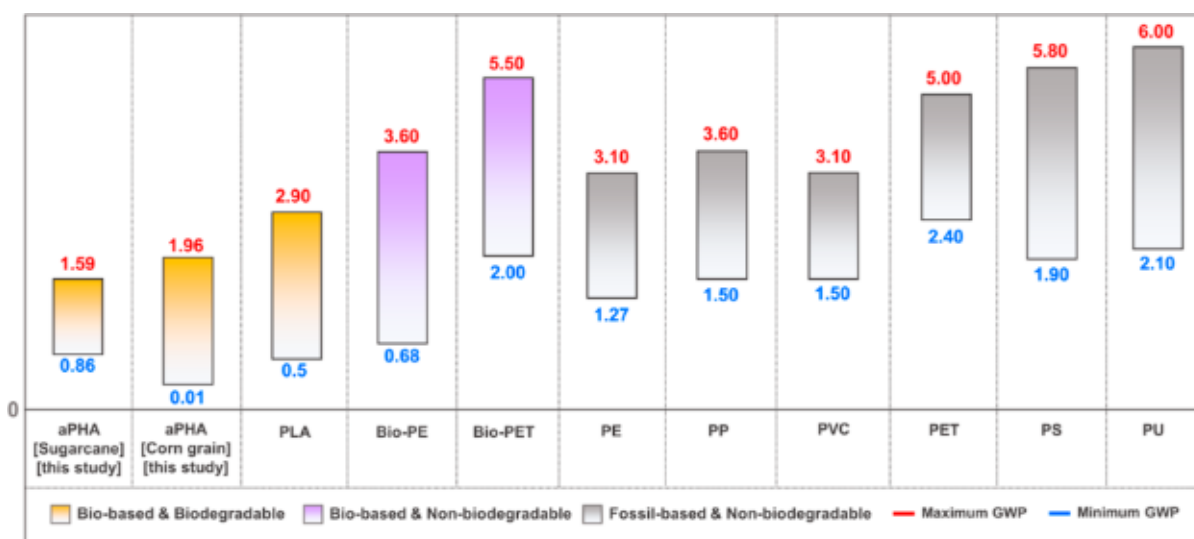


Figure 15. Comparison of GWP values from cradle-to-gate LCA for various plastics^{49,51,52}

In addition, PAS were conducted for cases 1 and 2 with a focus on GWP due to data limitations. As shown in Figure 13, the GWP value in the case of corn grain best was lower than that of sugarcane best. These results indicated that the proximity of the factory to the source of the raw material and the use of low-GWP energy sources among renewable energy sources were important factors for determining the GWP of the best case, whereas sustainable production of the feedstock was important in cases 1 and 2.

As previously mentioned, PHAs are reproduced from various feedstocks and are primarily divided into three generations. The first generation in this study was the production of PHA from food or feeds, such as sugarcane and corn grains. The second generation utilizes nonfood materials and waste as raw

materials. Finally, the third generation represents direct production by microorganisms, such as algae and cyanobacteria, which obtain energy through photosynthesis.⁴⁸ Figure 14 shows a comparison of the GWP values for PHA production methods reported in previous studies. As shown in Figure 14, the latest PHA production methods do not guarantee lower GWP values. For example, PHA production using gluconate¹⁶ and CO₂¹⁷ showed higher GWP values of 29.46 kg CO₂ equiv/kg PHA and 23.58 kg CO₂ equiv/kg PHA, respectively, compared to other production methods. This is because the use of NaOCl¹⁶ or chloroform¹⁷ for PHA extraction is not environmentally friendly. Additionally, the recovery of these solvents is energy intensive.¹⁷

The first-generation methods showed GWP values ranging from -0.45 to 4.10 kg CO₂ equiv/kg PHA. In this generation, the lowest GWP value was -0.45 kg CO₂ equiv/kg PHA.¹³ However, the nonrenewable energy consumption was also 15.7 MJ/kg PHA, which is smaller than that reported in other studies, because the pelletizing stage (which consumes approximately 43% of the total electricity) of PHA was not considered.

With the exception of Kim and Dale,¹³ other reported GWP values for the production of first-generation PHAs were higher than those obtained in this study. In this study, an environmental impact assessment for cases 1 and 2 was conducted based on CJ Cheil Jedang's actual production data to provide a realistic perspective. Therefore, the process of producing a PHA using sugarcane and corn grains in this study can be considered to have less impact on global warming than other PHA production methods.

Plastics can be categorized into four groups. Except for fossil-based and biodegradable plastics, such as polybutylene adipate terephthalate (PBAT), which has not been evaluated by environmental impact assessments,⁴⁹ the GWP results for the production of the remaining plastics are shown in Figure 15. As shown in Figure 15, biobased and biodegradable plastics, such as a PHA and PLA, generally have a lower GWP than fossil fuel-based plastics. Therefore, an increase in the use of biobased and biodegradable plastics is expected to address environmental problems related to global warming.

Further, considering the cost aspect, most conventional fossil-based plastics cost less than approximately USD 1 per kg, and the commercial price of PHA is known to be between approximately USD 1.5 and USD 5.0 per kg.⁵⁰ However, a PHA is still in the early stages of development; therefore, the price gaps are likely to be large. This gap is expected to be reduced as a PHA is commercialized, and technology advancements are achieved.

CONCLUSIONS

In this study, the environmental impacts of a PHA production using sugarcane and corn grains were analyzed using actual production data to provide realistic results. Consequently, the total single scores for cases 1 and 2 were 242.39 and 177.14 mPt, respectively, with case 2 showing small values. Of the three endpoints (Human Health, Ecosystems, and Resources), Human Health had the largest difference between cases 1 and 2. The most significant impact on Human Health was fine particulate matter formation, which is primarily due to the high proportion of raw materials in cases 1 and 2 and the long-distance transportation of sugar to the PHA production site in case 1. Therefore, for the sustainable production of a PHA, low-impact raw materials and the proximity of the factory to raw material sources are essential.

In terms of GWP, a minimum of 0.86 kg CO₂ equiv/kg a PHA and a maximum of 1.59 kg CO₂ equiv/kg a PHA were found for case 1 using sugarcane as feedstock. The GWP values of case 2, which used corn grains as feedstock, had a minimum of 0.01 kg of CO₂ equiv/kg of a PHA and a maximum of 1.96 kg of CO₂ equiv/kg of a PHA. In both cases, a PHA was produced in an environmentally friendly manner through electricity production using renewable energy, RNG production using wastewater, and sustainable feedstock production. These results showed that the first-generation method of PHA production seems to be the most sustainable in terms of GWP at the current level of technology development compared with

the second- and third-generation methods of PHA production and other fossil fuel-based plastics.

A cradle-to-gate LCA was performed owing to a lack of information regarding the "product use" and "product disposal or recycling" stages for a PHA. As previously mentioned, a PHA has higher biodegradability than other bioplastics, which can have a positive effect on the environment. Therefore, cradle-to-grave LCAs for a PHA are required to quantify the effect of biodegradability on the environment. In addition, the cost aspects of increased sustainability were not analyzed but are recommended for future investigation.

ASSOCIATED CONTENT

* Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acssuschemeng.3c04788>.

Physical properties of PHAs; collection method for primary database; relationship between midpoint impact categories and endpoints; BMP test result for wastewater; lifecycle inventory and impact category results for cases 1 and 2; and impact category results for parametric analyses 1 and 2 (PDF)

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ABBREVIATIONS

BMP biochemical methane potential
GHG greenhouse gas

GWPglobalwarmingpotential
 HB hydroxybutyrate
 LCIIlifecycleinventory
 LLClimitedliabilitycompany
 LNGliquefiednaturalgas
 MECMidAmericanEnergycompany
 Mt millionmetrictons
 PA parametricanalysis
 PBATpolybutylenedipateterephthalate
 PCLpolycaprolactone
 PE polyethylene
 PETpolyethyleneterephthalate
 PHApolylactide
 PLApolylacticacid
 PP polypropylene
 PS polystyrene
 PU polyurethane
 PVCpolyvinylchloride
 REC renewableenergycertificate
 RERrestoftheEurope
 RNGrenewablenaturalgas
 ROWrestoftheworld
 TRLtechnologyreadinesslevel
 USLCIUnitedstateslifecycleinventory
 VS volatilesolid
 aPHAamorphouspolyhydroxyalkanoate
 cPHAcrySTALLINEpolyhydroxyalkanoate
 scPHAsemicrystallinepolyhydroxyalkanoate

SYMBOLS

M_w molecular weight, kg/kmol
 T_g glass transition temperature
 T_m melting temperature

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