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Life cycle comparison of petroleum- and bio-based paper binder from distillers grains (DG)

Renil Anthony, Mahmoud A. Sharara, Troy M. Runge, Robert P. Anex*

Biological Systems Engineering, University of Wisconsin, Madison, WI-53706, United States

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ABSTRACT

This study presents a comparative cradle-to-gate life cycle assessment (LCA) of distillers grain (DG) gum, a bio-based paper coating binder, and polyvinyl alcohol (PVA). Non-renewable energy use, greenhouse gas (GHG) emissions, and eutrophication potential were assessed for each binder. Economic, mass, and energy allocation were used to allocate the impacts of DG gum production with co-products (ethanol and livestock feed). DG production non-renewable energy use (269 to 183 MJ) surpassed that associated with PVA production (168 MJ). GHG emissions from DG gum production under mass and energy allocations were 28% and 37% lower than PVA production emissions, respectively. Corn cultivation is responsible for 55% to 78% of the eutrophication impacts of DG gum production under energy and economic allocation, respectively. Changes to natural gas consumption and fertilizer runoff had the largest influence on total energy use, GHG emissions, and eutrophication potential of DG gum production.

1. Introduction

Paper coating is necessary to achieve desired properties such as gloss, smoothness, printability, ink-retention, wear resistance, and novel functionality (Challener, 2011). Coating formulations are composed of pigment(s) that provide the desired functionality; a polymeric binder to contain the pigment(s); and additives to enhance processability and handling (Khwaldia et al., 2010; Challener, 2011). The binder, the most expensive component in paper coating (Challener, 2011), is typically derived from non-renewable petroleum. The volatility of petroleum markets introduces price uncertainty to petroleum-derived commodities, such as binders, which introduces financial risk to production processes that utilize binders. Additionally, petroleum-derived chemical use results in greenhouse gas (GHG) emissions and water quality impacts. In addition, papers coated using polyolefins and other synthetic binders complicate recycling and composting (Aulin and Lindström, 2011).

In an effort to address the challenges associated with synthetic binder materials, binders derived from renewable plant and animal materials have been developed (Weber, 2000). Biopolymers such as hemicellulose, lignin, and chitosan were evaluated for synthesizing coatings and binders (Hansen and Plackett, 2008; Hult et al., 2013; Shogren, 1999; Tripathi et al., 2013). Distillers grains are promising sources of hemicellulose which can be used in the development of paper binders. Distillers grains (DG) and ethanol are co-products of the corn ethanol production process. DG is typically utilized as animal feed owing to its relatively high protein and fat content. However, high concentrations of hemicelluloses lower the quality of DG as animal feed (Keys et al., 1969; Liu et al., 2011). Thus, extraction of hemicelluloses from DG will likely improve its feed value and marketability. Additionally, on-site availability of ethanol, the most common organic solvent used in hemicellulose precipitation (Peng et al., 2012), reduces hemicellulose processing costs. Recovering hemicelluloses for higher value applications using available ethanol while simultaneously improving the feed value of DG will improve the overall value of the ethanol production process.

Previous research in our lab compared the properties of DG gum and polyvinyl alcohol (PVOH) as paper coating binders (Anthony et al., 2015). Polyvinyl alcohol is a common binder for paper and paperboard, specialty coatings and grease resistant coatings imparting high binding power, brightness and good printability (Mishra and Yagci, 2008). DG gum, on the other hand, is a long chain rheological modifier which consists primarily of arabinoxylan chains. The comparative study results indicated that the properties of the two binders were similar for equivalent coating weights suggesting that DG gum has the potential to replace PVOH as a paper coating binder (Anthony et al., 2015). In this study, we compare the environmental impacts of producing DG gum and polyvinyl alcohol through an attributional life cycle assessment of the two binders.

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* Corresponding author.
E-mail address: anex@wisc.edu (R.P. Anex).

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Life cycle assessment (LCA) is a standardized technique that analyzes and evaluates the environmental impacts of products and processes throughout their life cycles: production, use, disposal, recycling and/or landfilling (Weidema, 2000). Attributional LCA evaluates the average physical flows and impacts associated with a product or process, assuming that the production process is fixed. That is, it does not consider indirect effects on other systems or processes that might result from a change in the level of production of the process under consideration, for example through markets responses. In contrast, consequential LCA (CLCA) assesses the consequences of possible consumption decisions, such as changes in demand resulting from policy or technology choices (Plevin et al., 2014). CLCA requires modeling multiple systems of production and comprehensive data and market-based models (Weidema, 2003). Binders based on DG gum are not yet commercially available, so there are not market data such as substitution rates or elasticities that would be required to model market effects. In this study we seek to understand the environmental impacts of DG gum-derived binders relative to those of PVOH. Secondary impacts and indirect market effects on the ethanol and animal feed industries are not considered. This attributional LCA assesses and compares the environmental impacts of functionally equivalent quantities of DG gum and PVOH binders.

2. Goal and scope definition

The goals of this LCA are: (1) To evaluate the cradle-to-gate non-renewable energy use, greenhouse gas (GHG) emissions, and eutrophication potential impacts of producing DG gum from corn DDG obtained from a corn dry grind ethanol plant; (2) Compare the results of the DG gum and polyvinyl alcohol binder through cradle-to-gate LCAs. This study will assess the environmental potential of DG gum to replace polyvinyl alcohol as a binder in paper coating formulations.

3. Functional unit

Tensile strength, water resistance and optical property tests showed that the coating density (g/m^2 paper) of DG gum and PVOH formulations required to achieve equivalent coating performance were not significantly different (Anthony et al., 2015). Accordingly, the functional unit here is defined as 1 kg (dry weight) of binder (DG-gum or polyvinyl alcohol).

4. Scope

This study analyzes cradle-to-gate energy and the environmental impacts of producing DG gum and polyvinyl alcohol (PVOH) binders. The cradle-to-gate study scope is justified because the physical flows and impacts associated with the use and end-of-life stages of the two binders are considered indistinguishable. A primary use of such binders in the United States is in the production of Light Weight Coated (LWC) paper. As mentioned above, equal amounts of each binder type are required to achieve equivalent paper coating performance and the resulting papers have the same density. At end-of-life, according to the U.S. EPA, 38.6% of used LWC paper is recycled, 11.4% is incinerated and about 50% is sent to landfills (AD and PA, 2006). In the paper recycling process, both types of water soluble binder will exit the recycling plant with the wastewater effluent as organic compounds creating biological oxygen demand (BOD). A useful proxy for BOD is chemical oxygen demand (COD). The COD of PVA is approximately 1700 mg O_2/g while the COD of DG gum, modeled as starch, is approximately 1000–1200 mg O_2/g (Schonberger et al., 1997). Although only 11% of LWC paper is incinerated, the higher heating value of PVOH (23.31 kJ/g) (Walters, Hackett, and Lyon 2000) and DG gum (~17 kJ/g) are also quite similar. Landfilling of LWC paper should be unaffected by the paper binder.

Table 1
Corn cultivation process inputs.

Fertilizer	Herbicide	Insecticide	Energy
Urea-ammonium nitrate Diammonium phosphate Potash as K_2O Lime	Acetochlor	Chlorphyrifos	Diesel Gasoline LPG Electricity Natural gas

The system boundary for DG gum, as presented in Fig. 1, includes corn cultivation and harvesting, corn transport to mill, dry mill operations including extraction and processing of hemicelluloses, and animal feed production from wet distillers grains (WDG). The major corn-producing U.S. states, namely, Illinois, Indiana, Iowa, Minnesota, Nebraska, Ohio, Michigan, South Dakota, and Wisconsin were used as the basis for modeling corn production. These nine states account for approximately 80 and 90% of U.S. corn and ethanol production, respectively (Oliveira et al., 2005).

The system boundary for analyzing polyvinyl alcohol (Fig. 1) includes: production of vinyl acetate, polymerization of vinyl acetate to produce polyvinyl acetate (PVA) and further processing of PVA to polyvinyl alcohol through saponification.

The environmental impact categories evaluated were [1] nonrenewable energy consumption, [2] global warming potential and [3] eutrophication potential. We included the nonrenewable energy consumption indicator because one of the motivations for exploring this use of DG is to reduce the dependency of the paper industry on fossil fuels. Similarly, given that DG is a byproduct of corn ethanol production, we included nonrenewable energy consumption and global warming potentials to analyze whether this process furthers the goals of corn ethanol adoption, i.e., energy independence and GHG emission reduction. Eutrophication was included in this assessment due to the large impact of agriculture and in turn agricultural-based bioproducts on water quality and the integrity of aquatic ecosystems. Additional impact categories, while necessary to address the full impacts of the process, could not be assessed due to the lack of detailed data on emissions to air and water. It is our intention that this LCA serves as a screening step for the DG gum binder. More complete process development and testing at larger scale would be required to collect the data required to perform an LCA incorporating a more complete set of impact categories.

5. Methodology

Material and energy input data for corn cultivation were obtained from the National Agricultural Statistics Service database (NASS, 2005). A list of agronomic inputs for corn cultivation as modeled in this study (fuel, fertilizers, herbicides, and insecticides) is presented in Table 1. Based on Shapouri et al. (2002), the average transportation energy requirement for corn grain after harvest was assumed to be 6020 Btu/bushel of corn (250 MJ/Mt corn). The largest farm energy requirements were for harvesting, fertilization, spraying and drying. Water required for corn cultivation was assumed to be supplied entirely by rainfall. This study uses material and energy input values for 1995-97 because the data available for these years are comprehensive (Oliveira et al., 2005).

Nitrate and phosphate losses due to leaching were estimated as a fixed percentage of the total fertilizers applied (Powers, 2005). Similarly, air emissions, such as N_2O from fertilizer nitrogen and CO_2 from lime application, were estimated as a percentage of total applications (Wang et al., 2007; Powers, 2005). The 100-year time horizon global warming potentials (Intergovernmental Panel

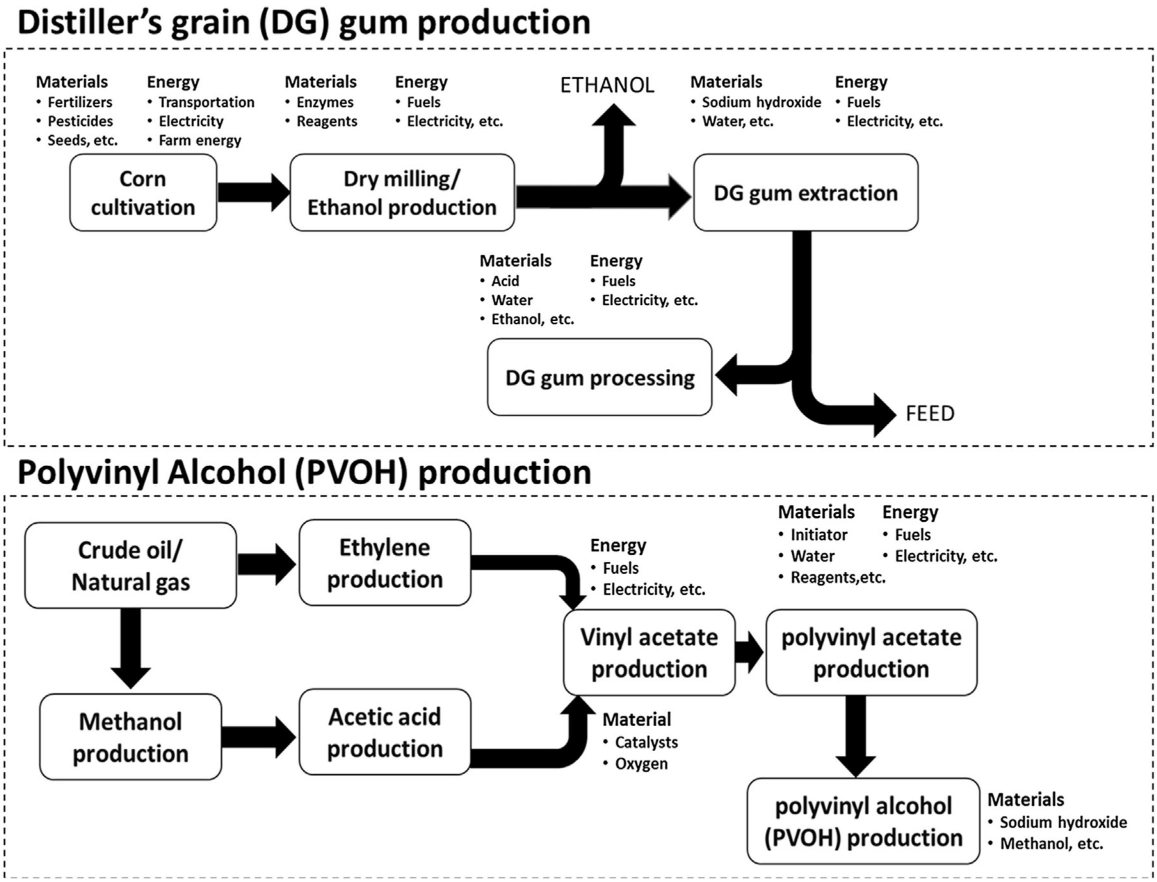

Fig. 1. Flowchart of production stages for distillers grain (DG) gum and polyvinyl alcohol (PVOH).

on Climate Change, 2001) were used to estimate global warming potential (GWP100) in units of equivalent CO_2 mass (kg CO_2 eq). Eutrophication impact was determined using the TRACI (Tools for Reduction and Assessment of Chemical and Other Environmental Impacts) methodology database developed by the National Risk Management Research Laboratory (2003). Eutrophication impacts are denominated in equivalent units of nitrogen mass released to water (kg N eq).

DG gum production was simulated using the process modeling software Aspen Plus v7.3. Production was modeled as two sequential sub-processes: (1) Ethanol/DDG production (dry milling) and (2) DG gum extraction from DDG. The process flow diagram detailing the Aspen Plus process model is presented in Fig. 2. Material inputs for the dry milling unit processes (i.e., liquefaction, saccharification, and distillation) were obtained from the model developed by Kwiatkowski et al. (2006). The material inputs for DG gum extraction and feed processing were based on factorial experiments carried out to optimize the yield and purity of DG gum (Xiang et al., 2014a). In the process model, DG gum extraction was accomplished by first extracting the alkaline-soluble (hemicellulose-rich) DG fraction using sodium hydroxide (NaOH), followed by pH adjustment using hydrochloric acid (HCl) and volume reduction using rotary evaporative drying. The DG gum was then precipitated and washed using a recyclable stream of ethanol (EtOH), and separated by centrifugation. Additional processes such as ethanol distillation and insoluble DG fraction washing and drying were added to model ethanol recycling and additional DG gum recovery. The Aspen model results were verified through comparison with lab-scale study results (Xiang et al., 2014b). Enthalpy requirements were used to estimate process heat requirements, and electricity consumption was estimated by sizing the electrical components used in the process. The Aspen model was linked to Microsoft Excel via a VBA script to iteratively estimate recycle streams.

The material and energy inputs for the production of polyvinyl alcohol were obtained from (Jungbluth et al., 2012). The inputs represent the different production stages: production of vinyl acetate, polymerization of vinyl acetate and further processing to produce 1 kg dry weight of polyvinyl alcohol (PVOH). Available industrial data for PVOH production were utilized, while input data for surrogate processes were used where data for the processes of interest were unavailable. The life cycle inventory data for producing DG gum and PVOH were primarily collected from the ecoInvent Database (version 3) (Weidema et al., 2013). Data that were unavailable in the LCI database were obtained from the literature. As with most LCA studies, energy and material flows associated with the construction of infrastructure, such as buildings, farm equipment and reactors have not been considered.

6. Allocation

Environmental burdens associated with DG gum production processes were allocated among the process coproducts based on a two-stage allocation methodology. The first stage consisted of allocating the burdens from corn cultivation and dry milling to ethanol and distillers grains. No impacts were allocated to corn stover since it is not customary for corn producers in the modeled corn region to harvest and process the stover and corn stover is not used in the dry grind ethanol process. In the second stage of allocation, the burdens associated with the distillers grains and additional environmental burdens associated with DG processing were allocated between the DG gum and feed products. Burdens associated with processes specific to DG gum or feed processing were identified and allocated

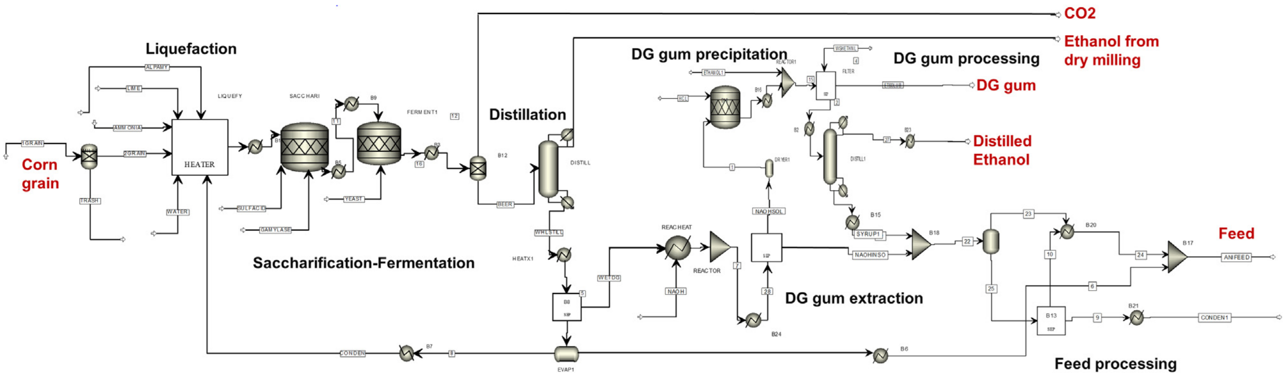

Fig. 2. Flow diagram for modeled process in an integrated corn ethanol dry milling and DG gum production facility.

Table 2
Products and allocation bases for DG gum production.

Products	Economic Allocation		Energy allocation		Mass Allocation
	Price[a], $/kg	Allocation	kJ/kg	Allocation	Allocation
Ethanol	0.75	63%	29700[b]	70%	50%
DG	0.44	37%	12890[a]	30%	50%
Allocation Stage 2: Distillers grain burden further allocated to DG gum and Feed					
DG gum	3.00	83%	16970[c]	32%	30%
Feed	0.25	17%	14270[a]	68%	70%

[a] Xiang et al. (2014a).
[b] Shapouri et al. (2002).
[c] Anderson and Eastwood (1989).

accordingly. At both stages of allocation, burdens were allocated in terms of economic value, energy density and mass ratios of the products. A detailed review of different types of allocation methods and their relative merits is provided (Kodera, 2007). Table 2 contains the percentage of burdens allocated to each process for the various allocation methods used.

6.1. Mass allocation

The mass of the total output of a corn dry milling plant is divided into 3 approximately equal parts as ethanol, distillers grains, and CO_2. Of these three outputs, only ethanol and distillers grains were included in the allocation process and were assumed to have equal mass (Kraatz et al., 2013). The mass output ratio for DG gum and feed after distillers grains processing was obtained from experiments (Xiang et al., 2014a) which yielded 30% DG gum from distillers grains for the extraction conditions assumed. Accordingly, the allocated burdens for DG from the first stage were further allocated based on a mass output ratio of 30: 70 (DG gum: feed).

6.2. Economic allocation

Economic allocation was performed based on the market value ($/kg) of each of the products. The typical price for dried distillers grains as animal feed is 0.25 $/kg. However, the cumulative price for DGs increased to 0.44 $/day due to the combined production of DG gum and feed (Xiang et al., 2014a). This new price of DG was used at allocation stage 1 to assign the burdens based on market value. At allocation stage 2 however, the feed value was assigned the price of 0.25 $/day, which was the typical price of DDG as animal feed during 2015. Since the rate of mass output at allocation stage 2 for the two products was different, Eq. (1) was used to accommodate the difference and calculate the percentage contribution of each product.

$$\%DGgum = \frac{P_{DG} \times M_{DG}}{[(P_{DG} \times M_{DG}) + (P_{Feed} \times M_{Feed})]} \tag{1}$$

Where, PDG = Price of DG gum, $/kg; M_{DG} = % yield of DG gum from DDG

P_{Feed} = Price of Feed, $/kg; M_{Feed} = % yield of Feed from DDG.

6.3. Energy allocation

Energy allocation was based on the application-based energy content (kJ/kg) of products. Ethanol, for example, is a fuel and hence its calorific value was used in allocation. Similarly, the energy contents of DDG and feed were calculated as the metabolizable energy using animal feed characterization techniques. The energy content of DG gum was modeled as that of gum arabic for allocation. Similarly, since the rate of mass output at allocation stage 2 for the two products was different, Eq. (2) was used to accommodate the difference and calculate the percentage contribution of each product.

$$\%DGgum = \frac{E_{DG} \times M_{DG}}{[(E_{DG} \times M_{DG}) + (E_{Feed} \times M_{Feed})]} \tag{2}$$

Where, E_{DG} = Energy content of DG gum, kJ/kg; M_{DG} =% yield of DG gum from DDG

E_{Feed} = Energy content of Feed, kJ/kg; M_{Feed} =% yield of Feed from DDG

No allocation was necessary in evaluating the environmental burdens of polyvinyl alcohol (PVOH) production because no byproducts are generated during its synthesis.

7. Results and discussion

7.1. Non-renewable energy use

Cradle-to-gate energy use and environmental impacts were calculated for DG gum using the three different allocation bases (economic, mass, and energy) and compared to the impacts of PVOH production. For both products, the impacts were broken down by production stage to determine their contribution to total impact. Fig. 3 presents the nonrenewable energy consumption, expressed in megajoules (MJ), to produce 1 kg of DG gum and 1 kg of PVOH. The allocation basis has a significant impact on the total energy consumption for DG gum production. Economic allocation resulted in the highest allocated energy consumption (269 MJ/kg DG Gum), followed by mass allocation (200 MJ/kg DG Gum), and energy allocation (183 MJ/kg DG Gum). As shown in Table 2, under economic (market value) allocation, the majority of non-renewable energy consumption associated with DDG in the first allocation stage is allocated to DG gum since the price of DG gum is much higher than

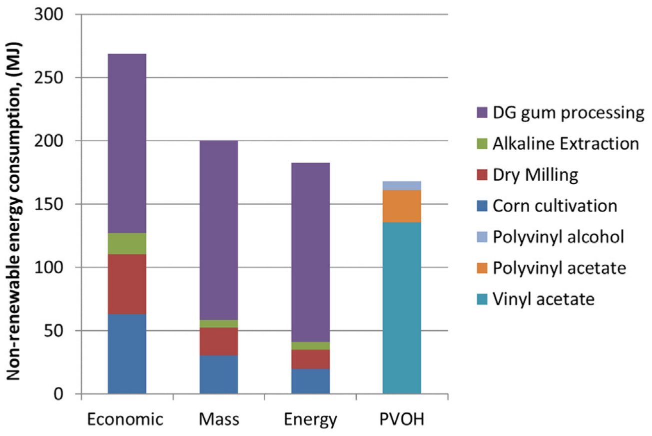

Fig. 3. Non-renewable energy consumption associated with production of a functional unit (1 kg) of DG Gum (under different allocations) and PVOH.

that of DG animal feed (3.0 $/kg and 0.25$/kg, respectively). Conversely, the DG gum energy consumption under mass and energy allocations were 25% and 32% lower, respectively, than under economic allocation since the feed has a higher mass output and energy content than DG gum.

The energy consumption associated with DG gum processing (142 MJ/kg DG Gum) is the same under all three allocation methods since the sub-processes, such as drying of alkaline soluble content and distillation of ethanol after precipitation, were specific to the DG gum stream and no allocation was necessary. DG processing bore the largest non-renewable energy consumption burden due to the energy-intensive nature of the processes involved (drying, distillation, and centrifugation) and energy consumption during the production of input chemicals (hydrochloric acid and ethanol).

Production of PVOH required 168 MJ, of which 81% was consumed during vinyl acetate production. The most widely used process to produce vinyl acetate is the vapor phase ethylene process, which is an oxidative reaction carried out by bubbling ethylene through acetic acid at 120 °C over a palladium chloride catalyst (OARM, 1989). Polyvinyl acetate production is accomplished via emulsion polymerization of vinyl acetate, requiring both continuous mixing and heating (van Herk and Heuts, 2002). The final stage, vinyl alcohol production, requires the least energy input (6.7 MJ/kg PVOH) since it is essentially a partial hydrolysis process in which the polyvinyl acetate ester group is replaced with a hydroxyl group (Hassan and Peppas, 2000).

7.2. GHG emissions

Similar to non-renewable energy use, the GHG emissions associated with DG gum production (Fig. 4) were highest with economic allocation. Furthermore, the rank order of contributions to total GHG emissions was similar to that observed in energy use for the two binders investigated. This was attributable to GHG emissions being closely associated with the consumption of fuels, electricity and petrochemicals. Alcohol fermentation during the dry grind process results in relatively large CO_2 emissions (0.299 Mt CO_2/Mt dry corn grain). This carbon emission, however, is of biogenic origin, i.e., it represents a recycling of atmospheric CO_2 and, therefore, was not included in the total GHG impacts. During corn cultivation, the GHG emissions were primarily from the production and use of fertilizers and combustion of fuel in various farming operations. Similarly, the processes specific to DG gum production contributed the largest fraction of GHG emissions (2.1 kgCO2e/kg DG Gum) primarily resulting from the use of hydrochloric acid and the drying and distillation processes, which were assumed to be fueled with natural gas.

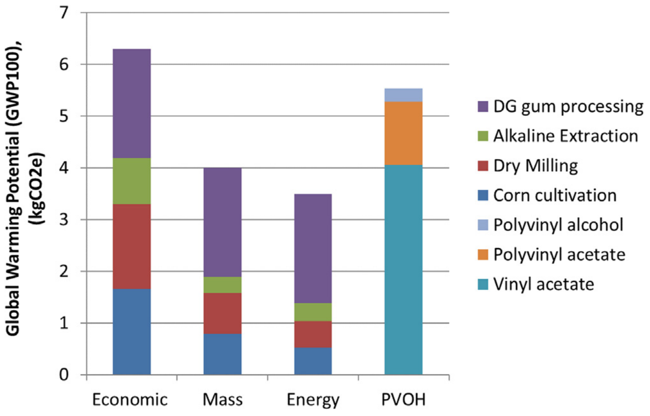

Fig. 4. Global warming potential (GWP100) associated with production of a functional unit (1 kg) of DG Gum (under different allocations) and PVOH.

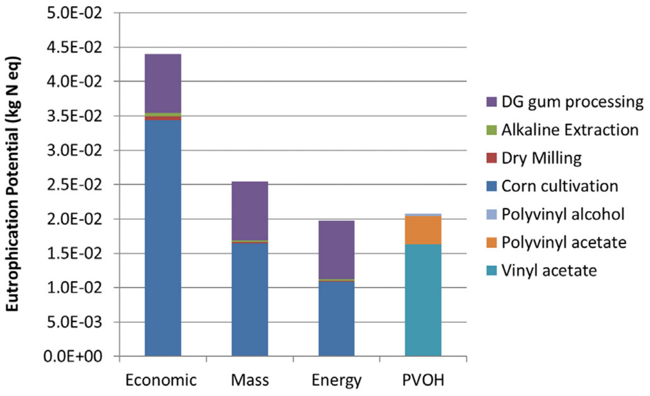

Fig. 5. Eutrophication potential associated with production of a functional unit (1 kg) of DG Gum (under different allocations) and PVOH.

Vinyl acetate production dominated the GHG impacts of PVOH, generating 73% of total emissions. Overall, the GHG emissions for DG gum production under economic allocation was 14% higher than PVOH, whereas under mass or energy allocation, DG gum GHG emissions were 28% and 37% lower than PVOH life-cycle GHG emissions. Thus the choice of allocation basis is a decisive factor in determining the relative life-cycle GHG emissions of the two binders.

7.3. Eutrophication potential

The potential eutrophication impacts of DG gum production were calculated and presented in Fig. 5. The major contributing stage, regardless of allocation basis, was corn cultivation. This contribution was dominated by the impacts of the production and application of urea and phosphate fertilizers, which were the major drivers of nitrogen and phosphorus loss to surface and ground water. DG gum processing life-cycle stage contributed as the second most eutrophication impacts, mainly owing to the production of hydrochloric acid (HCl) which was used in neutralizing the alkaline soluble DG fraction. The eutrophication impacts associated with hydrochloric acid production were primarily the result of large heat and electricity requirements of generating chlorine gas. In the case of PVOH production, the vinyl acetate production stage is the major driver of eutrophication impacts. The highest eutrophication impact in that step was attributed to acetic acid production. The eutrophication potential of DG gum under energy allocation was 5% lower than that of PVOH, while economic and mass allocation of the DG gum inventory yielded higher eutrophication potentials relative to PVOH, 112% and 23%, respectively. Similar to the GWP

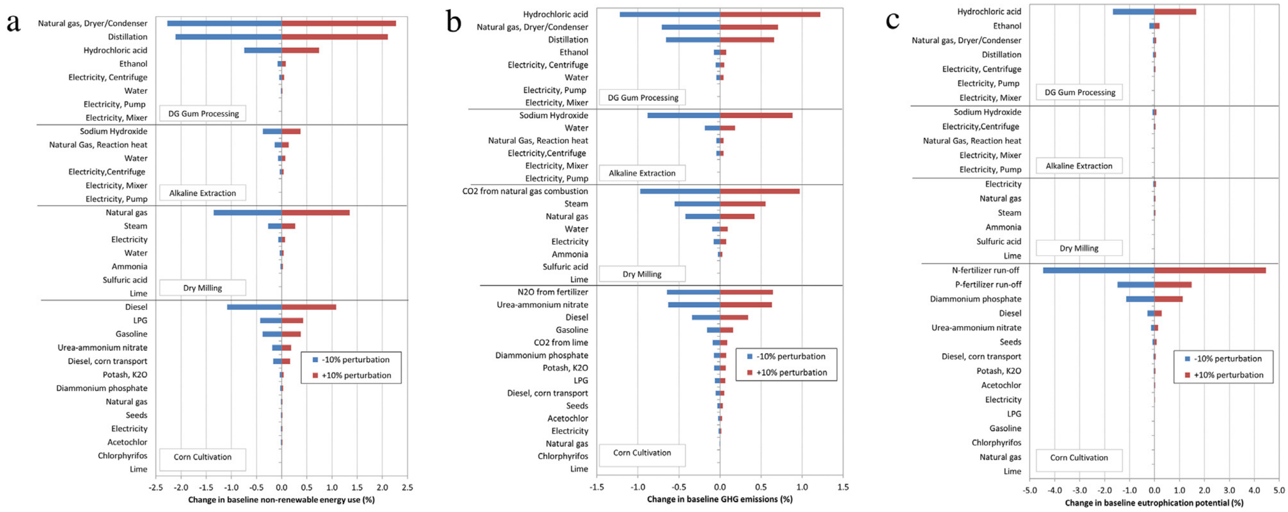

Fig. 6. a.) Sensitivity analysis for non-renewable energy use in producing DG gum under economic allocation. b.) Sensitivity analysis for GHG emissions of producing DG gum under economic allocation. c.) Sensitivity analysis for eutrophication potential of producing DG gum under economic allocation.

impacts, the allocation basis plays a decisive role in determining relative levels of eutrophication impacts of the two binders.

7.4. *Sensitivity analysis*

Sensitivity analyses were performed on DG gum production impacts, using as the baseline the earlier reported potentials for energy use, GHG emissions and eutrophication potential under economic allocation (Fig. 6a–c) [Figures for mass and energy allocations are reported in electronic annex (Supplementary material)]. The relative contribution of input use on each impact category was evaluated by computing the percentage change in life-cycle impact resulting from a 10% perturbation in input usage.

For the DG binder, sensitivities were computed for each allocation basis and impact category. Fig. 6a presents the sensitivity analysis for non-renewable energy use in DG gum under economic allocation. Changes to the energy requirements for the DG processing stage had the largest added impact on the overall process profile, with 5.27% change from the baseline. Process improvements in the heat-intensive steps of drying, distillation, and corn broth cooking, such as heat-integration and the use of renewable energy sources to generate process heat, would reduce the total non-renewable energy impacts associated with DG gum production. Similarly, using animal manure to meet a fraction of the fertilizer requirements for corn cultivation can reduce non-renewable energy use. The use of organic fertilizers like manure, however, can also increase GHG impacts due to high nitrogen volatilization rates sometimes associated with manure use as fertilizer (Xue et al., 2014).

GHG emissions impacts were most sensitive to changes in the gum processing stage which resulted in a 2.76% summed change from baseline GHG emissions (Fig. 6b). The largest single GHG contribution, resulting in a 1.22% change in total GHG emissions, is attributed to hydrochloric acid use. Reducing the intensity of non-renewable energy use typically translates to reductions in process GHG emissions. Furthermore, the GHG performance of the entire system could be improved by utilizing the biogenic CO_2 stream resulting from fermentation for beverage carbonization or as an industrial solvent.

Corn cultivation dominated the eutrophication impacts of DG gum production with a total of 7.73% change to the life-cycle impacts in response to a 10% change in corn cultivation inputs (Fig. 6c). N and P fertilizer loss are the key variables contributing to corn cultivation eutrophication impacts. In modeling corn cultivation N and P loss was assumed to be a fixed fraction of fertilizer applied. Conservation farming practices, such as no-till and contour farming, maintaining riparian buffer strips and growing cover crops can significantly reduce the eutrophication impacts of corn cultivation.

8. Conclusions

Allocation basis had a decisive effect on ranking of DG gum and polyvinyl alcohol GHG emissions and eutrophication potential. Neither binder was clearly superior in terms of GHG emissions or eutrophication potential. Corn cultivation and DG gum processing stages contributed the most to eutrophication potential, GHG emissions, and energy use. Conservation practices to minimize nutrient runoff, heat integration, chemicals recovery and recycling, and renewable energy use can lower the environmental impacts of DG gum production. The studied environmental impacts of DG gum binder production, i.e., global warming potential, energy use, and eutrophication potential were comparable to those for polyvinyl alcohol and with process improvements can be a low-impact alternative for paper production.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.indcrop.2016.11.014.

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