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## Impacts of pre-treatment technologies and co-products on greenhouse gas emissions and energy use of lignocellulosic ethanol production

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A R T I C L E I N F O

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#### ABSTRACT

Life cycle environmental performance of lignocellulosic ethanol produced through different production pathways and having different co-products has rarely been reported in the literature, with most studies focusing on a single pre-treatment and single co-product (electricity). The aim of this paper is to understand the life cycle energy use and greenhouse gas (GHG) emissions implications of alternative pretreatment technologies (dilute acid hydrolysis, ammonia fiber expansion and autohydrolysis) and coproducts (electricity, pellet, protein and xylitol) through developing a consistent life cycle framework for ethanol production from corn stover. Results show that the choices of pre-treatment technology and co-product(s) can impact ethanol yield, life cycle energy use and GHG emissions. Dilute acid pathways generally exhibit higher ethanol yields (20-25%) and lower net total energy use (15-25%) than the autohydrolysis and ammonia fiber expansion pathways. Similar GHG emissions are found for the pretreatment technologies when producing the same co-product. Xylitol co-production diverts xylose from ethanol production and results in the lowest ethanol yield (200 L per dry t of stover). Compared to producing only electricity as a co-product, the co-production of pellets and xylitol decreases life cycle GHG emissions associated with the ethanol, while protein production increases emissions. The life cycle GHG emissions of blended ethanol fuel (85% denatured ethanol by volume) range from -38.5-37.2 g CO2eq/MJ of fuel produced, reducing emissions by 61-141% relative to gasoline. All ethanol pathways result in major reductions of fossil energy use relative to gasoline, at least by 47%.

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1. Introduction

Interest in alternatives to fossil fuels for the transportation sector has motivated research, development and deployment of biofuels. In particular, ethanol produced from lignocellulosic feedstock has seen increasing attention as a light-duty vehicle fuel, giving rise to a large number of production pathways that have been examined in technological reviews (Chen and Qui, 2010; Mabee and Saddler, 2010). While there are "semi-mature" technologies that produce ethanol from corn and sugarcane, ethanol produced from lignocellulosic feedstock remains on the verge of

\* Corresponding author. Tel.: +1 647 223 7442. *E-mail address*: mohammad.pourbafrani@utoronto.ca (M. Pourbafrani). commercialization due to higher capital and operating costs (Stephen et al., 2011).

The potential contributions of lignocellulosic ethanol to reducing petroleum energy use and greenhouse gas (GHG) emissions have been identified in numerous life cycle studies (e.g., von Blottnitz and Curran, 2007) and are sensitive to impacts of different life cycle stages (Mullins et al., 2011; Spatari et al., 2010). However, most LCA studies examine a single biomass pre-treatment process and a single co-product. A broader approach utilizing a consistent analysis framework capable of considering co-product compatibility with a range of pre-treatment processes or comparing the relative benefits and disadvantages of potential pre-treatment technologies and co-product opportunities provides additional valuable insights.

Due to the relatively low market value of ethanol as a fuel, financially-viable ethanol production pathways are expected to







involve co-product production following a biorefinery concept (FitzPatrick et al., 2010). The co-production of value-added products with lignocellulosic ethanol is an emerging opportunity, due to the wide variety of possible products and their potential environmental and economic benefits. Biorefinery concepts in the literature have considered various biomass feedstocks and evaluated a variety of co-products, including electricity (e.g., Delivand et al., 2012), isolated lignin (Pan et al., 2006), acetic acid and hydrogen (Zhang, 2008), protein (Laser et al., 2009a), and antioxidants (Ekman et al., 2013). However, life cycle environmental impacts have only been quantified in a subset of the studies.

A smaller set of studies compared the environmental implications of multiple co-products. Uihlein and Schebek (2009) examined the life cycle of ethanol from wheat straw using dilute acid pre-treatment, with electricity, isolated lignin and xylitol as potential co-products. The study reported results in terms of human health, resource use and eco-system quality, and showed beneficial impacts for lignocellulosic biorefineries. Laser et al. (2009a) studied the environmental impact of the ammonia fiber expansion (AFEX) process considering various co-products (protein, electricity, Fischer-Tropsch liquids and hydrogen) and Switchgrass as feedstock. Laser et al. (2009a) found that the choice of co-product has a significant impact on the environmental performance of the AFEX process. Cherubini and Ulgiati (2010) studied ethanol from corn stover and wheat straw using an autohydrolysis pre-treatment with electricity and lignin-derived phenols as co-products. Ethanol production from both feedstock was reported to have lower life cycle GHG emissions than gasoline: however, corn stover showed better performance than wheat straw in terms of GHG savings, ozone depletion, photochemical oxidation and human toxicity. McKechnie et al. (2011) studied steam explosion (autohydrolysis) with different co-products (steam, electricity and pellet). Pelletizing lignin remaining following fermentation was found to have clear environmental advantage over using lignin for electricity generation. The scope of the above studies, however, is limited to evaluating a single pre-treatment process.

Biomass pre-treatment processes differ in terms of chemical and energy inputs, as well as effectiveness in liberating cellulosic material for subsequent hydrolysis to sugars. These factors impact the life cycle GHG emissions and energy use of lignocellulosic ethanol. As some co-products may be incompatible with some pre-treatment processes (Chiesa and Gnansounou, 2011), pretreatment technology selection may affect co-product options, with potential consequences for life cycle GHG emissions and energy use. Among the most promising pre-treatments under development are dilute acid hydrolysis, AFEX and autohydrolysis (steam explosion) (Cherubini and Strømman, 2011). Prior studies have typically considered a single pre-treatment technology when assessing lignocellulosic ethanol production, including: rice straw ethanol production via dilute acid pre-treatment (Delivand et al., 2012); switchgrass ethanol production via AFEX pretreatment (Bai et al., 2010); poplar ethanol production via autohydrolysis (McKechnie et al., 2011). Based on our knowledge, there are also a limited number of studies (Spatari et al., 2010; Wang et al., 2013) that considered different pre-treatments and a single co-product (electricity). Wang et al. (2013) found ethanol production using steam explosion (autohydrolysis) to provide greater reductions in GHG emissions compared to ethanol production employing dilute acid as a pre-treatment process. Spatari et al. (2010) concluded that AFEX pre-treatment showed more promise than dilute acid for reducing life cycle GHG emissions. Our literature survey was unable to locate a published study that considered both a range of pre-treatment technologies and a range of different co-products within a consistent life cycle framework.

Understanding the energy use and GHG emissions implications of lignocellulosic ethanol requires evaluation of alternative pretreatment technologies and co-products within a consistent life cycle framework. In this study, we compare three conversion pathways that are strong candidates for commercialization (dilute acid. AFEX and autohydrolysis) and four potential co-products (electricity, lignin pellets, xylitol and protein). A single lignocellulosic feedstock, corn stover, is considered to facilitate comparison of the pre-treatment technologies and potential co-products. We evaluate the energy and environmental consequences of these production decisions using a consistent life cycle-based ("well-towheel") framework. Results of the evaluations can inform energy sector stakeholders and government as to how cellulosic ethanol production decisions may impact eligibility under relevant renewable energy policies (e.g., Energy Independence and Security Act (2007)).

#### 2. Methods

#### 2.1. Life cycle assessment

Life cycle inventory analysis models are developed for the set of ethanol pathways described in Table 1. In each pathway, ethanol is the primary product and potential co-products include electricity, lignin pellets, protein, and xylitol. The ethanol product of each pathway is blended with gasoline to produce E85 (83%v/v of ethanol). The E85 pathways include activities associated with corn stover collection and transportation to a biorefinery, conversion of corn stover to ethanol in the biorefinery, blending of the ethanol with gasoline and its distribution, combustion of E85 in a flexible fuel light-duty vehicle and finally, utilizing biorefinery co-products (Fig. 1). Cradle-to-gate modules for energy and material inputs into the main life cycle stages are included in the boundaries (e.g., recovery and processing of petroleum, generation of regional electricity, production of process chemicals). The E85 pathways are compared with those of the gasoline vehicle reference pathway. The life cycle inventory of the reference pathway includes oil recovery, refining, transportation of gasoline and the combustion of gasoline in a light-duty vehicle.

Energy use in terms of total, fossil, and petroleum, as well as GHG emissions are examined. Total energy use includes energy from both renewable (e.g., solar, wind, hydro) and non-renewable (e.g., coal, petroleum, nuclear) sources in addition to the renewable energy in the corn stover feedstock itself. For GHG emissions, CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O emissions are reported as well as CO<sub>2</sub> equivalent

Table 1
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Pathway name	Pretreatment technology	Co-Product(s)	Electricity	Steam
DAEL	Dilute Acid (DA)	Electricity (EL)	Onsite	Onsite
DAPE	Dilute Acid (DA)	Lignin Pellets (PE)	US Midwest	Natural
			Average Mix	Gas
AXEL	Ammonia Fiber	Electricity (EL)	Onsite	Onsite
	Expansion (AFEX)			
AXPE	AFEX	Lignin Pellets (PE)	US Midwest	Natural
			Average Mix	Gas
AXPR	AFEX	Electricity, Protein concentrate (PR)	Onsite	Onsite
AHEL	Autohydrolysis (AH)	Electricity (EL)	Onsite	Onsite
AHPE	Autohydrolysis (AH)	Lignin Pellets (PE)	US Midwest	Natural
			Average Mix	Gas
AHXE	Autohydrolysis (AH)	Xylitol and	Onsite	Onsite
		Electricity (XE)		
AHXP	Autohydrolysis (AH)	Xylitol and Lignin	US Midwest	Natural
		Pellets (XP)	Average Mix	Gas

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