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# COMPARATIVE SYSTEMS ANALYSIS OF THERMOCHEMICAL AND BIOCHEMICAL RECYCLING OF ORGANIC WASTE TOWARDS INDUSTRIAL FEEDSTOCKS

.

BY

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#### DISSERTATION

Submitted to the University of New Hampshire in Partial Fulfillment of the Requirements for the Degree of

> Doctor of Philosophy in Earth and Environmental Sciences

> > May 2012

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#### ABSTRACT

# COMPARATIVE SYSTEMS ANALYSIS OF THERMOCHEMICAL AND BIOCHEMICAL RECYCLING OF ORGANIC WASTE TOWARDS INDUSTRIAL FEEDSTOCKS

BY

Philip Nuss University of New Hampshire, May 2012

Shifting the resource base for chemical and energy production from fossil feedstocks to renewable raw materials is seen by many as one of the key strategies towards sustainable development. The utilization of biomass for the production of fuels and materials has been proposed as an alternative to the petroleum-based industry. Current research and policy initiatives focus mainly on the utilization of lignocellulose biomass, originating from agriculture and forestry, as second generation feedstocks for the production of biofuels and electricity. These activities act on the assumption that significant amounts of biomass for non-food purposes are available.

However, given a certain productivity per area, the current massive growth in global biofuels demand may in the long term only be met through an expansion of global arable land at the expense of natural ecosystems and in direct competition with the foodsector. Although many studies have shown the potential of biofuels production to reduce both, greenhouse gas emissions and non-renewable energy consumption, these production routes are still linear processes which depend on significant amounts of agricultural or forestry production area.

Cascading use, i.e. when biomass is used for material products first and the energy content is recovered at end-of-life, may provide a greater environmental benefit than primary use as fuel. Considering waste and production residues as alternative feedstocks could help to further reduce pressures on global arable land.

This research focused on thermochemical and biochemical technologies capable of utilizing organic waste or forestry residuals for energy, chemical feedstock, and synthetic materials (polymers) generation. Routes towards synthetic materials allow a closer cycle of materials and can help to reduce dependence on either fossil or biobased raw materials. The system-wide environmental burdens of three different technologies, including (1) municipal solid waste (MSW) gasification followed by Fischer-Tropsch synthesis (FTS), (2) plasma gasification of construction and demolition (C&D) wood for syngas production with energy recovery, and (3) forest residuals use in a biorefinery for polyitaconic acid (PIA) production, were assessed using life-cycle assessment.

The *first two studies* indicated that MSW gasification and subsequent ethylene and polyethylene production via FTS has lower environmental impacts than conventional landfilling. In the future, as societies may shift towards the use of renewable energy, power offset by conventional waste-to-energy systems would not be as significant and chemicals production routes may then become increasingly competitive (in terms of environmental burdens) also to waste incineration. While production cost of FischerTropsch derived chemicals seems not yet competitive to fossil-based chemicals provision, future price increases in global oil prices as well as changes in waste tipping fees, and efficiency gains on site of the waste conversion systems, may alter the economics and allow carbon recycling routes to reach a price competitive to fossil-based production routes.

The third study found that plasma gasification of C&D wood for energy recovery has roughly similar environmental impacts than conventional fossil-based power systems. However, process optimization with respect to coal co-gasified, coke used as gasifier bed material, and fuel oil co-combusted in the steam boiler, would allow to significantly lower the system-wide environmental burdens.

The fourth study looked at PIA production from softwood hemicellulose in a stream integrated approach (with the partially macerated wood and lignin being used in other existing processes such as pulp & paper plants for conventional pulp and bioenergy production). The assessment indicated lower global warming potential, energy demand, and acidification, for the wood-based PIA polymer, when compared to corn-based PIA and fossil-based polyacrylic acid (PAA). However, water use associated with wood-derived PIA was found to be higher than for fossil-based PAA production and land occupation is highest for the wood-derived polymer.

It is hoped that results of this dissertation will add to the current debate on sustainable waste and biomass utilization and to establish future supply chains for green and sustainable chemical products.

#### **CHAPTER 1**

#### INTRODUCTION

#### **A.Background**

A growing world population is getting richer and demands more natural resources. Although materials are used more and more efficiently, an average of 90% of all biomass inputs and more than 90% of the non-renewable materials used are still wasted on the way to making products available to the end-user (Lettenmeier et al. 2009). Until to date there are no indications that overall material consumption will decline (Bringezu et al. 2004) and hence it is expected that large amounts of natural resources will be required and wastes continue to be generated in the future (Brunner et al. 2004).

With a growing demand for natural resources, environmental pressures on ecosystems worldwide are increasing. These are to a large extent due to 1.) Anthropogenic greenhouse gases (GHGs) being emitted to the atmosphere causing climate change, 2.) Large-scale landscape changes from the extraction and refining of abiotic resources and significant amounts of wastes generated, and 3.) Land use changes as a result of expanding agricultural lands and human developments (Bringezu 2011).

With the goal to mitigate climate change and reduce dependence on extracted fossil resources, governments and industry invest heavily into the use of biomass as feedstock for fuels, energy, and materials production. Many countries, such as the United States, the European Union, China, Brazil and India, have enacted national policies promoting the utilization of food and non-food biomass (Bringezu and Schütz 2008). These include e.g. mandates for blending biofuels into vehicle fuels and national biofuels production targets. As a direct result of the various biomass policies as well as triggered by high oil prices, global production of liquid biofuels has grown significantly in recent years (REN21 2009).

However, given a certain productivity per area, the current massive growth in global biofuels demand may in the long term only be met through an expansion of global arable land at the expense of natural ecosystems and in direct competition with the food-sector (Bringezu and Steger 2005; Bringezu, Schütz, Arnold, et al. 2009; Bringezu, Schütz, O'Brien, et al. 2009). Although many studies have shown the potential of biofuels production to reduce both greenhouse gas emissions and non-renewable energy consumption, the production routes are still linear processes which depend on significant amounts of agricultural or forestry production area (Bringezu 2009).

#### **B.Towards a More Sustainable Use of Biomass**

Against this background, the UNEP Resource Panel<sup>1</sup> presents a number of options for a more efficient and sustainable production and use of biomass (Bringezu, Schütz, O'Brien, et al. 2009). Although the majority of studies focus on the use of biofuels for transportation, many experts are convinced that our finite resources of biomass may be more effectively used for stationary electricity and heat production as well as material

<sup>&</sup>lt;sup>1</sup> http://www.unep.org/resourcepanel/

applications (Bringezu, Schütz, O'Brien, et al. 2009; Earley and McKeown 2009; Howarth and Bringezu 2008; Weiss et al. 2007). In particular, the use of residues and waste has the potential to widen and complement available biomass resources with the least environmental burden. The UNEP Resource Panel's biofuels report (Bringezu, Schütz, O'Brien, et al. 2009) resulted in the following set of recommendations:



Figure 1-1 Recommendations for more efficient and sustainable production and use of biomass outlined by the UNEP Resource Panel's report on biofuels.

Of particular interest to this dissertation are the recommendations made under the second point "efficient use of biomass". In this context, the concepts of biomass cascading means to use biomass as feedstock for the production of a material (e.g. construction material, chemicals and biobased polymers, etc.) first, before it is either recycled and used for further material applications, or the energy content is recovered from the final waste material at end-of-life (Arnold et al. 2009; Dornburg 2004). *Waste utilization* deals with the recovery of carbon stocks from the waste flow by means of

thermochemical (e.g. gasification) and biochemical (e.g. fermentation and anaerobic digestion) conversion technologies to produce chemical feedstock for further biopolymers production.

#### **C.Future Vision**

Both concepts, i.e. *biomass cascading and waste utilization*, represent the *motivation and backbone* for this study. They are part of a future vision for sustainable resource use in which, following industrial ecology principles, industrial systems use carbonaceous waste materials (including plastics) from household and industry within the socio-industrial metabolism not only for energy recovery (e.g. via incineration) but increasingly for materials production via carbon capture and reuse in a cyclical (cascading) fashion (Bringezu 2009). The systematic reuse of waste carbon<sup>2</sup> (if achieved at high efficiencies) would decrease the need for fuel crops and other non-food plantations and therefore reduce pressures on global arable land. It would also allow the carbon to remain longer in the use phase, hence delaying emissions to the atmosphere.

However, while a small body of literature is available on the economic and environmental implications associated with conversion routes from organic waste towards fuel and energy products (Bez et al. 2001; Chester and Martin 2009; Chiang 2005; S. Jones et al. 2009; Kalogo et al. 2007; Khoo 2009; Sakamoto 2004; Stichnothe and Azapagic 2009), information in this regard on routes towards chemicals and subsequent polymers is still limited. Implementing new technologies is always prone to

<sup>&</sup>lt;sup>2</sup> Metals will be recovered by means of physical recycling schemes and mineral wastes deposited back into the earth's crust.

burden shifting. A comprehensive system analysis is hence required to assess the lifecycle wide performance of carbon recycling compared to conventional systems of waste management and in light of future developments.

#### **D.Dissertation Overview**

This dissertation will investigate the potential system-wide environmental burdens associated with *three different technologies* capable of utilizing *organic waste*<sup>3</sup> or *forest residuals/byproducts* for the generation of chemical feedstock<sup>4</sup>. Life-cycle assessment is used in combination with other tools throughout the dissertation. Chapter two has been published as a book chapter, while chapters three, four, and five, have each been submitted for publication in technical journals.

*Chapter two* presents a detailed discussion of the concept of carbon recycling and its potential application in both developed as well as developing countries. This chapter also includes a preliminary system analysis looking at thermochemical conversion via Fischer-Tropsch synthesis (FTS) to convert municipal solid waste (MSW) feedstock into high density polyethylene – a durable plastic for further use in plastics applications. Results are compared to conventional landfilling operations. Results indicate that the use of organic waste feedstock may be beneficial if greenhouse gas (GHG) emissions associated with landfill diversion are considered.

<sup>&</sup>lt;sup>3</sup> Biodegradable municipal solid waste (BMSW) and construction and demolition (C&D) wood.

 $<sup>^4</sup>$  Syngas (a mixture of CO and H<sub>2</sub>) generated by the plasma gasification system (Chapter 4) is seen as 'chemical feedstock' in this context as it would, besides power production in a steam turbine, allow the generation of platform chemicals via catalytic pathways similar to other thermochemical systems assessed in this dissertation.

*Chapter three* discusses the Fischer-Tropsch synthesis in further detail and investigates the environmental burdens and costs associated with ethylene production from MSW feedstock in the United States. Results are compared to traditional landfilling and incineration (both with energy recovery). Due to the preliminary nature of the inventory data used, the chapter includes a comprehensive sensitivity analysis and uncertainty assessment using Monte-Carlo analysis. Results suggest that, while from an environmental perspective carbon recycling may not be able to compete with incineration with energy recovery under current conditions, if a renewable power mix (envisioned for the future) is assumed to be offset, carbon recycling may become increasingly competitive with conventional incineration systems.

*Chapter four* investigates the life-cycle environmental impacts associated with plasma gasification, a high-temperature process using an electric plasma torch, to convert construction and demolition derived biomass (CDDB) and forest residuals (from the U.S. Northeast) into syngas (CO and H2) for subsequent electricity production. Using inventory data from pilot plants and computer simulations, the study shows that the environmental burdens associated with CDDB and forest residue gasification may be similar to conventional electricity generation. Land occupation is lowest when CDDB is used. The study gives recommendations for further lowering system-wide environmental impacts. By producing a clean syngas, plasma gasification may be further advanced in the future to allow fuels, chemicals, and polymer provision via various catalytic pathways (e.g. Fischer-Tropsch (FTS) or Methanol-to-Olefins (MTO) synthesis).

Chapter five looks at the environmental burdens of producing polyitaconic acid (PIA), a water soluble polymer derived from itaconic acid identified by the U.S. Department of Energy as one of the top 12 value added chemicals, from U.S. Northeastern softwood biomass. The polymer is obtained via a stream-integrated approach in which wood hemicellulose is extracted from the softwood on site a pulp & paper plant and is then diverted to the PIA biorefinery, while following current practices it would be burned on-site together with lignin as 'black liquor'. The partially macerated wood and lignin can be used in existing processes (such as for conventional pulp and bioenergy production) within the pulp & paper plant. Results are compared to cornderived PIA and fossil-based poly acrylic acid (PAA) on the basis of 1 kg of polymer at the factory gate. Softwood-derived PIA is found to result in lower overall environmental burdens when compared to fossil-based polymer production, although water use  $5^{5}$ , eutrophication potential, and land occupation are higher in comparison to PIA's fossilbased counterpart. Wood-derived PIA production in an integrated biorefinery may be an interesting feedstock alternative to current fossil-based pathways and the use of food crops (e.g. corn) and could contribute to a future bio-based economy.

Finally, *chapter six* presents conclusions and an outlook for future research that may be completed using the existing datasets and results from the dissertation. This work may be carried out by the author after completion of this dissertation or by other

<sup>&</sup>lt;sup>5</sup> Water use and water depletion are used interchangeably throughout this dissertation. The latter term derives from the ReCiPe impact assessment method (Goedkoop et al. 2009).

researchers using the collected data together with additional information and/or modeling techniques (e.g. consequential life cycle assessment, system dynamics, etc.).

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#### **CHAPTER 2**

#### WASTE-TO-MATERIALS - THE LONGTERM OPTION <sup>6</sup>

#### A. Abstract

Managing solid waste is one of the biggest challenges in urban areas around the world. Technologically advanced economies generate vast amounts of organic waste materials, many of which are disposed to landfills. In the future, efficient use of carbon containing waste and all other waste materials has to be increased to reduce the need for virgin raw materials acquisition, including biomass, and reduce carbon being emitted to the atmosphere therefore mitigating climate change. At end-of-life, carbon-containing waste should not only be treated for energy recovery (e.g. via incineration) but technologies should be applied to recycle the carbon for use as material feedstocks. Thermochemical and biochemical conversion technologies offer the option to utilize organic waste for the production of chemical feedstock and subsequent polymers. The routes towards synthetic materials allow a more closed cycle of materials and can help to reduce dependence on either fossil or biobased raw materials. This

<sup>&</sup>lt;sup>6</sup> This chapter has been published in: Nuss, P., Bringezu, S., and Gardner, K. H. (2012). "Waste-to-Materials: The Longterm Option." *Waste to Energy: Opportunities and Challenges for Developing and Transition Economies*, A. Karagiannidis, ed., Springer London, London, 1-26.

chapter summarizes carbon-recycling routes available and investigates how in the longterm they could be applied to enhance waste management in both industrial countries as well as developing and emerging economies. We conclude with a case study looking at the system-wide global warming potential (GWP) and cumulative energy demand (CED) of producing high-density polyethylene (HDPE) from organic waste feedstock via gasification followed by Fischer–Tropsch synthesis (FTS). Results of the analysis indicate that the use of organic waste feedstock is beneficial if greenhouse gas (GHG) emissions associated with landfill diversion are considered.

#### **B. Evolution of Waste Management Practices and the Socio-Industrial Metabolism**

#### **B.1. Waste Generation and Management in a Development Perspective**

Since prehistoric times, human activities generated waste materials that were discarded because they were considered of low-value or useless. In the early days, the disposal of wastes did not pose a significant problem, as the population was small and land for the assimilation of wastes was widely available. However, as the human population grew and began to settle in villages and communities, the accumulation of waste became a rogue consequence of life (Tchobanoglous et al. 1993). Since then, the turnover of materials has increased dramatically. This is not only due to global population growth but also due to the enormous growth of goods and assets used per person, in particular in affluent countries. Thus, along with the benefits of technology have also come the problems of disposal of resultant wastes.

Today, approximately 745 kg of municipal solid waste (MSW) are produced per capita per year in the United States (EPA 2009a) and an average of 522 kg MSW in the EU-27 (Eurostat 2009). Modern man consumes between 30 and 75 tons of material per person per year in their companies and households (Bringezu et al. 2003). Of the materials consumed, an average of 90% of all biomass inputs and more than 90% of the non-renewable materials used are wasted on the way to making products available to the end-user (Lettenmeier et al. 2009). Although materials are used more and more efficiently, there are no indications that overall material consumption will decline (Bringezu et al. 2004) and as a result it is expected that large amounts of waste will continue to be generated in the future (Brunner et al. 2004). Although in developing countries the quantity of solid waste generated in urban areas is low when compared to industrialized countries, waste management still remains inadequate (Henry et al. 2006). Rapid economic growth and rise in community living standard in many of the low- or middle-income countries are likely to accelerate MSW generation as well as the complexity and variety in terms of substances present.

Managing these solid waste streams well and affordably is one of the key challenges of the Twenty first century (UN-HABITAT 2010). Traditionally, municipal solid waste management encompasses the functions of collection, transport, resource recovery, recycling, and treatment. The primary goal of MSW management is to protect the health of the population, promote environmental quality, develop sustainability, and provide support to economic productivity (Henry et al. 2006). In addition, climate change has drawn attention to the diversion of biodegradable municipal solid waste (BMSW), such as kitchen and garden waste, from landfills because it has the potential to form methane (a powerful greenhouse gas) under anaerobic conditions. According to the U.S. Environmental Protection Agency (EPA) the four basic options for integrated solid waste management include: (1) source reduction, including reuse, (2) recycling and composting, (3) combustion (waste-to-energy (WtE) facilities), and (4) landfills (Kreith and Tchobanoglous 2002). Examples from e.g. Denmark and Japan suggest that a sustainable waste management system furthermore consists of a stable mixture of technologies and institutions which work flexibly under a defined policy umbrella (UN-HABITAT 2010). Such sustainable waste management systems are designed to mimic an ecosystem that is robust and resilient. Taking a systems-perspective can help to e.g. determine whether materials currently regarded as wastes in one industrial sector could be viewed as raw materials by another sector.

#### **B.2. Future Perspectives for Sustainable Waste Management**

Ecosystems provide the best example of a system that works in a sustainable fashion (Ehrenfeld 2000). One of the central principles in industrial ecology is the vision that industrial systems can use materials extracted and metabolized in a cyclical manner, driven by renewable energy which is used in a cascading manner (R. U. Ayres and L. Ayres 2002). One important measure relates to the systematic reuse of waste products in order to minimize the need to extract virgin raw materials and deplete environmental services (Erkman 1997). However, to date recovery rates for materials such as metals, plastics, paper etc. from the municipal waste stream vary widely, even among industrialized countries. For example, in Germany in 2007 a total of 25% of all MSW generated was disposed to landfills and incinerators (DeSTATIS 2009), while in the United States a total of 67% of all MSW generated in 2008 was discarded (EPA 2009a). Furthermore, the EU landfill directive sets targets to progressively reduce the amount of BMSW disposed to landfills among the EU member states (including Germany), whereas in the United States large amounts of organic waste are sent to landfills. This happens despite the fact that organic waste, being rich in carbon, could serve increasingly as feedstock for thermal and biological processes recovering the carbon for further use as chemical feedstock ('carbon recycling').

The concept of carbon recycling is that, instead of releasing the carbon stored in biowaste into the atmosphere by applying conventional waste management practices such as incineration (for heat) or anaerobic digestion and landfilling (for biogas/landfill gas), the carbon inherent in the organic waste should be seen as a valuable feedstock resource (Bringezu 2009). Instead of carbon-capture and storage, which generally occurs at the beginning of the resource flows (e.g. at oil extraction sites to reduce fossil GHG emissions), the principle of carbon-capture and reuse could be further developed and applied throughout the whole socio-industrial metabolism. Specifically, technologies such as gasification, which allows the generation of a syngas, or anaerobic digestion, for the generation of an upgraded biogas (methane), could be applied<sup>7</sup>. Both syngas and biogas can then serve as feed e.g. for the Fischer–Tropsch synthesis (FTS) to produce base compounds such as Fischer–Tropsch (FT)-naphtha and a number of subsequent chemical products and fuels. In addition, hydrolysis followed by fermentation can be applied to generate a variety of different base chemicals.

When fuels (e.g. FT-diesel, methanol, ethanol, etc.) are produced from organic waste and oxidized by use in combustion engines, the carbon (originally captured in the waste feedstock) is emitted back to the atmosphere. Assuming that the system-wide environmental burdens along this process route are lower than those of conventional fossil-based fuels production routes, this process route would lead to a mitigation of environmental burdens. However, this route of using carbon as fuel is still a linear process through the socio-industrial system which depends on significant amounts of waste feedstock being available (Bringezu 2009).

If, in contrast, synthetic materials for the production of plastics could be synthesized, then the carbon would be kept longer in the use phase and add to the stock of durable goods in the technosphere. The plastic products could potentially be recycled at end-of-life to provide feedstock for either energy generation or as feedstock for the production of syngas in a cascading use scheme. Figure 2-1 exemplifies the concept of carbon recycling, making use of organic waste as feedstock for polyolefins production for the example of Fischer–Tropsch synthesis.

<sup>&</sup>lt;sup>7</sup> In addition, in the future other carbon recycling technologies such as the synthetic tree air-capture unit, developed by Klaus Lackner of the Earth Institute at Columbia University, that stands in the open and captures CO2 on its collector surfaces ("leaves") comprised of anionic resin (Lackner and Brennan 2009), may serve as source of carbon for chemicals feedstock synthesis.



Figure 2-1 Carbon recycling: making use of organic waste via the Fischer-Tropsch synthesis (FTS). Organic waste that could not be recovered via conventional waste recovery systems is gasified and transformed into a FT-naphtha (as well as by-products such as FT-diesel and electricity/heat). FT-naphtha is then transformed into olefins via conventional steam cracking. Polyolefins (PE, PP) and other polymers (PET, PVC, PS, etc.) are generated via polymerization and used for the production of plastic products. At end-of-life these products can either be disassembled and the plastic parts be reused (preferred option if less energy and resource intensive than subsequent FTS) or the carbon and energy recovered via gasification producing a syngas and therefore closing the cycle.

From an environmental perspective the use of waste would be advantageous as, in comparison to virgin greenwood biomass, it has no direct land-use requirement and collection and processing systems are oftentimes already in place. In addition, thermal treatment (i.e. gasification) has the further advantage of contributing to volume reduction, waste disinfection, and concentration of certain toxic elements (e.g. cadmium) in the gasification ash and slag produced<sup>8</sup>. A comprehensive system analysis is required to assess the life-cycle wide performance of carbon recycling compared to conventional systems of waste management.

#### C. Carbon Recycling and Increased Resource Efficiency

#### **C.1. Exemplary Routes of Carbon Recycling**

Organic waste refers to all carbonaceous waste fractions that can potentially serve as feedstock for the thermochemical and biochemical platforms. These include:

- Biodegradable municipal solid waste (BMSW);
- Municipal plastic waste
- Construction & demolition (C&D derived biomass; and
- Liquid waste (e.g. sewage sludge)

BMSW includes all waste fractions of biological origin such as food wastes, paper, cardboard, yard wastes, and bulky wood waste. Of this the cellulose and hemicelluloses fractions can serve as feedstock for hydrolysis with fermentation or anaerobic digestion (kitchen organic waste, green organic waste and paper and cardboard). Plastic waste includes durable goods made from fossil-based<sup>9</sup> plastics such as

<sup>&</sup>lt;sup>8</sup> The removal of hazardous substances from the waste via thermal treatment leads to an ash or slag rich in hazardous substances, potentially enabling efficient recycling of metals from the waste stream in the future (Brunner et al. 2004).

<sup>&</sup>lt;sup>9</sup> In the beginning the thermochemical platform would, amongst other feedstocks, utilize conventional fossil-based plastics as feedstock for the production of syngas and subsequent plastics via the methanol to olefins (MTO) or Fischer-Tropsch synthesis (FTS). However, as this platform is continuously applied to
PE, PP, PET, etc. C&D derived biomass originates from new construction sites and repairs and consists of treated and untreated wood fractions. Organic liquid wastes include municipal sludges such as sewage sludge and animal wastes that can be treated via anaerobic digestion or can be gasified after drying. In addition, industrial organic waste feedstock may be of interest as it often times is more homogeneous than waste from municipal sources.

Organic waste can serve as feedstock for the production of transportation fuels, chemical feedstock and bio-energy using biochemical and thermochemical conversion routes. Current research with regards to biorefineries focuses mainly on the utilization of lignocellulosic materials, originating from agriculture and forestry, as second generation feedstock for the production of bio-fuels and chemicals. Interest in the use of organic waste residues as feedstock is growing. Biochemical processes will either employ anaerobic digestion or hydrolytic mechanisms to break apart the structural polysaccharides (lignocellulose) of the biomass. Alternatively, thermochemical procedures can be used to dehydrate and volatilize the biomass feedstock. Research in bio-refining is proceeding quickly and commercial facilities are expected in the near-future (Hayes 2009). Figure 2-2 provides an overview of the conversion technologies available for the treatment of organic waste. The bold arrows indicate pathways of interest for the synthesis of industrial feedstocks including plastic polymers.

recycle plastic waste by gasification and to produce new plastics from them, this implies that the feedstock origin will slowly shift from fossil- to waste-based plastics (assuming that fossil-based feedstocks will become increasingly scarce over the course of the next decades). At the same time those plastics will slowly fade out that are less appropriate as feedstock or end-product of the recycling pathway.



Figure 2-2 The various conversion technologies possible for the treatment of organic waste (Source compilation adapted from (Hayes 2009)). Organic waste with high water content is treated in the biochemical platform in which either anaerobic digestion or acid/enzymatic hydrolysis are applied. Anaerobic digestion produces a biogas consisting mainly of CH4 and CO2 that can subsequently be converted into a syngas. Hydrolysis produces sugars which can be fermented into a variety of different base chemicals. Thermochemical processes apply gasification or pyrolysis of dry organic waste to derive at a syngas which serves as intermediate for the production of a potentially large number of chemicals (see Figure 2-3). Please note that thermochemical processes, in contrast to anaerobic digestion and hydrolysis with fermentation, are able to utilize a large number of dry organic feedstock sources, including BMSW, plastic waste and C&D waste. Arrows in bold indicate routes of interest for the production of basic chemicals and polymers that would allow cascading use and carbon recycling.

Generally, the thermochemical platform, using gasification, will be superior to the biochemical platform if an organic waste fraction with low water content is used, whereas biochemical conversion generally works better if biomass with high water content is utilized (B. Kamm et al. 2006).

#### C.1.1. Thermochemical Platform ('Dry' Carbon Recovery)

Thermochemical conversion for the production of fuels and chemicals uses either pyrolysis or gasification. Pyrolysis is the thermal treatment of biomass in the absence of oxygen and results in the production of bio-oil, gases, or bio-char. Gasification occurs at higher temperatures and in less oxygen-restricted conditions than pyrolysis and leads to the formation of a synthesis gas (syngas) rich in hydrogen and carbon monoxide. The intermediate products of both processes have the potential as a feedstock for fuel and chemical synthesis via various catalytic pathways (e.g. Fischer–Tropsch synthesis) (Figure 2-3).



Figure 2-3 The potential chemicals from syngas and some of the catalysts involved (Source: compilation adapted from (Spath and Dayton 2003)). Syngas serves as industrial feedstock for the production of a variety of base chemicals. With respect to durable goods for cascading use, FTS and Methanol-to-Olefins (MTO) routes are of particular interest. Both allow the production of olefins which can subsequently be polymerized to derive at polyolefins.

#### C.1.2. Biochemical Platform ('Wet' Carbon Recovery)

Biochemical conversion either uses acids or enzymes to catalyze the conversion of the carbohydrate portion of the biomass (hemicelluloses and cellulose) into intermediate sugars which are then fermented to ethanol and other products. The remaining lignin residue, not processed via the biochemical platform, can be used for heat and power production, or alternatively used in the thermochemical conversion process to produce additional fuels and chemicals.

Anaerobic digestion is a fermentation technique that results in a biogas consisting mostly of CH4 and CO2 but generally carrying impurities such as H2S, H2O, NH3, and particulate matter. Anaerobic digestion is the principal process occurring in landfills (producing what is typically referred to as Land Fill Gas or LFG) and occurs naturally in marshes, wetlands and manure lagoons (R. B. Williams 2007). CH4 for energy production can be obtained by upgrading the biogas. Syngas can be produced by steam reforming the upgraded syngas. Similar to the subsequent steps of the thermochemical platform, syngas can then be utilized for e.g. the production of methanol or FT Naphtha (Figure 2-3). Direct olefin production from upgraded biogas is potentially possible via oxidative coupling (Figure 2-2).

## **C.2.Chemicals and Polymer Production Pathways: What Is Potentially Possible?**

Both thermochemical and biochemical conversion platforms allow the production of a variety of base chemicals and subsequent plastic polymers from organic waste. The reason for looking at base chemicals for the production of synthetic materials rather than fuels is the possibility of a more efficient cascading use in which a durable good (plastic polymer) is produced first and its energy content recovered at end-of-life. Gasification and fermentation both seem to be complementary to each other in terms of polymers they can produce. However, gasification has the clear advantage to be able to utilize a broader variety of waste feedstock (not only lignocellulosic waste but also plastics and C&D waste) and seems to have the advantage that possible toxic substances can be extracted directly from the syngas rather than the organic waste feedstock.

From a traditional base utilizing the natural complex macromolecules of e.g. starch and cellulose as raw materials for the production of biopolymers, the polymer industry is turning attention towards synthetic polymers based on renewable raw materials. Key polymer building blocks include e.g. alcohols such as methanol (C1), ethanol (C2) for the production of polyethylene and polypropylene polymers, glycerol (C3) as a building block for the production of polyurethanes, C3–C6 carboxylic acids (e.g. lactic acid, succinic acid, and itaconic acid) as well as aromatic aldehydes (e.g. 5-hydroxy-methyl-furfural (HMF, C6) and Fischer–Tropsch Naphtha (C5–C12). Strategies differ between replacement of conventional fossil-fuel derived plastics and the development of novel building blocks using biochemical and thermochemical conversion.

In terms of current production volume, ethylene and propylene as well as their derivatives dominate the plastics industry by feeding the polyethylene, polypropylene, ethylene oxide, styrene, polyvinylchloride, and a number of other supply chains (Ren 2009; Skibar et al. 2009). With a production volume of more than 150 million tons, light olefins (e.g. ethylene and propylene) are currently the most important basic petrochemicals to produce plastics, fibers and other chemicals (Ren et al. 2008). In this regard, the methanol to olefins (MTO) route, Fischer–Tropsch Synthesis (FTS) towards FT-naphtha and biogas to olefins routes (either steam reforming or oxidative coupling) seem to provide interesting future pathways for olefins production from organic feedstocks. The MTO route as well as the route from methanol to acetic acid are wellestablished. FT-naphtha could play a key role as a base chemical, from which a variety of chemicals, including polymer building blocks can be obtained.

## **C.3. MSW Feedstock Quality Issues**

The quality of MSW as a feedstock for fermentation or gasification is important in terms of pre-treatment and conversion facility design. Barriers to fermentation and anaerobic digestion of MSW include the ability to effectively separate BMSW material from other wastes whereas gasification requires costly and possibly energy-intensive drying of moist feedstock as well as gas cleanup later in the process chain. Potential variations in feedstock quality and availability, as well as the cost of handling and competing uses such as recycling, compost, waste-to-energy (WtE) and landfill gas generation are further issues of concern.

The composition of MSW varies significantly among countries as well as among regions within individual countries (e.g. urban vs. rural areas). These variations are caused for example by differences in consumer habits, diet and disposal patterns and relate furthermore to the level of affluence and development of the country (Juniper Consultancy Services 2001). One of the biggest challenges faced by developers of waste conversion facilities is the heterogeneity of the feedstock. Varying MSW composition over time<sup>10</sup> is a challenge for most conversion facilities and performance will depend on their flexibility to cope with these changes and to be able to process a number of

<sup>&</sup>lt;sup>10</sup> This includes variations caused by e.g. changing houshold patterns due to the season (e.g. more garden waste in summer); unusual events such as Christmas, holidays; etc.

alternative feedstocks. Methods that can be applied to deal with these issues are: on-site storage and blending, mixing and shredding of the waste, compression and baling of the input, and integration with a Materials Recovery Facility (MRF) to obtain a more homogeneous waste feedstock (Juniper Consultancy Services 2001).

A number of studies on the use of MSW as raw material for the production of fuels and chemicals has been published to date (Aiello-Mazzarri et al. 2005; Champagne 2007; M. Green and Shelef 1989; A. Jones et al. 2007; S. Jones et al. 2009; Li and Khraisheh 2008a; b; Li et al. 2007; McCaskey et al. 1994; Mtui and Nakamura 2005; Sakai et al. 2003; A. Z. Shi, Koh, et al. 2009; J. Shi, Ebrik, et al. 2009; R. B. Williams 2007: 2007). With biochemical Zheng al. respect to conversion et (hydrolysis + fermentation), these studies indicate that, by optimizing BMSW pretreatment and hydrolysis procedures, more than 85% of the waste cellulose fraction can be converted into glucose (Li and Khraisheh 2009) which could be converted to fermentation products such as ethanol and other platform chemicals. Depending on the waste composition, pretreatment methods using dilute sulfuric acid or hydrochloric acid followed by enzymatic hydrolysis and steam and pressure pretreatment have been investigated. However, chemicals (e.g. biosurfactants and antimicrobials) present in the feedstock have the potential, if not removed, to inhibit enzymatic hydrolysis or fermentation resulting in lower yields of the intermediate-products (Li et al. 2007). Most studies looking at fermentation of BMSW have focused on the production of ethanol as biofuels for transportation purposes. All of these studies looked at the conversion of MSW on the lab- or pilot-scale. So far, no commercial plants applying hydrolysis followed by fermentation of the sugars are operating.

In contrast to fermentation, pyrolysis and gasification techniques are widely used for the processing of waste feedstock. As of 2001, there were 110 plants operating in 22 countries processing over 5 million tons of waste per year applying gasification and/or pyrolysis (Juniper Consultancy Services 2001). The majority of these efforts focus on the utilization of MSW and other dry waste fractions to recover energy. However, a wide range of technologies is emerging for the conversion of organic waste to biofuels. These technologies are able to use a wide variety of waste feedstocks, including C&D derived biomass as well as plastics waste.

Anaerobic digestion (AD) of BMSW has advanced mainly in Europe but facilities were recently also built in Canada, Japan, Australia and several other countries (Rapport et al. 2008). AD systems are applied in many wastewater treatment facilities for sludge degradation and stabilization and are used to treat those wastewaters prior to discharge. Some facilities are also employed at animal feeding operations to reduce the impacts of manure and to use it as a feed for energy production from biogas. Of the organic waste fraction of MSW, wet BMSW such as food and yard wastes can be treated in anaerobic digesters. AD therefore represents a commercially available alternative to fermentation techniques. European technologies all use extensive pre- and post-digestion processing units. These include visual manual or robotic sorting and removal of bulky or potentially harmful items, particle size reduction and separation (see (Rapport et al. 2008) for further details) adding to the cost of these technologies. Gasification seems to be favored over biochemical conversion due to the fact that contaminants (alkali metals, halides, sulfur gases, and tars) present in the biodegradable fraction of the waste can (in comparison to pre-sorting and steam-cleaning the biomass itself) be removed from the produced syngas before catalytic conversion (e.g. Fischer– Tropsch) to the intermediate products takes place. If not removed, these contaminants can poison the noble metal catalysts. In addition, gasification utilizes both the lignin as well as the cellulose and hemicelluloses fractions of the BMSW feedstock and has the potential to utilize additional waste fractions such as plastics and C&D waste.

Among the advantages of using organic waste as a primary feedstock for biofuels and bio-materials are that unlike other lignocellulosic feedstocks, MSW has an already well-established collection system and processing infrastructure and is generally available at a negative cost. In contrast to agricultural waste and energy crops which are harvested on a seasonal basis, BMSW provides a year-long supply of feedstock for the biochemical and thermochemical platform. Since the major fraction of MSW consists of organic waste, utilization of MSW provides environmental benefits, as for instance reduction of GHG emissions (CO2, CH4) and landfill space (landfill diversion).

#### C.4. Potentials for Developing and Emerging Countries

Waste gasification, anaerobic digestion and fermentation are technologies still under development. Implementation will require significant investments and initial investors will have to carry the risk of whether they are able to successfully introduce these technologies to the market. While gasification systems may be affordable in affluent countries such as Germany or the United States, they are unlikely to be either appropriate or financially affordable in developing countries in the short-term, simply because citizens have lower incomes and are therefore not be able to pay as much for waste management and carbon recycling. A modern gasifier designed for high-heating value European wastes is likely to require additional fuel inputs to gasify a typical highorganic and relatively wet waste in a developing country. Furthermore, the costs and expertise required to operate and maintain the system in a continuous manner is likely to restrict it to a few cities with most advanced waste collection and separation systems in place in developing or transitional countries. In addition, a novel conversion technology that has not yet been introduced to e.g. the European market is a risky choice for the developing world which requires systems that are guaranteed to be reliable in collecting, treating and disposing of the waste, all year around. Therefore, we envision these technologies to be first introduced in industrialized nations and mega-cities of emerging economies with high volume generation rates of organic waste feedstock.

However, it should be pointed out that in particular the thermochemical platform has the capability of combining safe waste handling of organic waste with the production of energy, fuels and chemical feedstock. According to UN-Habitat data, significant increases in the occurrence of sickness among children living in households where (organic) waste is dumped or burned in the yard can be observed (UN-HABITAT 2008). Organic waste materials can pollute surface and groundwater and therefore pose a threat to the health of people who depend on these water resources for drinking water. The potential of gasification technologies to destroy harmful microorganisms at high temperatures and concentrate hazardous metals in the slag and ash could become of increasing interest for developing countries in the future. In addition, feedstock flexibility would potentially allow utilizing both, organic waste as well as virgin green wood biomass as gasification feed. Finally, operating smaller decentralized conversion facilities would allow the production of energy, fuels and chemicals without having to build large refineries and power plants.

# D.Status of Knowledge: Waste as Feedstock for Thermo- and Bio-chemical Conversion

## D.1. Resource Potentials in the USA and Europe

A *high estimate* of MSW feedstock availability for the production of fuels and chemicals is based on total MSW generation (before recycling) assuming that the whole cellulosic and hemicellulosic fraction of the BMSW stream is used for fermentation into base chemicals or anaerobic digestion into biogas<sup>11</sup>. Gasification on the other hand would be able to utilize the dry fraction of BMSW as well as the plastic waste fraction. The *low estimate* uses the BMSW and plastic feedstocks available after recycling (BMSW and plastic waste disposed of) assuming that recycling and recovery programs remain intact.

Table 2-1 shows the amount of BMSW and plastic waste available each year in the United States, the EU-27 and Germany.

<sup>&</sup>lt;sup>11</sup> There are also typically much smaller amounts of ash, soluble phenolics, fatty acids and other minor components present in the biodegradable waste fraction (Li and Khraisheh 2009). However, as these represent only minor components they are not included in this estimate.

Country/ Region	MSW before recycling	BMSW before recycling	Plastic waste before recycling	MSW final disposal	BMSW final disposal	Plastic waste final disposal
USA (2008) <sup>1</sup>	226 (100%)	144 (64%)	27 (12%)	151	- 83 (55%)	25 (17%)
EU-27 (2007) <sup>2</sup>	259 (100%)	161 (62%)	28 (11%)	105 (100%)	- (no data available)	- (no data available)
Germany (2007) <sup>3</sup>	48 (100%)	18 (38%)	5 (10%)	12 (100%)	0.107	0.249 (2%)

Table 2-1 MSW generation and recovery in the United States and Europe.

All numbers are shown in *million metric tons per year*. MSW before recycling represents the total amount of waste generated per year. It is therefore the maximum amount of MSW available as feedstock for the thermochemical and biochemical platforms. BMSW and plastic waste before recycling represent the annual amounts of the biodegradable waste fraction (paper & board, yard trimmings, food scraps, and bulky wood items) and the plastic fraction, respectively. Waste discarded represents the waste stream after recycling took place and includes disposal to landfills and incineration plants. Please note that incineration plants may convert the waste feedstock into energy. C&D debris is not included. Sources: <sup>1</sup> Data for the year 2008 (EPA 2009a; b); <sup>2</sup> Data for the year 2007 (Eurostat 2009; Skovgaard et al. 2008). Of the waste generated in Germany, 2.3 million tons were bulky waste. It was assumed that 50% (1.15 million tons) of this consisted of wood (Gillner and Pretz 2007). ; <sup>3</sup> Data for the year 2007 (DeSTATIS 2009).

In the United States, 144 million metric tons of BMSW and 27 million tons of plastic waste were available before recycling (high estimate) but only 83 million tons and 25 million tons after recycling (low estimate), respectively. In Germany the situation is even more distinct, with about 18 million tons of BMSW and 5 million tons of plastic waste available before recycling (*high estimate*) but only 107 thousand tons of BMSW and 249 thousand tons available after recycling (*low estimate*).

In order to estimate the amount of waste feedstock available for each platform the moisture content of the organic fractions has to be taken into account. Thermochemical gasification will favor feedstock of low moisture content (0-20%), whereas fermentation can utilize wet feedstock. However, hydrolysis with fermentation cannot utilize the BMSW lignin fraction and lignin-encased biomass used for anaerobic digestion degrades

very slowly. (Li and Khraisheh 2009) state that typical BMSW fractions contain about 40-60% of cellulose, 20-40% of hemicelluloses and 10-20% of lignin and other small contents. It is here assumed that on average 15% of the "wet" kitchen and garden organic waste (food scraps and yard trimmings) cannot serve as feedstock for hydrolysis with fermentation and anaerobic digestion.

In order to obtain the total amount of organic feedstock available for both platforms, the reported wet tons have to be adjusted for known material moisture content. Taking this into account, Table 2-2 shows the *total amount of dry waste feedstock* available for each platform under the high and low estimate.

Under the high estimate 105 million metric tons of dry waste could potentially serve as feedstock for gasification and 18 million tons for fermentation in the United States alone. Under the low estimate, this number decreases to 65 million tons for gasification and 11 million tons, assuming that current recycling and recovery programs remain intact. Results for the EU-27 and Germany also show that significant amounts of dry waste are currently available for both platforms. However, recovery rates, particularly in Germany, are significantly higher with the result that only minimal amounts of feedstock are available under the low estimate. In Germany 13 million tons of dry organic waste would be available for gasification and pyrolysis and 2.7 million tons for biochemical conversion. This decreases significantly when recycling rates are taken into account, with the result that only 271 thousand tons of dry waste remain for thermochemical conversion and 23 thousand tons for biochemical conversion. It is

important to note that C&D debris generation and recycling is not yet included in this estimate.

The *results of this estimate* imply that without changing current recycling and recovery practices, the highest potential for the use of MSW as feedstock for the production of fuels and chemicals currently is in the United States. In Germany the advantages and disadvantages of MSW utilization for fuels and chemicals would need to be compared to current well-established recycling practices. The results also indicate that overall more feedstock would be available for gasification. This is mainly due to the high moisture content of certain waste fractions, such as food scraps and yard trimmings, resulting in less biomass per kg of waste available for biochemical conversion (which utilizes primarily wet organic waste). In contrast to biochemical routes, the thermochemical platform has the capability to utilize all dry BMSW fractions including lignin and plastics waste. The dry wood and plastics fraction of C&D waste can be utilized via gasification and would hence add to the amount of feedstock potentially available for thermochemical conversion. On the other hand, biosolids from sewage sludge can serve as feedstock for the biochemical platform (especially AD).

In addition, a number of other organic waste feedstocks could serve as feedstock for chemicals/polymer production. These include post-consumer wastes such as mixed plastic waste (MPW), tires and auto shredder residues as well as industrial residues (e.g. paper plant sludge, leather shavings) and agricultural and forestry residues.

	USA (in 2008) <sup>1</sup>			EU-27 (in 2007) <sup>2</sup>		Germany (in 2007) <sup>3</sup>						
	Before r	recycling	Disca	rded	Before r	recycling	Before r	ecycling	Disca	rded		
Organic waste fraction	wet weight	dry weight	wet weight	dry weight	wet weight	dry weight	wet weight	dry weight	wet weight	dry weight	Moisture content, % by weight <sup>4</sup>	Platform <sup>5</sup>
Paper & Paperboard	70.234	66.020	31.280	29.403	54.908	51.614	8.121	7.634	0.004	0.004	6%	Т
Wood	14.869	11.895	13.435	10.748	4.403	3.522	1.168	0.934	0.029	0.023	20%	Т
Food Scraps	28.839	8.652	28.114	8.434	100.751	30.225	4.411	1.323	0.028	0.008	70%	В
Yard Trimmings	29.846	11.939	10.523	4.209	0.777	0.311	4.509	1.804	0.046	0.018	60%	В
Plastics	27.261	26.716	25.338	24.831	27.454	26.905	4.975	4.876	0.249	0.244	2%	Т
Waste available for T (only dry) Waste	112.364	104.631	70.053	64.982	86.765	82.041	14.264	13.443	0.282	0.271		
available for B (only wet)*	49.883	17.502	32.841	10.747	86.299	25.956	7.582	2.658	0.063	0.023		
available for both platforms	162.247	122.133	102.894	75.729	173.064	107.997	21.846	16.101	0.345	0.294		

Table 2-2 Organic waste available for the thermochemical and biochemical platform (in million metric tons per year).

It is assumed that paper & paperboard, wood and plastics can be utilized using gasification whereas all wet biodegradable fractions (food scraps and yard trimmings) can serve as feedstock for biochemical conversion. C&D debris is not included. In order to obtain the dry weight of each organic waste fraction, the moisture content (in percent by weight, average values) was subtracted from the wet waste fractions. \*Since the biochemical platform can only utilize the cellulose and hemicelluloses fraction of organic waste, it is assumed that 15% (Li and Khraisheh 2009) of the garden and kitchen waste is comprised of acid and enzyme insoluble lignin and can therefore not be utilized by biochemical conversion. The numbers under "waste available for B" therefore represent the total amount of food and kitchen waste minus 15% (lignin) of the total. Unutilized lignin is often used to produce energy to power the conversion facility. Alternatively it could serve as feedstock for thermochemical conversion. Sources: <sup>1</sup> (EPA 2009b); <sup>2</sup> Assuming a total MSW generation of 259 million tons in 2007 (Eurostat 2009) the share of each waste fraction was calculated using information on the 2003 waste composition in the EU-27 from (Skovgaard et al. 2008). It was therefore assumed that the waste composition does not change over time. <sup>3</sup> (DeSTATIS 2009), 50% of bulk waste generated in Germany is assumed to consist of wood (Gillner and Pretz 2007); <sup>4</sup> Average moisture content percentages taken from (Tchobanoglous et al. 1993). <sup>5</sup> T = Thermochemical platform (Gasification), B = Biochemical Platform (Hydrolysis + Fermentation and Anaerobic Digestion).

## **D.2.** Resource Potentials with a Focus on Developing Countries

The data on solid waste generation and recovery rates in developing countries is scarce (Twardowska 2004). Even a rough estimate of waste amounts and composition as well as recovery and recycling rates is often not possible. When data exists it is difficult to do comparisons even within a city because of inconsistencies in data recording, collection methods and seasonal variations. However, a recent overview of a number of reference cities in developing and emerging countries is given in (UN-HABITAT 2010). According to this study, in low gross domestic product (GDP) cities, waste density can be as high as 400 kg per cubic meter due to high fractions of wet organic waste. A comparison of all reference cities indicates that organic waste is a very large part of the waste stream in all cities investigated. The organic fraction is often between 50 and 70 weight-% of MSW in developing countries. Low- and middle-income countries were found to have relatively high percentages of organic waste (above 45 weight-%) in cities such as Cairo, Cluj, Lima, Pune, and Quezon City<sup>12</sup>.

While in industrialized countries the value of organic waste often times is due to composting or incineration and anaerobic digestion for energy production, in cities of the developing world organic waste is used mainly to feed livestock (especially swine feeding) and to generate compost for land application (UN-HABITAT 2010). It is important to note that the informal sector does most of the recycling related to organic waste in developing countries. This includes street pickers, dump-pickers, itinerant waste buyers and junk shops that collect and deal with the waste feedstock as long as a market

<sup>&</sup>lt;sup>12</sup> See Key Sheet II in Chap. 4 of the (UN-HABITAT 2010) report.

for the product exists. This is, however, only partially true for organic wastes. While food waste may have a market value as animal feed, products made from compost are increasingly being replaced e.g. by chemical fertilizers.

Figure 2-4 shows the amounts of organic waste going to animal feeding, composting or land application in a number of cities around the world. As can be seen from the figure, still large amounts of organic waste feedstock remain unutilized. This fraction could potentially serve as feedstock for thermochemical or biochemical conversion technologies.



Figure 2-4 Destination of organic waste generated in MSW per year in a number of cities around the world (Source: compilation using data from (UN-HABITAT 2010)). For example, in Delhi, India, a total of 2.55 million metric tons of MSW are generated per year. Of this roughly 2.10 million tons consist of organic waste of which 8% (165,565 t) is diverted to composting or land application.

## **D.3. Environmental Performance Evaluation: Waste-to-Chemicals**

A limited number of studies looking at the life-cycle-wide environmental implication of the route MSW to fuels/chemicals have been carried out to date. These studies focus on the production of heat, electricity and fuel, including methanol (Bez et al. 2001), ethanol (Chester and Martin 2009; Kalogo et al. 2007; Stichnothe and Azapagic 2009) and synthesis gas (Khoo 2009) from MSW. All of these products are interesting intermediates on the way to synthetic materials. Methanol could serve as base chemical for the methanol-to-olefins (MTO) route, whereas ethanol can be transformed into

ethylene and subsequent polypropylenes. Syngas acts as a base chemical for a variety of routes including MTO and the FTS and can be used for the production of electricity.

These studies indicate that utilizing the organic fraction of MSW for energy recovery or material recycling may have advantages in terms of GHG emissions savings and to reduce fossil-energy consumption when compared to conventional use including current waste management practices (e.g. landfilling and incineration) and transportation purposes to replace fossil-based petrol. Material recycling through the provision of base chemicals (syngas, methanol, and ethanol) via fermentation and gasification seems to be possible. The overall environmental performance will largely depend on the choice of assumptions made and comparisons applicable (e.g. landfilling with or without landfill gas recovery, inclusion of MSW collection and classification, etc.). Key processes and their performance will be exemplified in the following.

## E. MSW Processes to High-Value Products

This section presents results from one case-study carried out on gasification routes from organic waste to chemical feedstock. We assessed the system-wide global warming potential (GWP) and cumulative energy demand (CED) associated with these routes using attributional life-cycle assessment (LCA). Data collection for the foreground system as shown in the case study below was gathered from available literature. All supplies of materials, electricity, energy carriers, services, etc. were modeled with best available background data from the ecoinvent database (Ecoinvent 2010), the U.S. LCI database (NREL 2008) and other published LCI data sources. SimaPro LCA software was used to calculate the life cycle inventory and carry out the impact assessment.

## E.1. Case Study: Waste-to-Olefins via Fischer-Tropsch Synthesis (FTS)

## E.1.1. Methodology

This analysis compares the use of organic waste for polyethylene (PE) production with the production process using crude oil in a conventional refinery. The goal is to estimate the life-cycle environmental burdens with regards to GWP<sup>13</sup> and CED associated with the production of 1 kg of PE at the factory gate. This analysis is based primarily on U.S. waste collection practices, technological parameters and background data. Electricity inputs to the foreground system (Figure 2-5) are assumed to come from the U.S. power grid<sup>14</sup>. An LCA model is developed following the ISO 14040 standards. It is assumed that organic waste needs to be disposed of and the environmental implications of the processes that generated the waste are therefore excluded. Utilizing MSW as feedstock implies a diversion of the waste, as opposed to the cultivation of additional feedstock (e.g. biomass).

<sup>&</sup>lt;sup>13</sup> Biogenic carbon present in the BMSW feedstock has been excluded from the analysis.

<sup>&</sup>lt;sup>14</sup> The process 'electricity, medium voltage, at grid from the ecoinvent database is used.



Figure 2-5 Process diagram of the MSW to polyolefin process (Source: own compilation). Mixed MSW enters the physical separation facility (MRF) in which the recyclable fractions are separated and a portion of the biodegradable fraction (BMSW) is sent to composting facilities (Waste Classification). The remaining fraction consisting of BMSW is further pre-treated and then converted into syngas via gasification. Additional steps include gas cleaning and conditioning followed by FTS. The products of FTS consist of hydrocarbons of various chain length (syncrude) of which the naphtha fraction (C5–C8) is converted into polyolefins using syncrude upgrading, steam cracking and polymerization.

The need to collect MSW regardless of its end-use implies no significant changes to the collection process and environmental burdens associated with collection are therefore excluded from the LCA. However, in order to process the waste and separate the organic fraction from the remaining waste stream, the waste feedstock needs to be pre-sorted and separated in a Materials Recovery Facility (MRF) (MSW classification)<sup>15</sup>. Further pre-treatment steps for comminution and drying are required (Figure 2-5).

<sup>&</sup>lt;sup>15</sup> (Kalogo et al. 2007) reported that there is some discussion as to whether MSW classification should be included in the analysis. Some authors share the opinion that this step does not need to be included in an LCA. They state the fact that MSW is anyways classified into the different waste fractions because it is economically feasible due to the value of recovered material and because of legal mandates for prior separation.

In this system, MSW is first separated to remove recyclables and shredded and milled to reduce size. It is then dried prior to gasification. The syngas is then cleaned to remove tars, dust, alkali, BTX (benzene, toluene and xylenes) and halons. The cleaning stages envisaged are suitable for subsequent FTS. The six main stages of the life cycle considered are: Classification (sorting), Pre-treatment (Fluff shredding/Drying), Gasification/FTS, FT Syncrude upgrading, Steam Cracking, and Polymerization. Technologies included represent existing processes that are available on pilot or demonstration scale (e.g. gasification system) as well as currently operated processes (i.e. naphtha steam cracker, etc.). The transport from the MSW classification plant to the conversion plant is taken as 50 km. The transport is performed by a 28 t truck.

The analysis exclusively considers MSW destined for landfills and incineration plants. This excludes recyclables which are reused as well as agricultural and forestry residues. Commercial scale FTS plants utilizing organic waste as feedstock do not yet exist, but mass and energy balances on syngas generation from waste feedstock including raw MSW, BMSW and refuse derived fuel (RDF) (Higham et al. 2001; S. Jones et al. 2009; Juniper Consultancy Services 2001; Niessen et al. 1996; Paisley et al. 1989) as well as data for subsequent FT syncrude production (Bechtel 1998; Van Bibber et al. 2007; Choi et al. 1997; S. Jones et al. 2009; Jungbluth, Chudacoff, et al. 2007; Jungbluth, Frischknecht, et al. 2007; Marano and Ciferno 2001) are available from the literature. In fact, several studies indicate that in particular RDF would be a suitable feedstock for gasification-based FTS (S. Jones et al. 2009; Paisley et al. 1989).

We assume that all BMSW destined for landfills and incinerators can be separated from the remaining waste either at the source or during the classification process (during which marketable aluminum, glass, steel and plastic material are recovered). The waste composition and energy content of the BMSW fluff diverted to the gasification plant is assumed to be similar to the U.S. average and is taken from (EPA 2009a; Tchobanoglous et al. 1993). The wet tons of MSW constitute the mass that must be treated in the classification plant. The classification process is modeled based on (Broder et al. 1993) assuming that electricity is used for meeting all of the energy requirements in the classification process<sup>16</sup>. The analysis assumes average recycling and recovery rates as given in (EPA 2009a). Energy use is allocated as follows: BMSW fluff (37%), recyclables (i.e. glass, ferrous, non-ferrous, etc.) (24%), compostable waste (9%), and scraps (30%). The input of BMSW fluff to the conversion plant is calculated based on the average energy content of the waste fluff after classification (11.588 MJ per kg wet BMSW fluff). The BMSW composition is: paper & paperboard (38%), wood (16%), food scraps (34%), and yard trimmings (13%).

Three technologies for converting BMSW fluff into synthetic gas (CO + H2) are selected and compiled from (Ciferno and Marano 2002; S. Jones et al. 2009; Jungbluth, Chudacoff, et al. 2007; Jungbluth, Frischknecht, et al. 2007; Khoo 2009; Niessen et al. 1996; Paisley et al. 1989). Low-temperature wet gas cleaning is envisaged as cleaning process after gasification. Various reports are available describing the in-depth technical

<sup>&</sup>lt;sup>16</sup> The study by (Broder et al. 1993) looked specifically at classification processes that would be able to generate a clean RDF suitable for biochemical ethanol synthesis. We assume that this sort of classification system will produce a pure organic feedstock that would be suitable for subsequent conversion towards chemicals via gasification and AD.

details of those technologies (Belgiorno et al. 2003; Juniper Consultancy Services 2001; Klein 2002; Malkow 2004) and therefore they are not explained in detail. Table 2-3 gives an overview of the conversion technologies selected.

U	, , ,	00	
Data Source	(Niessen et al. 1996; Paisley et al. 1989)	(Juniper Consultancy Services 2001; Niessen et al. 1996)	(Jungbluth, Frischknecht, et al. 2007)
Manufacturer	Battelle (BHTGS) <sup>a</sup>	MTCI ThermoChem <sup>b</sup>	Choren <sup>c</sup>
Туре	Circulating fluidized bed gasifier (CFB)	Bubbling fluidized bed gasifier (BFB)	Two-stage entrained flow gasification (Carbo- V process)
Direct/Indirect heating	Indirect	Indirect	Indirect
Pressurized/Atmospheric	Atmospheric	Atmospheric	Pressurized
Air/Oxygen/Steam- blown	Steam/Air	Steam	Oxygen
Temperature (°C)	766	843	400–600 (1st step) 1,300–1,500 (2nd step)
Feedstock	RDF	RDF	RDF <sup>d</sup>
Water content (%)	20	20	20
Scale	Demonstration	Semi-commercial	Semi-commercial

Table 2-3 Technologies chosen for syngas production using gasification of RDF.

<sup>a</sup>Battelle High Throughput Gasification System (BHTGS); <sup>b</sup>Manufacturing and Technology Conversion International, Inc (MTCI); <sup>c</sup>Only aggregated datasets for the generation of FT-liquids were available; <sup>d</sup> The study by (Jungbluth, Frischknecht, et al. 2007) looks at woody biomass (willow-salix) for FT Diesel production. We assume that pre-plant classification produces an organic feedstock (RDF) that would be acceptable for gasification and subsequent FT liquids production using the Carbo-V process.

Fischer–Tropsch synthesis followed by upgrading of the FT raw liquid yields mainly naphtha and distillate as well as electricity. The FT product of primary interest to this study is naphtha that can be sent to a petroleum refinery. For this study, a *combined credit/allocation approach* is used for allocation. The environmental burdens from conversion and hydrocarbon recovery of the syngas-based FT plants are allocated based on the ratio of the energy content (Lower Heating Value) of the specific fuel relative to the total product. However, electricity co-produced is sold to the grid and can therefore be considered an end-use for FT-liquids and syngas. In order to compensate for this, excess electricity is treated with the credit approach, whereby electricity is assumed to come from the U.S. medium voltage grid.

The Fischer-Tropsch synthesis (FTS) is based on modeling results from (Van Bibber et al. 2007; Jungbluth, Frischknecht, et al. 2007). Two different FT systems are investigated as part of this study. On the one hand, clean syngas generated by the BHTGS and MTCI gasification units is fed into a slurry-bed, iron-based catalyst FT-reactor system based on a model developed from public information and published in (Van Bibber et al. 2007). The FT-model used in their study is based on data originally published by Bechtel/Amoco in 1993<sup>17</sup>. On the other hand, syngas generated from the Choren Carbo-V process is converted into FT syncrude using a cobalt catalyst in a tubular-fixed-bed reactor (TFBR). This process is based on aggregated inventory data (due to confidentiality issues) directly taken from (Jungbluth, Frischknecht, et al. 2007; RENEW 2006). Although assumptions with regards to allocation and emissions profiles may vary somewhat from our LCA model, it was decided to use the aggregated dataset to cross-check results of the other two conversion systems investigated in this paper. Syngas characteristics and conditioning are critical for fuels and chemicals synthesis. High purity syngas (with low quantities of inert gas such as N2) is beneficial as it substantially reduces the size and cost of downstream equipment. Supporting process equipment (e.g. scrubbers, compressors, coolers, Water-Gas-Shift, etc.) can be applied to adjust the conditioning of the product gas. When using an iron catalyst the H2/CO ratio of the

<sup>&</sup>lt;sup>17</sup> Baseline Design/Economics for Advanced Fischer-Tropsch Technology, DOE Contract No. DE-AC22-91PC90027, Topical Report Volume 1, Process Design – Illinois No. 6 Coal Case with Conventional Refining, October, 1994.

syngas should be adjusted to approximately 0.6<sup>18</sup> while for cobalt catalysts a H2/CO ratio near 2.0 should be used (Ciferno and Marano 2002). An authothermal reformer (ATR) using steam and enriched air/oxygen with partial CO2 recycle is used for syngas preparation. It is important to point out, that varying calorific values of the product gas do not affect subsequent FTS as long as H2/CO and impurity levels are met (Ciferno and Marano 2002). No transportation is accounted for as it is assumed that the gasifier, syngas cleaning and FTS platforms are integrated and located within one conversion plant. The full amount of heat and the main part of electricity is used inside the conversion plant (note that excess electricity generated in the FTS platform is delivered to the gasifier to meet some or all of the energy requirements).

(Dancuar et al. 2003) investigated the suitability of FT naphtha for use as a steam cracker feedstock and found that the substance mix was extremely well suited for the production of olefins (ethylene and propylene) by steam cracking. Accordingly, this study assumes the use of conventional naphtha steam cracking for the generation of ethylene. Data from the CPM database (CPM 2010) and the ProBas database (UBA 2010) is used to model the FT naphtha steam cracking process.

The life cycle inventory for high density polyethylene (HDPE) resin production from FT-derived ethylene is based on data from the U.S. life cycle inventory database (Franklin Associates 2007; NREL 2008).

<sup>&</sup>lt;sup>18</sup> The iron-based F-T catalyst promotes the water–gas shift reaction which produces hydrogen for the F-T synthesis reaction (CO + H2O = CO2 + H2).

## E.1.2. Results

The results (see Figure 2-6, Figure 2-7, and Figure 2-8) summarize the systemwide GWP (Figure 2-6) and CED (Figure 2-7) that we estimate would occur if BMSW from the MSW stream were used as feedstock for HDPE production. Figure 2-8 considers the essential fact that the use of BMSW for chemical supply diverts waste from landfills and thus may relieve the overall GHG balance. Results are shown for the functional unit of 1 kg HDPE at the factory gate and are compared to conventional (fossil-based) HDPE production routes. Data for these comes from the U.S. LCI database (HDPE #1) and ecoinvent (HDPE #2).



Figure 2-6 Comparison of the system-wide global warming potential (GWP) of producing 1 kg of HDPE from MSW with its fossil-based counterpart. Data for fossil-based HDPE comes from the US LCI database (HDPE #1) and ecoinvent (HDPE #2). \*The process 'MSW Conversion' for the Choren plant includes gasification and FT-naphtha production (aggregated dataset).



Figure 2-7 Comparison of cumulative energy demand (CED) of producing 1 kg of HDPE from MSW with its fossil-based counterpart. Data for fossil-based HDPE comes from the US LCI database (HDPE #1) and ecoinvent (HDPE #2). \*The process 'MSW Conversion' for the Choren plant includes gasification and FT-naphtha production (aggregated dataset).



Figure 2-8 System expansion accounting for the fact that in a business-as-usual (BAU) case HDPE is produced from petroleum and BMSW is landfilled. The amount of BMSW going to landfills depends on the feedstock requirements of the carbon-recycling systems (Battelle, MTCI and Choren). U.S. landfill net emission factors from the WARM model are used. Data for fossil-based HDPE comes from the US LCI database (HDPE #1) and ecoinvent (HDPE #2).

The comparison shows that GWP associated with the waste-derived polymers is with 2.7-2.3 kg CO2-eq slightly higher than their fossil-based counterparts. A large share of total GWP is due to steam cracking, during which FT-naphtha is converted into ethylene feedstock, as well as MSW classification where the raw MSW is separated into BMSW fluff and other waste fractions. MSW conversion (gasification) leads to roughly 0.21-0.27 kg CO2-eq for the Battelle and MTCI systems investigated. Aggregated data for the Choren plant indicates a GWP of 0.463 kg CO2-eq associated with the conversion step from BMSW to FT-naphtha. During the FTS step, electricity is co-generated, most of which is used internally. However, the MTCI conversion system generates a small amount of excess electricity (0.23 kWh/kg FT liquids) which is assumed to offset conventional electricity from the U.S. national grid (therefore the negative GWP for FTnaphtha generation). The figure shows that the Battelle conversion-plant leads to the highest GWP, followed by the Choren and MTCI design. The reason for this is that, according to the data gathered, the Battelle conversion-plant requires slightly higher inputs of BMSW fluff (by energy content) and electricity to generate a clean syngas for use in the FTS platform. As a result, transportation and energy required for MSW classification contribute more towards GWP and excess electricity exported to the grid is minimal.

In contrast, results for CED are highest for fossil-based HDPE (77.3-81.0 MJ-eq). This is followed by 42.61 MJ-eq for the Battelle conversion facility and 35.97 and 35.57 MJ-eq for the Choren and MTCI systems, respectively. CED of the MSW-based routes is about half that of the conventional fossil-based routes. The CED indicator encompasses non-renewable (i.e. fossil and nuclear) as well as renewable (i.e. biomass, wind, solar, etc.) energy demand. However, renewables account for less than 1% of total CED. The reason that CED for the waste-derived polymers is lower than for their fossilbased counterparts is the fact that by definition the intrinsic energy content of the waste feedstock is not accounted for. In contrast, for fossil-based polymers the direct and indirect energy consumption of e.g. natural gas and crude oil resources used to synthesize the HDPE polymer (some of which is later present as 'feedstock energy' in the final product) are accounted for in the CED values. Similar to GWP, steam cracking and MSW classification, both being very energy intensive processes, account for a large share of CED. The magnitude to which MSW conversion and FTS contribute to total CED depends on the amounts of waste feedstock transported to the gasifier and further energy and materials requirements for the conversion facility. Both CED and GWP for the MSW classification step of the Choren plant are small compared to the Battelle and MTCI conversion systems. This is due to the fact that in the Choren design, which is optimized for FT diesel production, less naphtha is produced and therefore the largest part of CED and GWP associated with BMSW provision to the conversion system is allocated to the FT-distillate (for diesel).

## E.1.3. System Expansion: Avoided Landfilling

When paper, wood, food scraps and yard trimmings are landfilled, anaerobic bacteria degrade the materials, producing methane and carbon dioxide. Although landfills during use operate as net-carbon sink (and as a source afterwards), methane generated is

counted as an anthropogenic GHG because degradation would not take place if the BMSW were not landfilled. The impact of waste diversion from landfills is significant for landfills with no recovery equipment (i.e. landfill gas (LFG) recovery for flaring or electricity generation). In contrast to many countries in Europe, in the United States and many developing countries significant amounts of BMSW are sent to landfills. We use system expansion to compare: (1) GHG emissions associated with the production of 1 kg HDPE from BMSW ('carbon recycling') with (2). GHG emissions associated with landfilling the BMSW and the production of 1 kg of fossil-based HDPE ('business-as-usual (BAU)'). We use emission factors from the WARM model for the United States to estimate the GHG emissions from landfilling the BMSW fluff (EPA 2010a).

Given the U.S. national landfills average from the WARM model, the emissions avoided of removing 1 kg of wet BMSW (with the average waste composition mentioned above) from landfills equals 0.167 kg CO2-eq. As a result, the 'BAU—U.S. National Average' case would lead to a higher system-wide GWP of 5.5–3.1 kg CO2-eq per kg of HDPE produced when compared to the production of 1 kg of waste-derived HDPE (carbon recycling). With 3.1 kg CO2-eq, GHG emissions are lowest for the BAU case in which BMSW, otherwise used as feedstock in the Choren plant, is landfilled and HDPE is produced from fossil-fuels. This is due to the fact that most of the BMSW is used to produce distillate (for fuels) and less for naphtha (for HDPE), and thus a major part of landfill emissions in the BAU scenario are allocated to the distillate. The results indicate that, accounting for average landfills emissions in the U.S., carbon recycling may have the potential to lead to an overall reduction in GWP when compared to current (BAU) waste management and HDPE production practices (Figure 2-8).

However, the magnitude to which landfill diversion results in net GWP reduction depends significantly on whether landfill gas (LFG) recovery and energy recovery equipment is deployed and how effectively it is operated. Flares and generators on-site have the potential to convert methane into CO2, therefore reducing GWP. If only landfill systems without LFG recovery equipment are considered, system-wide emissions of the BAU case would amount to 17.6 to 7.3 kg CO2-eq per kg HDPE (BAU Landfills without LFG Recovery), while only considering landfills with LFG recovery equipment and electricity generation would amount to -11.6 to -2.9 kg CO2-eq in GHG savings (BAU Landfills with LFG Recovery & Electricity Generation), therefore competing with polymer production about the most beneficial use of BMSW to reduce GHG emissions (Figure 2-8). The difference between landfills without LFG recovery equipment and those with LFG recovery equipment illustrates the impact that assumptions on waste diversion can have on the net GWP of the expanded system.

## E.1.4. Discussion

Ignoring the fact that waste needs a safe final disposal, the use of BMSW for the production of polyethylene seems to result in only slightly higher GHG emissions as compared to conventional fossil-based routes. Under the same assumption, CED of the MSW-based polymer production routes was found to be roughly half that of conventional HDPE production. The impact of BMSW landfilling presents the greatest system

uncertainty. Depending on the landfill system chosen, these assumptions can change GHG emissions for the BAU scenarios from positive to negative. When using U.S. national landfills average data, carbon recycling systems investigated in this paper may have the potential to significantly reduce GHG emissions.

However, the required capital for methane recovery installations particularly in developing countries may be lacking, and the low price of commercially produced gas may not make methane recovery an economically viable option. In addition, landfill space may be limited, in particular in urban areas. These conditions could make carbon recycling technologies an attractive option for developing countries and emerging economies in the future. Furthermore, the WARM model makes key assumptions that are critical for the interpretation of our results. For instance, when LFG is recovered for energy production, co-product credits for the displacement of an equivalent amount of energy from the U.S. electricity grid, which is dominated by coal with high GHG emissions, are applied by the model. Therefore, considering a less carbon-intensive electricity mix (e.g. in a future scenario with larger shares of electricity being supplied by renewable energy systems) could change the balance more in favor of carbon recycling systems. Further investigations should also consider other end-of-life waste management techniques such as combustion and composting and include an uncertainty and sensitivity analysis (using e.g. economic allocation). Furthermore, the analysis should be expanded to other impact categories, including total material requirement (TMR), acidification, eutrophication and health impacts, as well as cost. Finally, we assumed that MSW classification is required to obtain a clean gasification feedstock. MSW classification

leads to roughly one fifth of these impacts and therefore excluding this process step from the LCA would result in further lowering of life-cycle wide impacts. This may be justified in circumstances where classification takes place solely due to the value of recovered material (e.g. plastics, metals, etc.) or because of legal mandates prior to separation.

## **F.Conclusion**

Carbon recycling, in which organic waste is recycled into chemical feedstock for material and fuel production, may have the potential to provide benefits in resource efficiency and a more cyclical economy – but may also create 'trade-offs' in increased impacts elsewhere. Preliminary LCA model results derived from the combination of various existing technologies (i.e. MSW classification, gasification, FTS, steam cracking, etc.) and considering landfill diversion, indicate that the use of biodegradable waste for HDPE production could lead to a reduction in system-wide GHG emissions when compared to conventional fossil-based production routes. However, as yet the conversion technologies assessed do not work in the integrated fashion modeled in this paper.

Developing pilot plants for the conversion of BMSW into base chemicals such as HDPE could be a future option in particular for mega-cities in developing countries and emerging economies with high-volume generation rate of organic waste and a lack of landfill gas recovery equipment and landfill space. Varying the process parameters of technologies such as FTS could allow the generation of both base chemicals (such as naphtha for polymers) and liquid fuels. The potential of gasification technologies to destroy harmful microorganisms at high temperatures and concentrate hazardous metals in the slag and ash could become of increasing interest for developing countries in the future.

In summary, the described technologies of carbon recycling may contribute to further develop the waste management sector towards a more sustainable resource management. Besides the recycling of carbon flows, also the extraction, use, recycling and disposal of all material resources should be considered when developing the physical basis of society and economy towards increased sustainable resource management.

## **CHAPTER 3**

## ENVIRONMENTAL IMPLICATIONS AND COSTS OF MUNICIPAL SOLID WASTE (MSW)-DERIVED ETHYLENE<sup>19</sup>

## A. Abstract

Carbon recycling, in which organic waste is recycled into chemical feedstock for material production, may provide benefits in resource efficiency and a more cyclical economy - but may also create "trade-offs" in increased impacts elsewhere. We investigate the system-wide environmental burdens and cost associated with carbon recycling routes capable of converting municipal solid waste (MSW) via gasification and Fischer-Tropsch synthesis into ethylene. Results are compared to Business-as-usual (BAU) cases in which ethylene is derived from fossil resources and waste is either landfilled with methane and energy recovery (BAU#1) or incinerated (BAU#2) with energy-recovery. Monte-Carlo and sensitivity analysis is used to assess uncertainties of the results. The study indicates that MSW-derived ethylene provision may lead to a reduction in global warming potential (GWP), cumulative energy demand (CED), total

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material requirement (TMR), and acidification when compared to BAU#1. In comparison to BAU#2 carbon recycling results in higher GWP, CED, TMR, acidification and smog potential, mainly as a result of larger (fossil-based) energy offsets from energy-recovery. However, if a renewable power mix (envisioned for the future) is assumed to be offset, BAU#2 will gain less credits for energy recovery and impacts may then be similar or higher than carbon recycling routes. This is due to a less carbon- and resource-intensive power mix being offset in the future. Production cost per kg MSW-derived ethylene range between US\$ 1.85 to 2.06 (Jan 2011 US\$). This compares to US\$1.17 per kg for fossil-based ethylene. Waste-derived ethylene breaks even with its fossil-based counterpart at a tipping fee of roughly \$42 per metric ton of waste feedstock. However, uncertainties of the results are high reflecting the preliminary nature of many of the system components.

**Keywords:** Waste-to-chemicals, Life-cycle assessment (LCA), Environmental life cycle cost analysis (LCC), Carbon recycling, Fischer-Tropsch synthesis (FTS)

#### **B. Introduction**

#### **B.1.Challenges**

The use of fossil-fuels is common in energy generation and in the production of chemical feedstocks such as olefins and their subsequent polymers. Shifting the resource base for chemical and energy production from fossil feedstocks to renewable raw materials is seen by many as one of the key strategies towards sustainable development. Current research and policy initiatives focus mainly on the utilization of lignocellulose biomass, originating from agriculture and forestry, as second generation feed-stocks for bioenergy production (i.e. fuels, heat and electricity) (Bringezu and Schütz 2008; Bringezu et al. 2007; Earley and McKeown 2009).

However, given a certain productivity per area, the current massive growth in global biofuels demand may, in the long term, only be met through an expansion of global arable land at the expense of natural ecosystems and in direct competition with the food-sector (Bringezu et al. 2009). Although many studies have shown the potential of biofuels production to reduce both greenhouse gas emissions and non-renewable energy consumption (Menichetti and Otto 2009; Zah et al. 2010), these production routes are still linear processes which depend on significant amounts of agricultural or forestry production area, and a growing demand may contribute to increased land use change and related tradeoffs (Bringezu et al. 2009).

## **B.2.Future Vision**

In the future, carbonaceous feedstocks and all other natural resources will need to be used much more efficiently and their use-phase within the technosphere prolonged. *Cascading use*, i.e. when biomass is used for material products (e.g. chemicals and subsequent polymers, pulp & paper, construction materials) first and the energy content is recovered at the end-of-life, may provide a greater environmental benefit than primary use as fuel (Bringezu et al. 2009; Dornburg 2004; Arnold et al. 2009; Weiss et al. 2007). Going hand in hand with biomass cascading, the *recycling of carbon flows* from *organic waste* (i.e. biodegradable municipal solid waste (BMSW), construction and demolition (C&D) derived biomass, plastics wastes, as well as industrial organic wastes) could help to further reduce pressures on global terrestrial ecosystems. Currently, vast amounts of organic waste are discarded to landfills and incinerators in industrialized countries (EPA 2009a; b; Eurostat 2009). Rapid economic growth and rise in community living standards in many of the low- or middle-income countries are likely to accelerate global waste generation and disposal rates (UN-HABITAT 2010). Disposal happens despite the fact that organic waste, being rich in carbon, could serve increasingly as feedstock for thermochemical (i.e. gasification and pyrolysis) and biochemical (i.e. fermentation and anaerobic digestion) technologies capable of recovering the carbon for further use as chemical feedstock ('carbon recycling').

Instead of releasing carbon stored in the organic waste into the atmosphere, by applying conventional waste management (WM) practices such as incineration, *carbon recycling* aims at capturing the carbon for use as material feedstock (Bringezu 2009, 2011). Technologies such as gasification and anaerobic digestion allow the generation of a syngas (CO and H<sub>2</sub>) or biogas (CH<sub>4</sub>) from organic waste that can serve as feed not only for energy recovery but also for catalytic conversion towards important base chemicals such as naphtha and olefins (Nuss et al. 2012). For example, the Fischer-Tropsch synthesis (FTS) (Spath and Dayton 2003) represents a widely applied route to provide base hydrocarbon mixtures from syngas which could then serve as an intermediate towards lower olefins (i.e. ethylene and propylene) (de Klerk et al 2005; Redman 2005;

Steynberg et al 2004; Dancuar et al. 2003). Ethylene, for instance, represents a chemical intermediate from which a variety of different products can be obtained, in particular polymers such as polyethylene, (PE), polypropylene (PP), polyethylene terephthalate (PET), polyvinylchloride (PVC) and polystyrene (PS) (Morschbacker 2009). Generating subsequent synthetic materials would allow the carbon to stay longer in the use-phase, therefore adding to the stock of durable goods in the technosphere. Polymers generated could be recycled at the end of product-life to provide feedstock for either energy generation or for the production of syngas for chemicals synthesis, therefore closing the loop (Nuss et al. 2012).

However, while a number of studies looked at the potentials of utilizing the organic waste fraction for both thermochemical and biochemical conversion (see (Nuss et al. 2012) for a summary of recent literature), and some recent studies investigated environmental burdens associated with energy and fuels production from organic waste feedstock (Khoo 2009; Bez et al 2001; Chester and Martin 2009; Kalogo et al. 2007; Stichnothe and Azapagic 2009; Münster and Lund 2009), knowledge of the system-wide environmental impacts and economic costs associated with the provision of chemical feedstock and subsequent polymers (not just bio-fuels and energy) is still limited.

#### **B.3. Research Question**

Against this background, the *central research question* addressed in this paper is: What are the life-cycle wide environmental burdens associated with the production of ethylene from BMSW via gasification/FTS and how do these compare to conventional fossil-based production routes as well as to current waste management (WM) practices (i.e. landfilling and incineration)? Furthermore the study includes a cost analysis of some of the carbon recycling routes investigated. Ethylene was chosen as it currently represents one of the most versatile intermediates and highly optimized production routes (therefore being a rigorous reference for comparison) towards industrial polymer production.

## **C.Methods**

#### C.1. Life Cycle Assessment (LCA)

The main research method used is LCA (ISO 2006a; b) to evaluate the systemwide environmental impacts of carbon recycling and comparative systems. The waste-toethylene (*foreground*) system includes: 1.) Physical sorting of the mixed waste feedstock (MSW classification) and diversion of the biodegradable fraction (BMSW) to the gasification/FTS plant, 2.) Gasification and syngas cleaning, 3.) Catalytic conversion (FTS) followed by syncrude upgrading, and 4.) Steam cracking to obtain the final ethylene product. Life cycle inventory (LCI) data comes from publically available sources including scientific publications and technical reports as well as personal communications with experts. Conversion systems are assumed to be located in the U.S. using region-specific inventory data with regards to waste composition, technological parameters and background data to the extent possible. Technologies investigated are existing processes for which data is available on pilot or demonstration scale (e.g. gasification and FTS) as well as currently operated processes (e.g. MSW classification and naphtha steam cracking). All supplies of materials, energy carriers, etc. were modeled with best available (*background*) data from the Ecoinvent (Ecoinvent 2010) and U.S. LCI database (NREL 2008), or other published LCI data sources.

SimaPro 7.3 LCA software was used to develop an attributional LCA model and carry out the impact assessment (LCIA). A combination of commonly used LCIA methods is used to asses global warming potential (GWP) and cumulative energy demand (CED) (Goedkoop et al. 2008), total material requirement (TMR) (MIPS<sup>20</sup> (Lettenmeier et al. 2009; Ritthoff et al. 2002), water use (Goedkoop et al. 2009), and acidification and smog (Bare et al. 2002).

The study does not account for carbon storage in the BMSW feedstock and subsequent biogenic  $CO_2$  losses resulting from the conversion of BMSW into ethylene. Giving *implicit* sequestration credits, assuming a biogenic carbon net flux of zero, can be justified when the carbonaceous feedstock gasified is not reducing carbon stocks, i.e. under the assumption that carbon stocks in a forest or on agricultural land are not affected (E. Johnson 2009); in other words land use patterns remain largely constant (*Appendix: section E.1*).

#### C.1.1.Comparison of carbon recycling with business-as-usual (BAU) scenario

We apply system expansion to compare carbon recycling, which diverts organic waste feedstock and generates 1 kg of ethylene (*functional unit*), to a 'business as usual' (BAU) case in which conventional waste treatment is applied and ethylene is derived

<sup>&</sup>lt;sup>20</sup> Material Input per Service Unit (MIPS)

from fossil resources (Figure 3-1). In this comparison, the environmental burdens associated with the use of MSW for producing 1 kg of ethylene (system [B]) is compared to the environmental load associated with conventional treatment (i.e. landfilling or incineration) of an equivalent amount of waste and the production of 1 kg fossil-based ethylene (system [A]).

Landfills and incinerators generate electricity offsetting conventional power from the U.S. average power grid (Ecoinvent 2010) (avoided burden credits). For carbon recycling systems, we use a combined allocation/credit approach in which environmental burdens from multi-output processes (i.e. MSW classification, FTS, and steam cracking) are allocated based on physical relationships, i.e. energy content for FT-diesel and FTnaphtha from FTS, and mass for outputs from MSW classification (i.e. BMSW, recyclables, compostables, scraps) and steam cracking (i.e. ethylene, propylene and other hydrocarbons). Similar to WM systems in the BAU scenarios, excess electricity coproduced from FTS is treated with the credit approach, whereby electricity is assumed to come from the U.S. average power grid. We include the physical separation of MSW (MSW classification) in the LCA model to be in line with system boundaries of our cost model (see below).



Figure 3-1 Production of ethylene as waste management (WM) option.

#### C.1.2. Uncertainty assessment and sensitivity analysis

We use Monte Carlo (MC) simulation to estimate combined LCI parameter uncertainties of our LCA model. For this, we must define the likely boundaries of each (input and output) parameter, within which variation may take place. We use reported data on the magnitude of uncertainty shown in *Appendix Table 3-4*. Due to a lack of sufficient uncertainty data for the gasification/gas cleaning step, respective probability distributions are based on a semi-quantitative uncertainty analysis based on the Ecoinvent procedure (Frischknecht and Jungbluth 2007) and using the pedigree matrix developed by (Weidema and Wesnæs 1996). Furthermore, we test the sensitivity of assumptions made on the LCA results by varying the input parameters. This includes assumptions made with regards to 1.) Allocation (physical vs. economic), 2.) Future low carbon energy mix (using energy shares from the BlueMap Scenario (IEA 2010)) and energy substitution (substitution of non-baseload<sup>21</sup> power in the U.S. (US EPA 2011a)), 3.) Increased conversion efficiency (from BMSW to FTS), and 4.) Energy inputs to the steam cracker (*Appendix: Section E.7*).

#### C.2. Environmental Life Cycle Costing (LCC) Analysis

LCC (Hunkeler et al 2008) is used to estimate the costs related to carbon recycling systems under investigation. Costs are given per *functional unit* of 1 kg of ethylene. The LCI provides the quantities of material and energy flows, and consumption-related costs are obtained by multiplying these quantities with the respective market prices. Included are also typical costs for residue disposal (e.g. waste water, ash, slag, etc.) in the U.S.

The cost perspective chosen for the LCC assessment is the cost for the product manufacturer. All costs are corrected for *inflation* and recalculated for January 2011 (base year) (USDOL 2011).

<sup>&</sup>lt;sup>21</sup> Using non-baseload power might provide a more accurate estimate of the marginal emission rates (which would most likely be offset by electricity produced as co-product e.g. from novel carbon recycling schemes and waste energy systems) than the average U.S. power mix.

One time *capital expenses* (i.e. equipment and engineering & contingency) (*Appendix: Section F*) are allocated over a depreciation period using the capital recovery factor (CRF):

$$CRF = \frac{[r(1+r)^m]}{[(1+r)^m - 1]}$$

In this equation, m is the number of years over which the cost is allocated and r equals the discount rate (Kirchain and Field 2000). For this study we use: period under consideration (m) = 20 years; and discount rate (r) = 8%. This results in a CRF of 10.19 %. The equipment was assumed to run for 8395 hours a year or 365 days a year and 23 hours a day. The time deducted takes into account possible unplanned downtime.

If not included in the capital cost figures, project contingencies were added to the equipment costs to cover project uncertainty and the cost of any additional equipment that could result from a more detailed design. The total capital investment (TCI) of a conversion plant is then equal to the sum of the equipment and contingency cost. Cost contingencies applied to economic analysis of gasification/FTS facilities range from 10 – 30% (Hamelinck 2004; Niessen et al 1996; Van Bibber et al. 2007; van Vliet et al 2009). For this study, a 15% project contingency is applied to the entire plant (in addition to installation factors given in the literature) and a process contingency of 25% applied to the FTS unit to reflect larger uncertainty relative to other sub-processes (Van Bibber et al. 2007).

*Operating cost* figures include material and energy cost (and revenues), waste treatment cost, operating labor, as well as maintenance cost and other overhead expenses (*Appendix: Section F*).

# **D. Inventory Data**

A brief description of LCI data sources and assumptions is given below. More details are provided in the Supporting Information in the Appendix.

#### **D.1. Carbon Recycling Systems**

We investigate carbon recycling systems using inventory data for three different gasification/FTS systems, namely *Battelle*, *MTCI*, and Choren (Table 3-1). While the Battelle and MTCI gasifiers underwent pilot runs using refuse derived fuel (RDF) (Niessen et al. 1996; Paisley et al. 1989; Jungbluth et al. 2007), inventory data for the Choren system is based on wood chips use only (Jungbluth et al. 2007; RENEW 2006). However, personal communications with Choren indicated that the Carbo-V gasifier was able to successfully convert dry stabilate as well as waste tires into a clean synthesis gas during test-runs in the past (Bilas 2010). This study assumes that MSW classification and subsequent drying will provide a feedstock of sufficient purity to be fed to the Carbo-V gasifier. The Choren datasets are used to cross-check results of the other two conversion systems investigated.

			~	
Route	Battelle	MTCI	Choren	
Subsystem	MSW classification			
Technology	Front-end classification system; Energy requirement: 33kWh/MT MSW			
Product outputs	BMSW, Recyclables, Compostables, Scraps			
Allocation	Based on MSW composition (i.e. BMSW fluff (37%), recyclables (24%), compostable waste (9%), and scraps (30%))			
Data Sources	(EPA 2009b; Broder et al. 1993; Tchobanoglous et al 1993)			
Subsystem	Gasification/Gas cleaning <sup>1</sup>	Gasification/Gas cleaning <sup>2</sup>	Gasification/Gas Cleaning/FTS <sup>3,4</sup>	
Technology	Battelle Circulating fluidized bed gasifier (CFB), BMSW input based on energy content of 11.59 MJ/kg	MTCI Bubbling fluidized bed gasifier (BFB), ), BMSW input based on energy content of 11.59 MJ/kg	Choren Two-stage entrained flow gasifier (Carbo-V process), Tubular-fixed-bed, Co-based, BMSW input based on energy content of 11.59 MJ/kg	
Product outputs	Syngas	Syngas	FT liquids (naphtha and distillate)	
Allocation	- Energy content			
Data Sources	(Niessen et al. 1996; Paisley et al. 1989; Jungbluth et al. 2007; Khoo 2009)	(Niessen et al. 1996; Jungbluth et al. 2007; Khoo 2009; Juniper Consultancy Services 2001)	(Jungbluth et al. 2007; RENEW 2006)	
Subsystem	FTS	FTS	-	
Technology	Slurry-bed Fe-based	Slurry-bed Fe-based	-	
Product	FT liquids (naphtha and	FT liquids (naphtha and		
outputs	distillate), electricity	distillate), electricity	-	
Allocation	Based on energy content (FT liquids), avoided burdens (electricity)	Based on energy content (FT liquids), avoided burdens (electricity)	-	
Data Sources	(Jungbluth et al. 2007; Van Bibber et al. 2007; Marano and Ciferno 2001)	(Jungbluth et al. 2007; Van Bibber et al. 2007; Marano and Ciferno 2001)	-	
Subsystem	Steam cracking			
Technology	Naphtha steam cracker			
Product outputs	Ethylene, propylene, other hydrocarbons			
Allocation	Based on mass (ethylene (48.3 %), propylene (17.6 %), C5+ liquids (16.9 %), and others (17.2 %))			
Data Sources	(CPM 2010; Dancuar et al. 2003; UBA 2010)			

Table 3-1 Life-cycle inventory data sources for carbon recycling systems.

FU: Functional unit. <sup>1</sup>Battelle High Throughput Gasification System (now SilvaGas supplied by Rentech Inc.). <sup>2</sup>Manufacturing and Technology Conversion International, Inc (MTCI) (now TRI technology). <sup>3</sup>Only aggregated dataset for the generation of of FT-liquids were available. <sup>4</sup>The study by (Jungbluth et al. 2007; RENEW 2006) looks at woody biomass (willow-salix) for FT-diesel production. We assume that pre-plant classification produces an organic feedstock acceptable for the gasification/FTS using the Carbo-V process.

#### D.2. Business-as-Usual (BAU) Cases

Carbon recycling systems are compared to a BAU case in which 1 kg ethylene stems from fossil fuels (NREL 2008) (i.e. natural gas in the U.S.) and BMSW is either landfilled or incinerated (both with energy recovery) (Figure 3-1). When organic waste is landfilled, anaerobic bacteria degrade the materials, producing a landfill gas (LFG) consisting mainly of methane and carbon dioxide. Methane not recovered is counted as an anthropogenic greenhouse gas (GHG) because degradation would not take place if the BMSW were not landfilled. GHG emissions associated with U.S. average landfills receiving BMSW come from the WARM model (EPA 2010a) according to which 0.166 kg CO<sub>2</sub>-eq are generated per kg BMSW<sup>22</sup>. All other impacts are derived using ELCD data in which 22% LFG is recovered for flaring and 28% used for energy generation (0.10 kWh/kg BMSW) offsetting conventional grid power (European Commission 2011).

Inventory data for *waste incineration* is taken from the ELCD database (European Commission 2011). Most of the waste-to-energy plants in the U.S. produce electricity (EPA 2010a). Given a combustion efficiency for RDF facilities of 16.3%<sup>23</sup> (EPA 2010a) and an energy content of 11.588 MJ/kg BMSW (Table 3-1), 0.53 kWh/kg BMSW are delivered to the grid offsetting conventional power. *TMR data* for both incineration and landfilling were obtained from (Schmidt 2003).

 $<sup>^{22}</sup>$  Includes energy offsets (U.S. average power, 0.775 kg CO<sub>2</sub>-eq/kWh). U.S. average landfills include systems with/without landfill gas (LFG) recovery equipment either for flaring or energy recovery.

<sup>&</sup>lt;sup>23</sup>This includes a 5% line loss rate for WTE facilities utilizing RDF according to (EPA 2010a).

#### **D.3. Energy Systems**

We use data from (Ecoinvent 2010) for current U.S. average power (0.76 kg  $CO_2$ eq/kWh) and from (Ecoinvent 2010; IEA 2010; Koornneef et al. 2008) for a future lowcarbon power mix (*IEA BLUE Map Scenario*, 0.206 kg  $CO_2$ -eq/kWh)<sup>24</sup> (see sensitivity analysis), to model energy in- and outputs (offsets) as part of the foreground system The marginal power mix (1.31 kg  $CO_2$ -eq/kWh)<sup>25</sup> comes from plants that are more likely to respond to incremental changes in electricity supply and demand. We approximate emissions factors from a marginal electricity mix consisting of 100% coal using data from (NREL 2008) (Appendix: Section D.7).

#### **E.Results and Discussion**

A brief description of LCIA results is given below. More details are provided in the Supporting Information in the Appendix.

#### E.1. Environmental Assessment

Table 3-2 summarizes the system-wide GWP, CED, TMR, acidification, smog, and water use that are estimated to occur if BMSW were used as feedstock for 1 kg ethylene production. The BAU scenarios show the environmental burdens of generating an equivalent amount of ethylene from fossil raw-materials in the U.S. and take into account that BMSW would be either landfilled or incinerated. Relative contributions of

<sup>&</sup>lt;sup>24</sup>The goal of including the BLUE Map mix in our analysis is not to exactly estimate its potential future impacts, but rather use it as a widely accepted proxy to investigate how an envisaged low-carbon energy mix, based on large shares of renewables, will impact results of our LCA model.

<sup>&</sup>lt;sup>25</sup>Consisting of electricity from coal-fired power plants.

unit processes to carbon recycling and BAU scenarios are shown in Figure 3-2 and Figure

3-3, respectively.

Impact category	Unit	MSW Route	SD	2.5%	97.5%	BAU#1, Landfilling	<b>BAU#2</b> , Incineration
Battelle							
IPCC GWP 100a	kg CO2 eq	1.959E+00	3.130E-01	1.400E+00	2.700E+00	4.884E+00	-5.031E+00
CED	MJ eq	3.035E+01	9.250E+00	1.860E+01	5.440E+01	8.214E+01	-3.142E+01
TMR	kg	3.696E+00	5.520E-01	2.760E+00	5.000E+00	1.441E+01	5.924E-01
Acidification	H+ moles eq	7.499E-01	1.250E-01	5.440E-01	1.030E+00	1.547E+00	-4.438E-01
Smog	g NOx eq	9.172E-03	1.600E-03	6.520E-03	1.290E-02	8.319E-03	1.156E-03
Water use	m3	1.069E-02	1.980E-03	7.290E-03	1.500E-02	1.665E-03	4.203E-02
МТСІ							
IPCC GWP 100a	kg CO2 eq	1.786E+00	3.140E-01	1.310E+00	2.550E+00	4.404E+00	-4.199E+00
CED	MJ eq	2.745E+01	8.040E+00	1.700E+01	4.570E+01	8.129E+01	-1.725E+01
TMR	kg	3.259E+00	7.440E-01	2.030E+00	4.930E+00	1.302E+01	1.029E+00
Acidification	H+ moles eq	6.900E-01	1.710E-01	3.940E-01	1.050E+00	1.542E+00	-1.855E-01
Smog	g NOx eq	8.746E-03	2.520E-03	4.280E-03	1.410E-02	7.619E-03	1.403E-03
Water use	m3	1.029E-02	3.400E-03	4.280E-03	1.750E-02	1.464E-03	3.649E-02
Choren							
IPCC GWP 100a	kg CO2 eq	1.990E+00	2.360E-01	1.570E+00	2.510E+00	3.059E+00	-1.903E+00
CED	MJ eq	2.779E+01	7.850E+00	1.910E+01	4.380E+01	7.895E+01	2.187E+01
TMR*	kg	-	-	-	-	9.179E+00	2.232E+00
Acidification	H+ moles eq	6.095E-01	1.270E-01	4.360E-01	9.310E-01	1.528E+00	5.274E-01
Smog	g NOx eq	7.864E-03	2.730E-03	4.620E-03	1.530E-02	5.685E-03	2.084E-03
Water use	m3	1.261E-02	2.030E-03	8.910E-03	1.690E-02	9.086E-04	2.120E-02

Table 3-2 Environmental burdens associated with the production of 1 kg ethylene from MSW and when compared to respective BAU scenarios.

\*TMR has been excluded due to the aggregated nature of the Choren dataset. SD: Standard deviation. 2.5%/97.5% represents the 95% confidence interval according to MC analysis. A total of 1000 MC runs were carried out for each parameter and system under investigation.



Figure 3-2 Relative contributions of unit processes to system-wide environmental burdens of carbon recycling technologies. Negative burdens indicate energy offsets from excess electricity co-produced at the FTS unit. \*TMR for the Choren system has been excluded due to the aggregated nature of the dataset. \*\*FTS for the Choren plant includes gasification and FT-naphtha production (aggregated dataset). AP: Acidification potential; WU: Water use.



Figure 3-3 Relative contributions of unit processes to system-wide environmental burdens of the Battelle-related BAU scenarios (BAU#1: Landfilling; and BAU#2: Incineration). The BAU case is based on MSW required for the provision of 1 kg of ethylene via the Battelle system (18.9 kg BMSW per kg ethylene) and include: 1.) Fossil-based ethylene production, 2.) Conventional waste treatment, and 3.) MSW classification. The overall trend is similar for all three BAU scenarios (i.e. Battelle, MTCI and Choren), with differences in absolute environmental burdens between carbon recycling routes and respective BAU scenarios (Table 3-2), being due to varying amounts of MSW required per kg of ethylene. Negative burdens indicate avoided energy production due to LFG recovery or waste incineration. AP: Acidification potential; WU: Water use.

Results of the assessment show that for all impact categories, except smog and water use, the BAU#1 scenario in which ethylene is generated from fossil resources and BMSW is landfilled leads to the highest environmental burdens (Table 3-2). Applying allocation based on physical relationships, 9.5 kg BMSW (Choren), 16.4 kg BMSW (MTCI), and 18.9 kg BMSW (Battelle) are required for the generation of 1 kg of ethylene at the factory gate. The lower feedstock requirements of the Choren plant is due to a higher lower-heating value (LHV) conversion efficiency of 53% (from BMSW to FTliquids) when compared to only 27% and 31% for the Battelle and MTCI systems, respectively (*Appendix: Table 3-8*). Furthermore, the Choren plant is optimized to generate FT-diesel rather than naphtha, and therefore a major part of the energy and material inputs are allocated towards the production of fuel.

BAU#1 (landfilling) impacts in all categories investigated are positive, even if the avoided burden credits of LFG energy recovery are taken into account (Figure 3-3). Hence, BAU#1 impacts are higher for the Battelle and MTCI landfilling scenario, diverting the largest amount of BMSW, than for the Choren landfilling scenario (Table 3-2).

On the other hand, larger energy offsets associated with BMSW incineration (BAU#2) drastically reduces environmental impacts to all categories, except water use (Figure 3-3). As a result, BAU#2 impacts are lower when compared to their respective carbon recycling routes (Table 3-2). Negative GWP, CED, and acidification are due to the avoided impacts of fossil energy offsets being larger than the cumulative environmental burdens of fossil-based ethylene production and MSW classification. However, using ELCD data, water use was found to be highest for BAU#2 scenarios. In general, BAU#2 impacts are lowest (though highest for water use) and a function of the amount of BMSW diverted.

Impacts to GWP, CED, and TMR are, to a large extent, due to energy inputs (heat) to the steam cracker (Figure 3-2), converting FT-naphtha into light olefins. During the FTS step, electricity is co-produced, most of which is used internally. However, both the MTCI and Battelle conversion systems generate a small amount of excess electricity (0.26 and 0.17 kWh/kg FT-liquids) offsetting conventional electricity from the U.S. national grid (negative burdens in Figure 3-2). Impacts from MSW classification to all categories are due to electricity requirements (33kWh/MT) to the front-end sorting system.

Impacts from gasification are largest for TMR, acidification, smog, and water use. For TMR, this is due to on-site inputs of gasifier bed materials, chemicals (syngas cleanup) and feedstock transport. Acidification impacts are largely a result of on-site emissions of sulfur dioxide and nitrogen oxides during gasification, FTS, and steam cracking (off-gases). Similarly, smog potential is mainly due to nitrogen oxides and volatile organic compounds emitted on-site. While typical off-gas emission profiles from gasification are based on actual organic waste treatment (Khoo 2009) and those of FTS on biomass gasification (Marano and Ciferno 2001), we use emissions profiles from conventional fossil-based naphtha steam crackers using crude-oil because other data are unavailable<sup>26</sup> (CPM 2010; Dancuar et al. 2003; UBA 2010). While FT-naphtha is generally free of sulfur and nitrogen compounds (de Klerk 2007, 2008), fossil-based naphtha is likely to carry higher amounts of these pollutants. The contributions of naphtha steam cracking to acidification and smog may therefore be rather conservative estimates of actual emissions. This is reflected in uncertainties, in particular with regard to smog (Table 3-2).

<sup>&</sup>lt;sup>26</sup> However, yields are based on FT-naphtha steam cracking test runs for the production of light olefins.

Water use is dominated by the gasification process. This is due to direct inputs of feedwater for steam production, cooling, and use in wet scrubbers (syngas cleaning). In addition, water is used in many of the chemicals inputs to gas cleaning such as sodium hydroxide and sulfuric acid (Ecoinvent 2010).

Although not assessed in a quantitative manner, MSW-to-ethylene related impacts to *land occupation* and indirect *land use* change are likely to be minimal when compared to biomass-based routes. Carbon recycling systems can utilize and divert organic waste feedstock that would otherwise go to landfills and incinerators. Carbon stored in chemical end-products and plastics would delay atmospheric GHG emissions. Their reuse by the very same systems (gasifiers may use any type of organic waste including the plastic fraction) reduces the area of land required for carbon uptake to produce virgin biomass.

However, current *carbon recycling efficiencies* from BMSW to chemical feedstocks are only about 19% for the Battelle, 24% for the MTCI, and 33% for the Choren system<sup>27</sup>. With improvements in conversion efficiencies (see sensitivity analysis) this percentage may be increased and more carbon kept within the technosphere.

#### E.2. Marginal Power Mix and Future Energy Scenario

Assuming marginal<sup>28</sup> energy offsets for MSW-ethylene routes and respective BAU scenarios (instead of U.S. average offsets (Table 3-2)) leads to minimal changes in environmental burdens associated with carbon recycling (Appendix: Section E.7.2). This

<sup>&</sup>lt;sup>27</sup> Carbon conversion efficiency = Chemical Feedstock  $[Kg_{carbon out}]/Waste Feedstock [kg_{carbon in}] x 100\%$ .

<sup>&</sup>lt;sup>28</sup> Non-baseload power from coal-fired power plants.

is due to only small amounts of power co-produced by the FTS unit. For BAU scenarios (landfilling and incineration), replacing more carbon-intensive energy at the margins, leads to lower environmental impacts.

Under a *future energy scenario*, using power shares from the IEA Blue Map scenario, carbon recycling may lead to lower impacts with regards to TMR, acidification, and water use (as well as CED for the Choren route) when compared to BAU#2 (incineration) (*Appendix: Section E.7.2*). GWP, CED, and smog potential associated with carbon recycling routes are now only slightly above BAU#2 impacts. This is due to a less carbon and energy intensive power mix being offset. This indicates that while under current conditions carbon recycling may only be environmentally superior to conventional landfilling, *in the long-term (i.e. by 2050 according to the BLUE Map scenario) the shift towards alternative sources of energy could lead to carbon recycling having similar or slightly higher impacts than WtE production via conventional incineration<sup>29</sup>.* 

#### E.3. Sensitivity Analysis

Results of our analysis were found to be sensitive to varying conversion efficiencies and energy inputs to the steam cracker, as well as to the choice of allocation for by-products generated (*Appendix: Section E.7*). Optimizing current system configurations to increase conversion efficiencies (up to 50% from BMSW to FT-liquids) and reduce heat inputs to the steam cracker could lead to a significant reduction in

<sup>&</sup>lt;sup>29</sup> This does not take into account possible future changes in efficiencies for both conventional WM systems as well as carbon recycling routes.

environmental burdens. Considering economic instead of physical allocation may exclude environmental burdens associated with MSW classification (due to the low value of BMSW feedstock reflected in landfilling) therefore resulting in a further reduction in environmental burdens, in particular with regards to GWP and CED.

#### **E.4. Environmental LCC Analysis**

When all parameters are set at their 'base-value', the production costs of 1 kg ethylene at the factory gate are US\$ 2.06 for the Battelle and US\$ 1.85 for the MTCI system, respectively (Jan 2011 US\$)<sup>30</sup>. This compares to currently US\$ 1.17 per kg fossil-based ethylene in the U.S. (CMAI 2011). The lower price for ethylene produced via the MTCI route is due to a slightly higher conversion efficiency which is, however, offset to some extent by less revenue obtained from BMSW tipping fees.

Parameters largely affecting the final ethylene costs are shown in Table 3-3. We vary each of these parameters between the ranges given in the table to determine the sensitivity of the cost figures. Figure 3-4 shows results of the sensitivity analysis for the Battelle system as example.

<sup>&</sup>lt;sup>30</sup> The Choren system has been excluded as cost data are given elsewhere (RENEW 2006).

Table 3-3 Main parameters used for the cost analysis and the ranges for the sensitivity analysis.

Parameter	Unit	Value	Range
Capital cost	\$/kg ethylene	3.11 (varies per concept)	70 - 130 %
BMSW tipping fee	\$/metric ton	30	15 - 45
FT-diesel	\$/kg	0.99	0.6 - 1.4
Propylene	\$/kg	1.57	0.94 - 2.19
Other hydrocarbons	\$/kg	0.96	0.5 - 1.4
Discount rate (r)	%	8	4 - 12
Depreciation period (m)	years	20	10 - 30
Load factor	hours/year	8395	6716 - 8760

Product prices indicate revenue streams. BMSW represents a revenue stream because a tipping fee would be collected by waste managers to landfill or incinerate the waste feedstock.



Figure 3-4 Sensitivity of the production cost of the Battelle carbon recycling system to the parameter variation.

Possible price fluctuations in revenues associated with by-products such as FTdiesel and other hydrocarbons as well as variations in cost model parameters such as capital cost and load factors were found to have a significant effect on the ethylene production cost (Figure 3-4). In this regard, further research in the effects of price volatility and anticipated future oil prices on the production cost of carbon recycling would be of interest. Due to the preliminary nature of equipment cost this factor represents a major uncertainty.

Furthermore, the feedstock price for the carbon recycling technologies was varied from \$-70 to \$+50 per metric ton. Here, the negative prices indicate a tipping fee which is likely to vary depending on the region (e.g. highly populated areas are likely to charge a higher tipping fee than rural areas). On average, conventional biomass costs \$48.5 per dry MT (Valkenburg et al. 2008) and hence we choose a maximum price of \$50/metric ton. Results of the analysis are shown in Figure 3-5.



Figure 3-5 Ethylene production cost versus waste feedstock price for the Battelle and MTCI system. Negative prices indicate revenues from tipping fees.

The figure shows that at current LHV conversion efficiencies (from BMSW to FT-liquids) a tipping fee of roughly \$42 per metric ton, both waste-to-ethylene systems may become economically competitive to fossil-based ethylene production. However, if carbon recycling systems were able to achieve higher conversion efficiencies of 50%, as reported in the literature for biomass-based FTS systems, ethylene production could be economically viable even if no tipping fee is charged. In the figure, flatter slopes at higher conversion efficiencies indicate that less waste feedstock is required per kg of ethylene and hence production cost is less impacted by changes in BMSW cost. At a feedstock price of \$50 (e.g. if traditional biomass is used as feedstock) carbon recycling at higher conversion efficiencies may allow ethylene production at costs of \$3.10 to \$3.55.

## **F.Conclusion**

Our study has shown that carbon recycling, in which organic waste is recycled into naphtha for chemical feedstock production, may have the potential to reduce environmental burdens with regards to GWP, CED, TMR, and acidification when compared to conventional landfilling and fossil-based ethylene production in the U.S. (BAU#1). However, when compared to incineration with energy-recovery, as it is implemented in European countries, carbon recycling based on currently available technologies results in higher system-wide impacts with regards to GWP, CED, TMR, acidification, and smog potential – mainly as a result of large energy offsets associated with waste incineration. However, in the future energy offsets may not be as significant if power comes increasingly from renewable low-carbon sources and hence carbon recycling routes may become increasingly competitive to conventional incineration systems.

While production costs seem not yet competitive to fossil-based ethylene provision, our cost comparison did not account for costs of BMSW treatment in a comparative BAU scenario. Under current conditions, waste-derived ethylene seems to breakeven with its fossil-based counterpart at a tipping fee of roughly \$42 per MT of waste feedstock (and at a tipping fee of \$5 to \$10 at higher conversion efficiencies of 50% (from BMSW to FT-liquids)).

The modeled carbon recycling routes are based on existing technologies available today (though not in an integrated fashion). Uncertainty ranges associated with our results, derived from a combination of existing data, semi-quantitative approaches and sensitivity analysis, are large and may differ from those obtained in more detailed engineering design studies. Absolute results of our study should therefore be used with caution and many uncertainties will remain until the actual operation of similar integrated systems. However, our study was able to indicate the sources of large uncertainties, and identify the subsystems of the life-cycles responsible for the highest environmental burdens and costs. It is hoped that the study will assist interested parties in the further improvement of similar systems and foster interest in carbon recycling as potential future option for waste-to-materials routes.

# APPENDIX <sup>31</sup>

# ENVIRONMENTAL IMPLICATIONS AND COSTS OF MUNICIPAL SOLID WASTE (MSW)-DERIVED ETHYLENE

## A. Abstract

This supplement contains further details on LCA methodology, the assumptions used for the carbon recycling systems along with tables of the foreground life cycle inventories, and contribution analysis for each carbon recycling system and impact category. In addition, detailed figures for the sensitivity analysis are provided.

#### **B.Glossary**

BAU:	Business as Usual
BMSW:	Biodegradable Municipal Solid Waste
DQI:	Data Quality indicator
FT:	Fischer-Tropsch
FTS:	Fischer-Tropsch Synthesis

<sup>&</sup>lt;sup>31</sup> The appendix has been submitted as supporting information with the paper: Nuss, P., Gardner, K.H., and Bringezu, S. (In review). "Environmental Implications and Costs of Municipal Solid Waste (MSW)-Derived Ethylene." *Journal of Industrial Ecology.* 

GHG:	Greenhouse Gas
GWP:	Global Warming Potential
HHV:	Higher Heating Value
LCA:	Life Cycle Assessment
LCC:	Life Cycle Costing
LCI:	Life Cycle Inventory
LFG:	Landfill Gas Recovery
LHV:	Lower Heating Value
MC:	Monte-Carlo analysis
MRF:	Materials Recovery Facility
MSW:	Municipal Solid Waste
RDF:	Refuse Derived Fuel
RENEW:	Renewable Fuels for Advanced Powertrains
TMR:	Total Material Requirement
WM:	Waste Management
WTE:	Waste-to-Energy

# C.Life Cycle Assessment (LCA) Methodology

# C.1. Impact Assessment

The *impact assessment* was carried out using a combination of commonly used life-cycle impact assessment (LCIA) methods, such as TRACI (Bare et al. 2002), the

SimaPro methods library (Goedkoop et al. 2008), MIPS (Lettenmeier et al. 2009; Ritthoff et al. 2002) and ReciPe (Goedkoop et al. 2009), to evaluate global warming potential (GWP), cumulative energy demand (CED, total material requirement (TMR), water use, acidification, and smog.

- <u>GWP</u> accounts for the greenhouse gases (GHGs) emitted to the atmosphere. Characterization factors are based on the Intergovernmental Panel on Climate Change (IPCC). The method contains the climate change factors of IPCC with a timeframe of 100 years (Goedkoop et al. 2008).
- <u>CED</u> refers to the equivalents of primary energy (fossil, nuclear, renewables) consumed. The method to calculate CED is based on the method published by ecoinvent version 1.01 and expanded by PRé Consultants for energy resources available in the SimaPro database (Goedkoop et al. 2008).
- <u>TMR</u> refers to the quantity of resources that has to be extracted or moved in order to obtain e.g. 1 kg of ethylene at the factory gate (Material Input per Service Unit (MIPS)). TMR is based on Material Intensity (MI) factors published (Lettenmeier et al. 2009) and are distinguished into the following categories: abiotic resources, biotic resources, water, air, and earth movements. TMR comprises abiotic and biotic resources plus earth movements and thus accounts for all primary resources required without water and air, mainly<sup>32</sup> solid extractions from the earth crust, both for energy and non-energy purposes, all measured in mass units. The

<sup>&</sup>lt;sup>32</sup> Also including natural oil and gas.

reciprocal (i.e. Service Unit/Material Input) gives insights about the resource productivity, i.e. it can be calculated how much use can be obtained from a certain amount of natural resources (Ritthoff et al. 2002).

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- <u>Water use</u> quantifies the total freshwater consumed by the conversion systems. It is expressed as the volume of water consumed (m3) and based on the characterization factors of the ReciPe (H) v1.04 method (Goedkoop et al. 2009). Water use may be a limitation for the implementation of novel systems in arid regions and developing countries.
- <u>Acidification and Smog</u> were accounted for using the Tool for the Reduction and Assessment of Chemical and other environmental Impacts (TRACI), a standalone computer program developed by the U.S. Environmental Protection Agency (Bare et al. 2002). TRACI uses region-specific characterization factors for North America. Air pollutants, contributing to both impact categories, are released e.g. from flue gas during gasification due to the inhomogeneous nature of the waste feedstock and possible contamination with inert materials and heavy metals.

Although eutrophication and toxicological impacts (e.g. carcinogenics, ecotoxicity, etc,) due to e.g. landfilling of gasifier ash would be of interest, these impact categories were not included due to a lack of data on the detailed waste composition and contaminant concentrations present in the organic waste feedstock, as well as the 'real-life' performance of the gas cleaning systems modeled. In addition, waste composition

and hence contaminant levels are likely to vary depending on factors such as region, consumer habits, and season.

#### C.2. Uncertainty Assessment

Within the life-cycle inventories (LCIs) compiled, the amounts of inputs and outputs are described with single parameters (mean values). Uncertainty of these parameters is due to empirical inaccuracy (imprecise measurements), unrepresentative data (incomplete or outdated measurements), lack of data (no measurements) and model uncertainty (using for instance linear instead of non-linear modeling) (Sonnemann et al. 2003). Reporting single values therefore fails to capture the variability and uncertainty inherent in LCA (Lloyd and Ries 2007).

We use Monte Carlo (MC) simulation to assess the uncertainty associated with the results of the environmental assessment. MC simulation provides a powerful tool to calculate the distribution of the predicted output values reflecting combined parameter uncertainties. Scenario uncertainty includes choices regarding allocation procedures and expected technology trends (Lloyd and Ries 2007) (see next section).

In order to use MC simulation, we must define the likely boundaries of each (input and output) parameter, within which variation may take place. Ideally, this is based on provided data from manufacturers but for pilot- and demonstration facilities such information is frequently not available. The LCI's presented in this paper are based on publically available data and are complemented with data from ecoinvent and U.S. LCI. While the ecoinvent LCA database includes quantitative uncertainty values for

parameters in most of its processes, inventory data for the carbon recycling systems are mostly provided as single values stemming from chemical models (e.g. Aspen Plus®) and pilot-plant runs. We use reported data on the magnitude of uncertainty from the literature (Table 3-4) for the processes including municipal solid waste (MSW) classification, FT-synthesis and FT-naphtha steam cracking.

The probability distributions for the gasifier/gas cleaning unit are based on a *semi-quantitative* uncertainty analysis based on the ecoinvent procedure (Frischknecht and Jungbluth 2007). The approach was chosen due to a lack of sufficient uncertainty data on this process. The procedure uses the pedigree matrix developed by (Weidema and Wesnæs 1996) in which data sources are assessed according to six qualitative characteristics including 'reliability', 'completeness', 'temporal correlation', 'geographic correlation', 'further technological correlation' and 'sample size'. Basic uncertainty factors according to (Frischknecht and Jungbluth 2007) are applied to each inventory parameter. Results of this semi-quantitative approach allow one to estimate the square of the geometric standard deviation.

$$SD_{95} = \sigma_a^2 = exp^{\sqrt{[\ln(U_1)]^2 + [\ln(U_2)]^2 + [\ln(U_3)]^2 + [\ln(U_4)]^2 + [\ln(U_5)]^2 + [\ln(U_6)]^2 + [\ln(U_b)]^2}}$$

with: U<sub>1</sub>: uncertainty factor of reliability

- U<sub>2</sub>: uncertainty factor of completeness
- U<sub>3</sub>: uncertainty factor of temporal correlation
- U<sub>4</sub>: uncertainty factor of geographical correlation

U<sub>5</sub>: uncertainty factor of other technological correlation

U<sub>6</sub>: uncertainty factor of sample size

Ub: basic uncertainty factor

When interpreting the results of this analysis, the reader must be aware of the following:

- The true uncertainty of the MSW-gasification model inputs was not derived from available data, but rather was estimated using a simplified qualitative assessment using data quality indicators and expert judgment regarding the basic uncertainty of these inputs (e.g., it is assumed that CO2 releases in general show much lower uncertainty compared to CO releases (Frischknecht and Jungbluth 2007)).
- Uncertainty analyses were performed using the *ecoinvent* database only, because the U.S. LCI database does not contain uncertainty estimates with its data.
- The estimation of uncertainty for background processes is based on a European context (i.e., uncertainty estimates available in the *ecoinvent* database were not modified to reflect the North American context).
- The error bars represent only the uncertainty in the inventory table. The uncertainty in the characterization scores are not taken into account.

Cause (type of uncertainty)	Magnitude & Data Source	Explanation
Variations in energy requirements for MSW classification	±27.5% (Broder et al. 1993)	Based on energy requirements of five MSW classification plants. Assuming normal distribution.
Uncertainties in inputs and outputs to/from gasifier and gas cleaning	DQI <sup>1</sup> (Frischknecht and Jungbluth 2007; Weidema and Wesnæs 1996)	Semi-quantitative uncertainty analysis based on the ecoinvent procedure (Frischknecht and Jungbluth 2007).
Uncertainties in flue gas emissions to air	DQI <sup>1</sup> (Frischknecht and Jungbluth 2007; Weidema and Wesnæs 1996)	Semi-quantitative uncertainty analysis based on the ecoinvent procedure (Frischknecht and Jungbluth 2007).
Uncertainties in input and output parameters to/from FTS and naphtha steam cracker	±10% (PriceWaterhouseCoopers LLP, Shell International Gas Limited 2003)	Uncertainty in all data values, except efficiencies of combustion processes (steam and gas turbine) for which SD of ±2.5% is assumed. All data values follow normal distribution.

Table 3-4 Sources and magnitude of uncertainty in the data.

<sup>1</sup>DQI: Data Quality Indicators.

## C.3. Sensitivity Analysis

We have carried out a sensitivity analysis under the following headings. The effect of each sensitivity on the LCIA results is assessed.

<u>Physical vs. economic allocation</u>: While for the standard scenario, allocation of environmental burdens is based on physical relationships<sup>33</sup> (mass or energy content) we assess the impact of economic allocation for the following product systems: MSW classification, FTS, and steam cracker. These are based on January 2011 market prices (recalculated using the CPI index (USDOL 2011)).

<sup>&</sup>lt;sup>33</sup> With the exception of small amount of electricity co-produced by FTS, for which system expansion is applied.

- <u>Energy substitution</u>: The substitutional value of energy from waste (biodegradable municipal solid waste (BMSW)) combustion or landfilling with energy recovery equipment, and power co-produced by the carbon recycling schemes affects the system-wide environmental impacts. The importance of energy assumptions has been evaluated by looking at the substitution of non-baseload <sup>34</sup> power in the United States according to (US EPA 2011a).
- Future low carbon energy mix: The impact of using an envisaged future electricity mix consisting of 48% renewables (hydro, wind, and photovoltaic), 24% nuclear, and the remainder coal-fired power plants applying carbon-capture & storage (CCS) to meet the power requirements of the foreground system is assessed. This includes energy substitution by electricity co-produced. The energy mix follows the BLUE Map Scenario of the IEA (IEA 2010). We assume that such an energy mix may be available under a future scenario in the United States.
- Increased conversion efficiency (based on LHV) for the combined gasification/FTS system: Current conversion efficiencies of the modeled MSW-based carbon recycling systems range between 27-31% (from BMSW to FT-liquids). However, technically it is possible to reach much higher conversion efficiencies of 50-60% (e.g. for pressurized gasification/FTS systems). We assess the impact of an increased efficiency for the MSW-based plants of 50% from BMSW to FT-liquids under a future scenario assuming plant optimization.

<sup>&</sup>lt;sup>34</sup> Using non-baseload power might provide a more accurate estimate of the marginal emission rates (which would most likely be offset by electricity produced as co-product e.g. from novel carbon recycling schemes and waste energy systems) than the average U.S. power mix.
• <u>Steam cracker energy inputs</u>: The impact of varying energy inputs to the ethylene production step (steam cracker) is assessed.

#### **D.Inventory Data**

# D.1. Municipal Solid Waste (MSW) Classification

The life-cycle starts with MSW classification during which mixed MSW is separated into different waste fractions including recyclables, compostables, BMSW, and scraps. We assume that electricity is used for meeting all of the energy requirements (33kWh/MT MSW) in MSW classification (Broder et al 1993). The wet tonnes of MSW constitute the mass that must be treated in the classification plant. Energy requirements are *allocated by weight* to the different waste fractions recovered, i.e. BMSW fluff (37%), recyclables (24%), compostable waste (9%), and scraps (30%) (EPA 2009b). The input of BMSW to the gasification plants is calculated based on the average energy content of the waste fluff after classification (11.588 MJ per kg wet BMSW fluff). The waste composition is based on U.S. average data for 2008 (EPA 2009b), while typical energy contents are taken from (Tchobanoglous et al 1993). This analysis assumes that all BMSW destined for landfills and incinerators can be separated from the remaining waste either at the source or during the classification process (during which recyclable materials are recovered).

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#### **D.2.** Gasification and Fischer-Tropsch Synthesis (FTS)

Commercial scale MSW-based Fischer-Tropsch synthesis (FTS) plants do not yet exist, but preliminary mass and energy balances on syngas generation from MSW, BMSW and refuse derived fuel (RDF) (Jones et al 2009; Juniper Consultancy Services 2001; Niessen et al 1996; Paisley et al. 1989) as well as inventory data for FT syncrude production from syngas (Bechtel 1998; Choi et al. 1997; Jones et al 2009; Jungbluth et al. 2007; Marano and Ciferno 2001; Van Bibber et al. 2007) are available from the open literature.

LCI data for three gasifiers (Battelle, MTCI, and Choren) capable of converting biowaste into syngas are selected and compiled from (Niessen et al 1996; Paisley et al. 1989; Juniper Consultancy Services 2001; Jungbluth et al 2007; Khoo 2009). The transport distance from the MSW classification plant to the gasification plant is taken as 50 km via a combination truck using an U.S. average fuel mix (NREL 2008). Various reports are available describing the in-depth technical details of these technologies (see e.g. (Belgiorno et al. 2003; Juniper Consultancy Services 2001; Klein 2002; Malkow 2004)) and therefore they are not explained in detail. The cleaning process envisaged after gasification is low temperature wet gas cleaning (Jungbluth et al. 2007).

Table 3-5 and Table 3-6 show the compiled life cycle inventories (LCIs) for the Battelle and MTCI gasifiers. Inventory data tables for the Choren system (gasification + FTS) are available from (Jungbluth et al. 2007).

Category	Parameter	Unit	Amount	SD95	DQI	Comments	Source	Biog. C- content [kg]	Biog. C- balance [kg
INPUTS:									1.6
Technos	BMSW fluff (wet)	kg	2.58E+00	1.640	2,4,5,1,3,5	Based on energy content	(Niessen et al. 1996)	2.72E-01	7.03E-01
Technos	Transport	kgkm	1.29E+02	2.095	4,5,na,na,na,na	50 km transport distance	Own assumption	-	-
Technos.	Electricity	kWh	1.14E-01	1.406	2,3,4,5,3,5	Includes energy for rotary		-	-
						dryers, crushing, air compression, pumping.	(Jungbluth et al. 2007)		
Technos.	Sand	kg	5.06E-02	5.435	2,3,4,5,3,5	Circulating sand matrix	(Jungbluth et al. 2007)	-	•
Technos.	Dolomite	kg	4.09E-02	1.406	2,3,4,5,3,5	Gas cleaning	(Jungbluth et al. 2007)	-	-
Technos	H2SO4	kg	2.35E-03	1.467	2,5,4,5,3,5	Based on typical raw syngas impurities (wood)	(Belgiorno et al. 2003)	-	-
Technos.	NaOH	kg	3.13E-03	1.467	2,5,4,5,3,5	Based on typical raw syngas impurities (wood)	(Belgiorno et al. 2003)	-	•
Technos.	Feedwater in	kg	5.15E-01	1.640	2,4,5,1,3,5	• • •	(Niessen et al. 1996)	-	-
Technos	Infrastructure	р	1.05E-09	1.406	2,3,4,5,3,5	Based on Ecoinvent infrastructure process	(Ecoinvent 2010)	-	-
OUTPUTS:								•	-
FU	Syngas (dry)	kg	1.00E+00				(Niessen et al. 1996)	3.58E-01	3.58E-01
Solid Waste	Ash	kg	2.76E-01	1.640	2,4,5,1,3,5	Dry, to landfill	(Niessen et al. 1996)	2.70E-01	7.45E-02
Air	Biog. CO2 (flue gas)	kg	9.91E-01	1.640	2,4,5,1,3,5		(Niessen et al. 1996)	2.73E-01	2.70E-01
WW	Wastewater to discharge	kg	4.48E-01	1.640	2,4,5,1,3,5	WW composition based on data from Ecoinvent	(Niessen et al. 1996)	-	-
Air	NOx	kg	4.53E-04	1.941	2,5,5,5,3,5		(Khoo 2009)	-	-
Air	SO2	kg	1.05E-05	1.694	2,5,5,5,3,5		(Khoo 2009)	-	•
Air	CO	kg	1.16E-04	5.435	2,5,5,5,3,5		(Khoo 2009)	4.29E-01	4.97E-05
Air	HCI	kg	1.86E-05	1.941	2,5,5,5,3,5		(Khoo 2009)	•	-
Air	HF	kg	1.16E-07	1.941	2,5,5,5,3,5	Based on average data for	(Khoo 2009)	-	-
Air	Dust	kg	6.97E-06	1.941	2,5,5,5,3,5	BMSW gasification	(Khoo 2009)	•	-
Air	Cđ	kg	8.01E-09	5.435	2,5,5,5,3,5		(Khoo 2009)	-	-
Air	Hg	kg	1.16E-07	5.435	2,5,5,5,3,5		(Khoo 2009)	-	-
Air	Pb	kg	1.16E-07	5.435	2,5,5,5,3,5		(Khoo 2009)	-	-
Air	Dioxin	kg	5.81E-14	3.379	2,5,5,5,3,5		(Khoo 2009)	-	-
C(in), pre-pro	duct (calculated)	)							7.03E-01
C(out), emiss	ions								7.46E-02
C(out), proce	ss-output								3.58E-01 2.70E-01

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FU = Functional unit. DQI: Data Quality Indicators. SD95: geometric standard deviation (95% percentile).

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Category	Parameter	Unit	Value	SD95	DQI	Comment	Source	Biog. C- content [kg]	Biog. C- balance [kg]
INPUTS:									
Technos.	BMSW fluff	kg	2.24E+00	1.640	2,4,5,1,3,5	Based on energy content	Niessen et al. 1996)	2.47E-01	5.53E-01
Technos.	Transport	kgkm	1.12E+02	2.095	4,5,na,na,na,na	50 km transport distance Includes energy for rotary	own assumption	-	
Technos.	Electricity	kWh	9.90E-02	1.406	2,3,4,5,3,5	dryers, crushing, air compression, pumping.	(Jungbluth et al. 2007)	•	•
Technos	Limestone	kg	4.38E-02	1.406	2,3,4,5,3,5	Limestone matrix	(Jungbluth et al. 2007)	-	•
Technos	Dolomite	kg	3.54E-02	1.406	2,3,4,5,3,5	Gas cleaning	(Jungbluth et al. 2007)	-	-
Technos	H2SO4	kg	2.35E-03	1.467	2,5,4,5,3,5	Based on typical raw syngas impurities (wood)	(Belgiorno et al. 2003)	-	-
Technos.	NaOH	kg	3.13E-03	1.467	2,5,4,5,3,5	Based on typical raw syngas impurities (wood)	(Belgiomo et al. 2003)	-	-
Technos.	Feedwater in	kg	5.93E-01	1.640	2,4,5,1,3,5		Niessen et al. 1996)	•	-
Technos	Infrastructure	р	1.05E-09	1.406	2,3,4,5,3,5	Based on ecoinvent infrastructure process	(Ecoinvent 2010)	•	•
OUTPUTS:									•
FU	Syngas (dry)	kg	1.00E+00			FU	Niessen et al. 1996)	3.58E-01	3.58E-01
Solid Waste	Ash	kg	2.03E-01	1.640	2,4,5,1,3,5	Dry, to landfill	Niessen et al. 1996)	3.13E-02	6.36E-03
Air	Biog. CO2 (flue gas)	kg	6.90E-01	1.640	2,4,5,1,3,5		Niessen et al. 1996)	2.73E-01	1.88E-01
WW	Wastewater to discharge	kg	3.28E-01	1.640	2,4,5,1,3,5	based on treatment plant class 2 data from Ecoinvent	Niessen et al. 1996)	-	•
Air	NOx	kg	4.530E-04	1.941	2,5,5,5,3,5		(Khoo 2009)	-	-
Air	SO2	kg	1.050E-05	1.694	2,5,5,5,3,5		(Khoo 2009)	•	•
Air	CO	kg	1.160E-04	5.435	2,5,5,5,3,5		(Khoo 2009)	4.29E-01	4.97E-05
Air	HC1	kg	1.860E-05	1.941	2,5,5,5,3,5		(Khoo 2009)	-	-
Air	HF	kg	1.160E-07	1.941	2,5,5,5,3,5	Based on average data for	(Khoo 2009)	-	-
Air	Dust	kg	6.970E-06	1.941	2,5,5,5,3,5	BMSW gasification	(Khoo 2009)	-	-
Air	Cđ	kg	8.010E-09	5.435	2,5,5,5,3,5		(Khoo 2009)	-	-
Air	Hg	kg	1.160E-07	5.435	2,5,5,5,3,5		(Khoo 2009)	•	•
Air	Pb	kg	1.160E-07	5.435	2,5,5,5,3,5		(Khoo 2009)	•	•
Air	Dioxin	kg	5.810E-14	3.379	2,5,5,5,3,5		(Khoo 2009)	•	•
C(in), pre-pro	duct (calculated)								5.53E-01
C(out), emissi	ions								6.41E-03
C(out), proces	ss-output								3.58E-01
C(out), emissi	ions, CO2								1 88E-01

# Table 3-6 Unit process data for the process: 'clean synthetic gas from RDF, MTCI ThermoChem gasification unit'.

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FU = Functional unit. DQI: Data Quality Indicators. SD95: geometric standard deviation (95% percentile).

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The FTS is based on energy and material balances given for two different FT systems (Van Bibber et al. 2007; Jungbluth, Frischknecht, et al. 2007). On the one hand, clean syngas generated by the BHTGS and MTCI gasification units is fed into a *slurry-bed, iron-based catalyst FT-reactor* based on a model developed from public information (Van Bibber et al. 2007). The FT-model used in their study is based on data originally published by Bechtel/Amoco in 1993<sup>35</sup>. On the other hand, syngas generated from the Choren Carbo-V process is converted into FT syncrude using a *cobalt catalyst in a tubular-fixed-bed reactor (TFBR)*. This process is based on aggregated inventory data (Jungbluth, Frischknecht, et al. 2007; RENEW 2006).

The energy and material balance for the process is based on the concept 2 plant from (Van Bibber et al. 2007) (excluding gasification). Oxygen is assumed to have been produced by the fractional distillation of air in a process that uses electricity.  $CO_2$ emissions from fuel gas combustion were derived from a carbon balance around the FT plant. All other air emissions associated with fuel gas consumption were taken from Table 24 in (Marano and Ciferno 2001) and recalculated per kg of FT-liquid produced. The composition of the FT-catalyst is based on (Jungbluth, Frischknecht, et al. 2007). The compiled LCI is shown in Table 3-7.

<sup>&</sup>lt;sup>35</sup>Baseline Design/Economics for Advanced Fischer-Tropsch Technology, DOE Contract No. DE-AC22-91PC90027, Topical Report Volume 1, Process Design – Illinois No. 6 Coal Case with Conventional Refining, October, 1994.

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Category	Parameter	Unit	Amount	Comments	Source	Biog C- content	Biog C- balance
INPUT						[*6]	[*6]
Technos	Clean Syngas	kg	6.03E+00	H2/CO = 0.81	(Van Bibber et al. 2007)	3.58E-01	2.16E+00
Technos	Water to FTS	kg	9.11E-02		(Van Bibber et al. 2007)	-	-
Technos	Steam to ATR	kg	2.14E-01		(Van Bibber et al. 2007)	-	-
Technos	Oxygen to ATR	kg	5.15E-02	Fractional distillation of air	(Van Bibber et al. 2007)	-	•
Technos	Syngas Compressor	kWh	1.22E+00		(Nexant 2006)	•	•
Technos	FT processes	kWh	8.90E-02		(Van Bibber et al. 2007)	-	-
Technos.	Transformer Losses	kWh	7.05E-03		(Van Bibber et al. 2007)	-	-
Technos	FT catalyst	kg	1.42E-04	Typical value from RENEW project	(Jungbluth et al. 2007)	-	•
Technos	FT fuel-synthesis plant	p	2.47E-10	Conversion plant from RENEW	(Jungbluth et al. 2007)	•	-
OUTPUT				p j			
FU	FT-Naphtha	kg	4.13E-01		(Van Bibber et al. 2007)	8.46E-01	3.50E-01
BP	FT-Distillate	kg	5.87E-01		(Van Bibber et al. 2007)	8.46E-01	4.96E-01
Air	CO2 (from CO2- removal)	kg	4.28E+00		(Van Bibber et al. 2007)	2.73E-01	1.17E+00
Air	CO2 (in flue gas)	kg	5.50E-01	Calculated to close carbon balance		2.73E-01	1.50E-01
BP	Electricity output (Total)	kWh	2.24E+00		(Van Bibber et al. 2007)	-	-
				Total output - Total use = Net	, , , , , , , , , , , , , , , , , , ,		
BP	Electricity output (Net)	kWh	8.58E-01	output; Used internally with excess sold to grid	(Van Bibber et al. 2007)	-	-
Air	CH₄	kg	2.34E-05	Fuel gas consumed	(Marano and Ciferno 2001)	7.50E-01	1.76E-05
Air	N <sub>2</sub> O	kġ	3.60E-05	Fuel gas consumed	(Marano and Ciferno 2001)	•	•
Air	NOx	kg	1.15E-03	Fuel gas consumed	(Marano and Ciferno 2001)	-	-
Air	CO	kg	2.77E-04	Fuel gas consumed	(Marano and Ciferno 2001)	4.29E-01	1.19E-04
Air	VOC	kg	4.86E-05	Fuel gas consumed	(Marano and Ciferno 2001)	N.A.	-
Air	PM	Kg	2.45E-05	Fuel gas consumed	(Marano and Ciferno 2001)	-	•
C(in), pre-prod	uct (calculated)						2.16E+00
C(out), emissio	ns						-
C(out), process	-output						8.46E-01
C(out), emissio	ns, CO2						1.32E+00

Table 3-7 Unit process raw	data for the generation	of 1 kg FT liquids	(from syngas)	at the factory gate
Table 5*7 Onit process raw	uata for the generation	UT I KE I I IIQUIUS	(nom syngas)	at the factory gate.

FU = Functional unit; BP = Byproduct; ATR: Autothermal Reforming. The inventory is based on ASPEN modeling results (Van Bibber et al. 2007). Air emissions come from (Marano and Ciferno 2001).

#### **D.3. Carbon Capture and Storage (CCS) – and Reuse (CCU)**

FT systems provide the opportunity for carbon capture and storage (CCS) or reuse (CCU), during which CO<sub>2</sub> is extracted from the product gas stream in the absorption tower using an amine acid gas removal process. CO<sub>2</sub> vented from the absorption tower is compressed and sent to sequestration (or alternatively for use as chemical feedstock). Following (Larson and Tingjin 2003), CO<sub>2</sub> drying and compression requires about 97.8 kWh per tonne of CO<sub>2</sub> released. Due to the large uncertainties associated with subsequent carbon sequestration (e.g. storage underground), we do not account for the environmental impacts associated with CCS. In general, operating CCS equipment would demand additional power. While a small fraction of this could be supplied internally from power produced as byproduct during FTS, the bulk would come from the U.S. power grid<sup>36</sup>. A recent investigation on CCS applied to FTS systems utilizing natural gas and coal in the United States can be found in (Jaramillo et al. 2008). With more knowledge on the environmental implications of CCS this could be included as a possible option in the LCA model in the future.

## **D.4.** Comparison of FT Plant to Literature

When compared to performance data from the literature, the LHV conversion efficiency<sup>37</sup> of the combined gasification/FTS systems is with 27% (Battelle) and 31% (MTCI) slightly below the range of 33-40% found for atmospheric gasification systems

<sup>&</sup>lt;sup>36</sup> In a commercial FTS plant applying CCS, the product distribution (FT-liquids vs. power) could be optimized so that the bulk of the energy for CCS could be supplied internally (assuming a carbon-intensive power grid mix).

<sup>&</sup>lt;sup>37</sup> Here calculated as: (LHV<sub>FT-liquids</sub> + LHV<sub>Excess Electricity</sub>) / (LHV<sub>BMSW input</sub>).

(Tijmensen et al. 2002) utilizing conventional biomass feedstock (Table 3-8). The Choren system scores with 53% (LHV eff%) slightly above the range of 42-50% found for pressurized gasification systems utilizing biomass feed (Tijmensen et al. 2002). The lower overall efficiency of the waste conversion systems is due to a number of reasons:

- Pressurized concepts have higher overall efficiencies than atmospheric designs (such as the Battelle and MTCI configurations), mainly due to high electricity consumption associated with syngas compression prior to FTS (Tijmensen et al. 2002) (see respective LCI tables for the energy requirements of compression).
- The MSW gasifiers have significantly lower thermal efficiencies when compared to biomass gasifiers described in the literature. For example, biomass gasifiers assessed in (van Vliet et al. 2009) show cold gas efficiencies (CGE) of 77% or above (based on LHV) while biomass gasification systems investigated in (Tijmensen et al. 2002) have CGEs of 80-89%. In contrast to this, the MSW gasifiers in this study show a CGE of only 69-72%).
- The mass and energy balance of the MSW gasifiers is based on publically available data. These data were derived from pilot tests and modeling runs in the 1990s and may not represent state-of-the art performance of modern MSW gasifiers.

We investigate the impact of varying conversion efficiencies on the overall environmental burdens as part of the *sensitivity analysis*.

Concept All numbers in MJ	Battelle CFB gasifier + Slurry bed Fe-based FTS	MTCI BFB gasifier + Slurry bed Fe-based FTS	Choren Two- stage EFG gasifier (Carbo-V process)	Battelle IH, airblown, atmospheric, Full conversion FTS, Reformer used	Battelle IH, airblown, atmospheric, Full conversion FTS	500 MW plant, based on model	Indirectly heated gasifier + Full recycle FTS	Indirectly heated gasifier + Once- through FTS
Source	(Niessen et al 1996; Van Bibber et al. 2007)	(Niessen et al 1996; Van Bibber et al. 2007)	(Jungbluth et al. 2007)	(Tijmensen et al. 2002)	(Tijmensen et al. 2002)	(Hofbauer and Schönberger 2008)	(Larson and Jin 1999)	(Larson and Jin 1999)
RDF	100.00	100.00	-	-	-	-	-	-
Biomass			100.00	100.00	100.00	100.00	100.00	100.00
FT- distillate	15.59	18.00	44.09	25.32	13.65	36.59	36.90	19.78
FT- naphtha	10.90	12.59	8.97	8.44	4.55	23.39	12.50	6.70
FT-liquids (total)	26.49	30.59	53.06	33.76	18.20	59.98	49.40	26.48
Power	0.37	0.66	-	11.28	17.71		-	17.20
eff. % (LHV)	26.85	31.25	53.06	45.04	35.91	59.98	49.40 <sup>1</sup>	43.68 <sup>1</sup>
Notes:	Based on LCA model used in this study	Based on LCA model used in this study	Assuming 17% naphtha/83% distillate output	Assuming 25% naphtha/75% distillate output (Kerosene mode)	Assuming 25% naphtha/75% distillate output (Kerosene mode)	Assuming 39% naphtha/61% distillate output		

Table 3-8 LHV energy efficiencies for the gasification/FTS systems under investigation (left three columns) and compared to literature data.

All numbers are in MJ.<sup>1</sup>Higher Heating Value (HHV) eff%. CFB: Circulating Fluidized Bed; BFB: Bubbling Fluidized Bed; EFG: Entrained Flow Gasifier; IH: Indirectly Heated. Distillate includes kerosene, diesel and waxes.

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#### **D.5. Steam Cracking for Light Olefins Production**

Ethylene production from FT naphtha is assumed to take place via conventional steam cracking. FT naphtha, for use as a steam cracker feedstock, was found to be extremely well suited for the production of lower olefins (Dancuar et al. 2003). This was attributed to the high feed paraffins content and almost total absence of aromatics. Accordingly, this study assumes the use of conventional naphtha steam cracking (Dampfpyrolyse).

We use a combination of data from the CPM database (CPM 2010) and the ProBas database (UBA 2010) to model the resource and energy in- and outputs from the FT naphtha steam cracking process. The yields are based on data from real FT-naphtha steam cracking by (Dancuar et al. 2003). During the cracking process a number of chemical substances, including ethylene, propylene, 1,3-butadiene and C4+ liquids, paraffins etc., are generated. The product distribution depends on the chemical nature of the naphtha feedstock and was found to be very similar to typical petroleum-based light naphtha. The product yields of the FT-naphtha steam cracking tests carried out by (Dancuar et al. 2003) are shown in the following table. FT-naphtha steam cracking yields can also be found in (PriceWaterhouseCoopers LLP, Shell International Gas Limited 2003).

In our study, methane and hydrogen (Heizgas) produced are not accounted for as product outputs as both are assumed to be used internally for heat production (following (UBA 2010)). Allocation percentages are: ethylene (48.3 %), propylene (17.6 %), C5+ liquids (16.9 %), and others (17.2 %). Since information on emissions to air and water is

limited in ProBas, these are complemented with data from CPM on typical naphtha steam

cracking. The following table shows the compiled life cycle inventory.

Category	Parameter	Unit	Amount	Comments	Source	Biog. C- content [kg]	Biog. C- balance [kg]
INPUT						1 07	1 8
Technos.	Naphtha	kg	2.50E+00		(Dancuar et al. 2003)	8.46E-01	2.12E+00
Technos.	Thermal Energy	MJ	2.59E+01	From natural gas	(UBA 2010)	-	-
Technos. OUTPUT	Water	kg	2.35E+00		(UBA 2010)	-	-
FU	Ethylene	kg	1.00E+00		(Dancuar et al. 2003)	8.57E-01	8.57E-01
BP	Propylene	kg	3.65E-01		(Dancuar et al. 2003)	8.57E-01	3.13E-01
BP	1,3-Butadiene	kg	1.18E-01		(Dancuar et al. 2003)	9.23E-01	1.08E-01
BP	C5+ Liquids	kg	3.50E-01		(Dancuar et al. 2003)	8.27E-01	2.89E-01
BP	Other olefins	kg	8.25E-02		(Dancuar et al. 2003)	8.57E-01	7.07E-02
BP	Other Paraffins	kg	1.00E-01		(Dancuar et al. 2003)	8.27E-01	8.27E-02
BP	Alkylenes	kg	6.00E-02		(Dancuar et al. 2003)	8.27E-01	4.96E-02
Air	Benzene	kg	2.82E-04		(UBA 2010)	9.23E-01	2.60E-04
Air	CH4	kg	1.33E-03		(CPM 2010)	7.50E-01	1.00E-03
Air	CO	kg	1.67E-04		(CPM 2010)	4.29E-01	7.14E-05
				to close			
Air	CO2	kg	1.26E+00	carbon		2.73E-01	3.43E-01
				balance			
Air	HC	kg	8.00E-08		(CPM 2010)	9.23E-01	7.38E-08
Air	N2O	kg	9.33E-06		(CPM 2010)	-	-
Air	NMVOC	kg	1.40E-05		(CPM 2010)	N.A.	-
Air	NOx	kg	1.33E-03		(CPM 2010)	-	-
Air	SO2	kg	2.33E-03		(CPM 2010)	-	-
Air	VOC	kg	8.33E-04		(CPM 2010)	N.A.	-
Waste	Prod. Waste	kg	5.22E-03	Chemical refinery waste	(UBA 2010)	-	-

Table 3-9 Life cycle inventory (before allocation) for the generation of 1 kg ethylene from FT naphtha.

Data was compiled from (CPM 2010; UBA 2010). The product yields are based on (Dancuar et al. 2003).

According to (Ren et al. 2006), typical specific energy consumptions<sup>38</sup> (SECs) for naphtha steam crackers are between 26-31 MJ/kg ethylene<sup>39</sup> produced (or 20-40 MJ/kg

<sup>&</sup>lt;sup>38</sup> SEC is the sum of fuel, steam and electricity in primary terms used for reactions and all subsequent processes.

<sup>&</sup>lt;sup>39</sup> MJ/kg ethylene means that all energy use is allocated to ethylene not considering useful by-products generated.

ethylene (maximum)). The heat input of 25.9 MJ/ kg ethylene (before allocation) equals a cumulative energy demand (CED) (Ecoinvent 2010) of 42.3 MJ-eq, indicating that the inventory compiled might represent a rather conservative estimate (likely due to older data sources utilized). Hence, varying energy inputs to the ethylene production step are further investigated in the *sensitivity analysis*.

## **D.6. Off-Gas Emission Profiles**

Data on the emission profiles from waste gasification facilities and integrated FTS refineries are rarely available. An integrated facility, as described in prior chapters, would burn tail gases, synthesis gases, and light hydrocarbon fractions for the generation of power and heat. On-site air pollutants are released during syngas cleanup, FTS, and steam cracking. These will be influenced mainly by filter technologies (as a result of economic considerations and legal standards). With the present state of knowledge being limited, emission profiles are based on publically available data on MSW gasification (Khoo 2009), catalytic conversion via FTS (Jungbluth, Frischknecht, et al. 2007; Marano and Ciferno 2001), and conventional naphtha steam cracking (CPM 2010; Dancuar et al. 2003; UBA 2010). These are likely to vary depending on the type of waste feedstock utilized and even with time as consumer habits and legal requirements change. Emissions from steam cracking are based on conventional fossil-based naphtha which is likely to carry significantly higher amounts of SO<sub>x</sub> and other air pollutants than FT-naphtha (de Klerk 2007, 2008) contributing to system-wide acidification and smog impacts.

Therefore, emissions from the steam cracking process may represent a rather conservative estimate.

### **D.7. Energy System**

The choice of the energy system in LCA can have a major influence on the results. This is in particular true for industrial processes that can produce energy as byproduct which can offset conventional fossil-based energy. We use data from (Ecoinvent 2010) for the current U.S. average power grid (0.775 kg  $CO_2$ -eq/kWh) and power shares from the IEA BLUE Map Scenario (IEA 2010), according to which a future low-carbon energy mix (0.206 kg  $CO_2$ -eq/kWh) would consist of 48% renewables, 24% nuclear, and 17% coal power using CCS (Table 3-10), to model energy in- and outputs (offsets) as part of the foreground system. The goal of including the BLUE Map mix in our analysis is not to exactly estimate its potential future impacts, but rather use it as a widely accepted proxy to investigate how an envisaged low-carbon energy mix will impact results of our LCA model.

Table 3-10 Future low-carbon energy mix using energy shares from the IEA BLUE Map scenario (IEA 2010).

Fuel Type	kWh	% of energy mix
Hydro	0.1753	16
Photovoltaic	0.1753	16
Wind	0.1753	16
Nuclear	0.2630	24
Coal (CCS) <sup>1</sup>	0.1863	17
Coal	0.1205	11
TOTAL	1.0958 <sup>2</sup>	100

The scenario assumes that an energy mix using renewables, nuclear power, and plants equipped with CCS would be available in the year 2050. We use data from Ecoinvent to model power inputs from renewables (hydro, PV, wind), nuclear and traditional coal power. <sup>1</sup>Coal-fired power plants equipped with CCS can reduce carbon emissions to 0.234 kg CO<sub>2</sub>-eq per kWh of electricity produced, but may also result in environmental trade-offs e.g. in increase in human toxicity, ozone layer depletion and fresh water ecotoxicity (Koornneef et al. 2008). For our model, we use a *simplified approach* accounting only for GHG reductions of CCS but excluding differences in all other impact categories compared to current coal-fired power plants (LCI data taken from Ecoinvent). <sup>2</sup>Accounts for transmission losses of 9.58%.

Carbon recycling systems described in previous chapters and conventional waste management (WM) schemes can generate electricity (and steam) as beneficial byproducts substituting conventional power. Since the current energy system relies to a large extent on fossil fuels including nuclear power, system-wide GWP is strongly affected by the substitutional value of energy generated from BMSW. However, instead of U.S. average power, waste management models such as EPA WARM (EPA 2010a) assume that energy generated by Waste-to-Energy (WtE) systems would rather offset electricity generation at a marginal, fossil-fuel-only<sup>40</sup> environmental burdens rate. As a result, the model assumes that marginal demands are met by fossil sources, leading to higher GHG emissions per kWh than U.S. average power. In order to investigate the

<sup>&</sup>lt;sup>40</sup> For simplicity, WARM currently uses the national weighted average of fossil-fuel plants as a proxy for the fuels displaced at the margin when electricity is displaced by electricity from WtE systems (EPA 2010a). The fossil-fuel mix does not include nuclear power. According to the EPA WARM model, nonfossil sources are expected to meet baseload requirements because of the financial incentive to operate these at capacity.

stability of our model results with regards to energy assumptions we include substitution of marginal power offsets in the sensitivity analysis. The marginal power mix (1.31 kg  $CO_2$ -eq/kWh) comes from coal-fired power plants (NREL 2008), assumed to be more likely to respond to incremental changes in electricity supply and demand.

In summary, the modeled energy scenarios include:

- Substitution of 100% marginal (non-baseload power) under current conditions following assumptions of the EPA WARM model (EPA 2010a). Data on coalfired power plants in the United States comes from U.S. LCI (NREL 2008).
- Substitution of electricity from the *IEA BLUE Map scenario* by waste-derived energy using inventory data from (Ecoinvent 2010; Koornneef et al. 2008) and power grid shares from (IEA 2010).

Finally, the following table compares the life-cycle wide environmental burdens of the current, marginal, and future energy mix.

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Impact category	Unit	Average power mix, USA, year 2011	Marginal power mix, USA, year 2011	Future power mix, IEA BLUE Map scenario, year 2050
GWP 100a	kg CO2 eq	7.754E-01	1.310E+00	2.059E-01
CED	MJ eq	1.284E+01	1.405E+01	1.044E+01
TMR	kg	1.510E+00	2.898E+00	9.596E-01
Acidificati on	H+ moles eq	2.747E-01	4.312E-01	1.264E-01
Smog	g NOx eq	1.486E-03	2.818E-03	8.336E-04
Water use	m3	2.149E-03	2.834E-03	1.755E-03

Table 3-11 System-wide environmental burdens associated with the production of 1 kWh of electricity now and under a future scenario.

<sup>1</sup>This is slightly higher than the 67 g/kWh according to (IEA 2010) and may be due to the fact that we use currently existing LCI data (from Ecoinvent) to model the future mix (i.e. we do not account for possible efficiency gains in future power production plants).

# **E.Life Cycle Impact Assessment Results**

## E.1. Global Warming Potential (GWP)

Impacts to GWP (Figure 3-6) are mainly due to energy inputs to the steam cracker (1.35 kg  $CO_2$ -eq), converting FT-naphtha into light olefins such as ethylene and propylene. This is followed by energy requirements for waste classification (Battelle: 0.478, MTCI: 0.415, Choren: 0.240 kg  $CO_2$ -eq) separating BMSW from the mixed MSW stream. The difference in impacts is due to larger amounts of waste feedstock gasified in the Battelle and MTCI systems, which both have a lower LHV (lower heating value) conversion efficiency (Table 3-8) when compared to the Choren system. Gasification leads to 0.159-0.181 kg  $CO_2$ -eq for the MTCI and Battelle system, respectively. Aggregated data for the Choren plant indicates a GWP of 0.397 kg  $CO_2$ -eq associated with the conversion step from biomass to FT-naphtha. During the FTS step, electricity is co-produced, most of which is used internally. However, both the MTCI and Battelle conversion systems generate a small amount of excess electricity (0.26 and 0.17 kWh/kg

FT-liquids) which is assumed to offset conventional electricity from the U.S. national grid.



Figure 3-6 GWP (kg  $CO_2$ -eq) of producing 1 kg ethylene from MSW compared to the production of an equivalent amount of fossil-based ethylene and treatment of the BMSW via conventional landfilling (BAU #1) or incineration with energy recovery (BAU#2). Electricity is assumed to offset energy from the U.S. power grid. Error bars show the 95% confidence interval. Biogenic carbon storage in the final ethylene product is not accounted for in the GWP impact category.

Figure 3-7 shows a Sankey diagram for the MTCI process showing how unit

processes are linked to each other and their relative contribution to total GWP.



Figure 3-7 Sankey diagram showing the contributions of various MTCI unit processes to GWP. The functional unit is 1 kg ethylene at the factory gate. The width of the arrows is shown proportional to contributions to GWP. Green arrows indicate avoided burdens. A cut-off of 4.4% was applied, meaning that only unit processes contributing more than the cut-off are displayed.

GWP associated with waste-derived ethylene is with 1.77 to 1.97 kg CO<sub>2</sub>-eq significantly smaller than for all  $BAU#1^{41}$  landfilling scenarios (2.91 to 4.88 kg CO<sub>2</sub>-eq). Besides fossil-based ethylene production (1.26 kg CO<sub>2</sub>-eq) and MSW sorting (0.24 to 0.478 kg CO<sub>2</sub>-eq) as part of the BAU scenarios, BMSW landfilling results in the bulk of GHG emissions (1.58 to 3.15 kg CO<sub>2</sub>-eq). This is due to anaerobic digestion of BMSW within the landfill over time, some of which is not captured by LFG recovery equipment. As discussed in (Nuss et al. 2012), the choice of the comparative system (i.e. landfills U.S. national average vs. landfills with or without any type of LFG recovery equipment) can drastically alter results since methane emissions depend on the amount of LFG recovered for flaring or energy purposes. However, for this assessment EPA national average data on LFG emissions is used, according to which about 0.166 kg CO<sub>2</sub>-eq are emitted per kg of BMSW landfilled (EPA 2010a). With 3.08 kg CO<sub>2</sub>-eq, GHG emissions are lowest for the BAU#1 case in which BMSW, otherwise used as feedstock in the Choren plant, is landfilled and ethylene is produced from fossil-fuels. This is due to the fact that most of the BMSW is used to produce distillate (for fuels) and less for naphtha (ethylene), and thus a major part of landfill emissions in the BAU#1 scenario are allocated to the distillate. Results indicate that under current conditions carbon recycling may have the potential to lead to an overall reduction in GWP when compared to current landfills and ethylene production systems in the U.S. (BAU landfilling).

<sup>&</sup>lt;sup>41</sup> The BAU scenarios consist of the sum of environmental burdens associated with 1.) conventional waste treatment (i.e. BMSW landfilling or incineration), 2.) MSW classification, and 3.) fossil-based ethylene production in the United States (from natural gas).

On the other hand, if compared to a <u>BAU#2 case (BMSW incineration for energy</u> <u>recovery</u>), carbon recycling leads to a much higher GWP. With a combustion efficiency of 16.3% (EPA 2010a) the electricity generated by BMSW incinerators in the BAU scenarios has the potential to offset a large share of power from the U.S. electricity grid, therefore reducing the overall environmental impacts (avoided burdens). The offset is largest for the Battelle and MTCI BAU#2 cases both requiring larger amounts of BMSW than the Choren plant (ironically, the less efficient technologies). However, the analysis assumes that energy generated replaces current carbon-intensive electricity from the U.S. power grid (0.775 kg CO<sub>2</sub>-eq/kWh). The choice of a less carbon-intensive energy mix using the BLUE Map scenario assumptions is hence investigated in later sections (sensitivity analysis).

**Carbon sequestration:** The WARM landfill model (EPA 2010a) makes assumptions impacting results of the GWP comparison. These include: 1.) CH4 is counted as anthropogenic GHG while  $CO_2$  is excluded because BMSW degradation under natural conditions would only yield biogenic  $CO_2$ ; 2.) Carbon in un-decomposed waste fractions is stored in the landfill and credits are provided for this (anthropogenic carbon sink); 3.) Energy generated via LFG is assumed to offset power from the U.S. average power grid.

In our basic LCA model we did not account for the fact that by generating a chemical product output (1 kg ethylene) carbon present in the BMSW feedstock is in fact captured and therefore removed from the atmosphere. However, to allow a fair

comparison with the WARM model the impact of carbon sequestration is investigated in

Figure 3-8.



Figure 3-8 Gross GHG take-up and emissions per kg of ethylene. The uptake of carbon present in the final ethylene product (0.857 kg carbon per kg ethylene) is subtracted from GWP as it represents an anthropogenic carbon sink. The system-wide GWP then becomes: -1.19 kg CO<sub>2</sub>-eq (Battelle), -1.36 kg CO<sub>2</sub>-eq (MTCI), and -1.16 kg CO<sub>2</sub>-eq (Choren).

Accounting for carbon capture leads to significantly lower GHG emissions associated with ethylene production for all three waste-conversion systems. GWP equals only -1.19 kg CO<sub>2</sub>-eq for the Battelle system (compared to 4.88 and -5.03 kg CO<sub>2</sub>-eq for BAU#1 and BAU#2), -1.36 kg CO<sub>2</sub>-eq for the MTCI system (vs. 4.40 and -4.20 kg CO<sub>2</sub>eq for BAU#1 and BAU#2), and -1.16 kg kg CO<sub>2</sub>-eq for Choren (compared to 3.08 and -1.59 kg CO<sub>2</sub>-eq for BAU#1 and BAU#2). The significance of carbon capture will depend on subsequent use of the chemical feedstock. If used for the production of long-living polymers (PE, PET), carbon recycling could contribute towards GHG savings from a system-wide perspective.

In addition, the fact that PE or PET can be recycled into new polymers via the very same carbon recycling technologies offers the unique opportunity to constantly keep a fraction of the carbon within the technosphere, therefore closing the loop on carbon emissions. Current carbon recycling efficiencies from BMSW to chemical feedstocks are about 19% for the Battelle, 24% for the MTCI, and 33% for the Choren system<sup>42</sup>. With improvements in conversion efficiencies (see sensitivity analysis) this percentage may be increased and more carbon kept in the loop. For the remainder of the results section, the effect of biogenic carbon capture on GWP impacts is not taken into account.

# E.2. Cumulative Energy Demand (CED)

CED of the MSW-based routes (Figure 3-9) is 27.8 (Choren) to 30.3 (Battelle) MJ-eq; less than half that of fossil-based ethylene (75.7 MJ-eq). The CED indicator encompasses non-renewable (i.e. fossil and nuclear) as well as renewable (i.e. biomass, wind, solar, etc.) energy demand. However, renewables account for less than 1% of total CED. This is due to the fact that the intrinsic energy content of the waste feedstock is not captured. In contrast, for fossil-based ethylene the direct and indirect energy consumption of e.g. natural gas and crude oil resources used to synthesize the ethylene product (some of which is later present as 'feedstock energy' in the final product) are accounted for in the CED indicator. Energy offsets from LFG recovery are not enough to offset this. As a

<sup>&</sup>lt;sup>42</sup> Carbon conversion efficiency = Chemical Feedstock [Kg<sub>carbon out</sub>]/Waste Feedstock [kg<sub>carbon in</sub>] x 100%.

result, all three carbon recycling systems score significantly lower than their respective BAU#1 (landfilling) counterparts.

In BAU#2, energy recovery from incineration, leading to offsets of carbonintensive (U.S. average) electricity, exceeds the energy requirements of producing fossilbased ethylene and MSW sorting, resulting in a negative CED of -31.4 (Battelle) and -17.2 MJ-eq (MTCI), respectively. In contrast to this, BAU#2 of the Choren system has a positive CED of 21.9 MJ-eq, roughly similar to the corresponding carbon recycling route (27.3 MJ-eq). The reason is a lower requirement in BMSW feedstock due to higher conversion efficiency as discussed before.

Similar to GWP, steam cracking and MSW classification account for a large share of CED. The magnitude to which MSW conversion and FTS contribute to total CED depends on the amounts of waste feedstock transported to the gasifier and further energy and materials requirements of the conversion facility.



Figure 3-9 CED (MJ-eq) of producing 1 kg ethylene from MSW compared to the production of an equivalent amount of fossil-based ethylene and treatment of the BMSW via conventional landfilling (BAU #1) or incineration with energy recovery (BAU#2). Electricity is assumed to offset energy from the U.S. power grid. Error bars show the 95% confidence interval.

### E.3. Total Material Requirement (TMR)

For TMR, carbon recycling seems beneficial if compared to BAU#1 (landfilling) but worse in comparison to current BAU#2 systems (incineration) (Figure 3-10). Again, energy offsets of U.S. power (TMR: 1.51 kg/kWh (Ritthoff 2011)) significantly lowers the impacts of both BAU cases. For the carbon recycling routes, a large share of TMR is due to steam cracking (1.61 kg per kg ethylene) followed by gasification (Battelle: 1.27 kg; MTCI: 1.13 kg) and MSW classification (Battelle: 0.93 kg; MTCI: 0.81 kg) due to energy requirements associated with waste sorting and separation. During FTS small amounts of excess electricity generated offsets average U.S. power (-0.11 kg (Battelle) and -0.28 kg (MTCI) per kg ethylene).

The comparative BAU#1 scenarios range between 13.0 kg (MTCI) and 14.4 kg (Battelle) with most of the TMR being due to construction materials required for the landfill site. Although, electricity is generated from LFG recovery, the kWh offsets are not sufficient to significantly reduce overall TMR (0.51 kg/kg waste). On the other hand, waste incineration, generating significantly more electricity per kg BMSW, leads to a slightly negative TMR of -0.22 kg/kg waste, therefore reducing overall TMR in BAU#2. It should be noted that material intensities for both waste incinerators and landfills originate from typical sites in Germany (Schmidt 2003) and overall results will vary depending on the type of construction and O&M materials used, as well as assumptions made on plant life-time and treatment capacity over time.



Figure 3-10 TMR (kg) of producing 1 kg ethylene from MSW compared to the production of an equivalent amount of fossil-based ethylene and treatment of the BMSW via conventional landfilling (BAU #1) or incineration with energy recovery (BAU#2). Electricity is assumed to offset energy from the U.S. power grid. \*TMR for the Choren system has been excluded due to the aggregated nature of the dataset. Error bars show the 95% confidence interval.

# **E.4.** Acidification

Impacts to acidification (Figure 3-11) are due to on-site emissions of sulfur dioxide and nitrogen oxides during gasification, FTS, and steam cracking (off-gases) as well as electricity inputs to MSW classification. While the off-gas emission profile from gasification is based on actual organic waste treatment (Khoo 2009) and those of FTS on biomass gasification (Marano and Ciferno 2001), FT-naphtha steam cracking comes from conventional fossil-based naphtha steam crackers using crude-oil due to a lack of better emissions data <sup>43</sup> (CPM 2010; Dancuar et al. 2003; UBA 2010). While FT-naphtha is generally free of sulfur and nitrogen compounds (de Klerk 2007, 2008), fossil-based naphtha is likely to carry higher amounts of these pollutants contributing to the acidification potential. The contributions of naphtha steam cracking to acidification may therefore be rather conservative estimates of actual emissions. With 0.609 kg H+ moleseq the Choren system leads to the lowest acidification potential compared to 0.690 and 0.750 kg H+ moles-eq for the MTCI and Battelle system, respectively. Steam cracking contributes roughly 0.283 kg H+ moles-eq to all routes and the impacts of gasification range between 0.210 (MTCI) and 0.217 kg H+ moles-eq (Battelle). Impacts from FTS are 0.050 and 0.081 kg H+ moles-eq for the MTCI and Battelle systems respectively, while the cumulative burdens of gasification/FTS for the Choren configuration is 0.241 kg H+ moles-eq. The impacts of MSW classification depend on the amounts of BMSW feedstock required for gasification and range between 0.085 and 0.169 kg H+ moles-eq.

<sup>&</sup>lt;sup>43</sup> However, yields are based on FT-naphtha steam cracking test runs for the production of light olefins.

When compared to BAU#1 (landfilling), carbon recycling scores significantly better with regards to the acidification potential, even when uncertainties associated with the inventory input parameters are taken into account. The bulk of impacts associated with BAU#1 are due to fossil-based ethylene production.

In contrast, energy offsets in BAU#2 result in negative acidification potentials of -0.438 and -0.180 kg H+ moles-eq for Battelle- and MTCI-related scenarios, and a positive impact of 0.629 kg H+ moles-eq for the Choren BAU scenario. Uncertainties associated with waste-derived ethylene production via the Choren plant do not allow a clear determination as to whether this route may result in significantly lower emissions of acidification-causing substances than its corresponding BAU#2 case.



Figure 3-11 Acidification (kg H+ moles-eq) of producing 1 kg ethylene from MSW compared to the production of an equivalent amount of fossil-based ethylene and treatment of the BMSW via conventional landfilling (BAU #1) or incineration with energy recovery (BAU#2). Electricity is assumed to offset energy from the U.S. power grid. Error bars show the 95% confidence interval.

Substances contributing the largest share to the acidification potential are shown in Figure 3-12. These include sulfur dioxide and nitrogen oxides.



Figure 3-12 Share of substances contributing to the acidification potential of waste-based ethylene production via the Battelle route. A cut-off of 0.1% was applied.

The following figure shows that the acidification impacts are not only due to onsite emissions at the MSW conversion facility but to a large extent emissions occurring up-stream in the supply chain, e.g. where conventional electricity is produced from fossil raw materials (i.e. coal, natural gas, etc.), commercial palladium production used as catalysts for the chemical synthesis, and transportation e.g. for BMSW.



Figure 3-13 Process contributions to the acidification potential of the Battelle system. The functional unit is 1 kg ethylene at the factory gate. A cut-off of 2.2% was applied.

# E.5. Smog

On-site smog potentials are mainly a result of gasification and FT-naphtha steam cracking. Gasification of MSW leads to the emissions of nitrogen oxides and volatile organic compounds (VOCs), as well as other smog-causing substances present in the waste feedstock. The combined smog potential of gasification and FTS is 0.0054 and 0.0057 g NO<sub>x</sub>-eq for the MTCI and Battelle conversion systems respectively. This is only slightly above the smog potential of 0.0048 g NO<sub>x</sub>-eq given for the Choren plant. It should be noted that the RENEW project used to obtain Choren data is based on different assumptions with regards to emissions profiles (see (Jungbluth, Frischknecht, et al. 2007; RENEW 2006) for the detailed RENEW LCIs) as their study assumes the use of short rotation wood (i.e. willow-salix) instead of BMSW. However, as the Choren dataset is used to cross-check our results, it was decided not to modify existing emission profiles.

With 0.0079-0.0092 g NO<sub>x</sub>-eq the three carbon recycling routes score only slightly above their respective BAU#1 (landfilling) scenarios (0.0052-0.0083 g NO<sub>x</sub>-eq) (Figure 3-14). However, as indicated by the 95% confidence interval, uncertainties associated with smog are large, reflecting e.g. the influence of the choice of data originating from related technologies in various parts of the world (DQIs) and the state of implementation.

When compared to BAU#2, carbon recycling leads to a much higher smog potential. With 0.0012 to 0.0022 g  $NO_x$ -eq (Battelle-Choren), all three incineration scenarios result in the lowest smog potentials. As for previous impact categories, electricity offsets from waste incineration contribute to the low impacts.



Figure 3-14 Smog (g NOx-eq) of producing 1 kg ethylene from MSW compared to the production of an equivalent amount of fossil-based ethylene and treatment of the BMSW via conventional landfilling (BAU #1) or incineration with energy recovery (BAU#2). Electricity is assumed to offset energy from the U.S. power grid. Error bars show the 95% confidence interval.

Finally, Figure 3-15 shows a Sankey diagram for the Battelle system showing the relative contributions of various unit processes to the smog potential.



Figure 3-15 Sankey diagram showing the contributions of unit processes of the Battelle process to smog potential. The functional unit is 1 kg ethylene at the factory gate. The width of the arrows is proportional to contributions to GWP. A cut-off of 4.3% was applied.

# E.6. Water Use

Water use is dominated by inputs to the gasifier (Figure 3-17). These include direct inputs of feedwater for the generation of steam, cooling, and wet scrubbers (syngas cleaning). In addition, water is used in many of the chemicals inputs such as sodium hydroxide and sulfuric acid (Ecoinvent 2010). Overall water use for all three systems equals 0.0107 m3 (Battelle), 0.0103 m3 (MTCI), and 0.0126 m3 (Choren) per kg ethylene generated. This compares to only 0.000899-0.00167 m3 water for the corresponding BAU#1 scenarios (landfilling). Clearly, carbon recycling seems more water intensive than landfilling.

However, when compared to BAU#2 in which BMSW feedstock is incinerated, results indicate that carbon recycling results in less system-wide water use. Water use in BAU#2 ranges between 0.0194-0.0421 m3 with most being due to flue gas treatment<sup>44</sup> (wet scrubbers, quenching, etc.), cooling, etc. during incineration.

<sup>&</sup>lt;sup>44</sup> According to ELCD (European Commission 2011), the modeled incineration plant represents a mix of wet and dry flue gas cleaning (EU average conditions).



Figure 3-16 Water use (m3) of producing 1 kg ethylene from MSW compared to the production of an equivalent amount of fossil-based ethylene and treatment of the BMSW via conventional landfilling (BAU #1) or incineration with energy recovery (BAU#2). Electricity is assumed to offset energy from the U.S. power grid. Error bars show the 95% confidence interval.

The following figure shows a Sankey diagram with the relative contributions of

the Battelle life cycle to water use.



Figure 3-17 Sankey diagram showing the contributions of unit processes of the Battelle process to water use. The functional unit is 1 kg ethylene at the factory gate. The width of the arrows is proportional to contributions to GWP. A cut-off of 5.5% was applied.

# E.7. Sensitivity Analysis

We test the sensitivity of assumptions made on results of the LCA by varying the input parameters. This includes assumptions made with regards to 1.) Allocation (physical vs. economic), 2.) Future low carbon energy mix and energy substitution, 3.) Increased conversion efficiency, and 4.) Energy inputs to the steam cracker.

## E.7.1. Physical vs. Economic Allocation

The choice of allocation can have an impact on upstream environmental burdens. For the results discussed in earlier sections of this chapter, we applied allocation based on physical relationships, including mass and energy. Co-product credits were given only for electricity produced during FTS, assumed to offset conventional fossil-based power. We test the robustness of our assumptions below by applying allocation based on the economic value of all (co-)products produced.

Table 3-12 shows how allocation percentages change if market prices are applied.

		MSW class	ification <sup>1</sup>		FTS (B MTC	attelle, CI) <sup>2</sup>	FTS (CI	noren)²	FT-naphtha Steam Cracking			
	BMS W	Recyclable s	Compos -tables	Scrap s	Naphtha	Diesel	Naphth a	Diesel	Ethylene	Propy- lene	Other HCs <sup>3</sup>	
Amount [kg]	0.37	0.24	0.09	0.30	0.41	0.59	0.17	0.83	1.00	0.37	0.71	
LHV [MJ/kg] Physical	-	-	-	-	44.43	43.96	43.70	44.00	-	-	-	
Allocation [%]	37.00	24.00	9.00	30.00	41.58	58.42	16.90	83.10	48.19	17.59	34.22	
Price [Jan 2011 [\$/tonne]	0.00	50.00	20.00	0.00	963.00	994.17	963.00	994.1 7	1168.45	1565.28	963.00	
Economic Allocation % I	0.00	86.96	13.04	0.00	40.55	59.45	16.56	83.44	48.21	23.57	28.21	

Table 3-12 Allocation percentages according to physical relationships (mass and energy) vs. economic value of the various product outputs.

All prices are given in January 2011 U\$D. <sup>1</sup>It is assumed that the price of all waste fractions going to landfills or incinerators (i.e. BMSW and scraps) equals zero. In reality, consumers may have to pay a tipping fee representing a revenue stream. <sup>2</sup>Electricity co-produced is treated via co-product credits. <sup>3</sup>The price of other hydrocarbons (HCs) is assumed to be equal to January 2011 naphtha prices. Prices originate from the following sources: Recyclables and compostables (own assumption); Naphtha (www.icispricing.com); Distillate (www.eia.gov); Ethylene and propylene (CMAI 2011).
In this table, we assume that the market price for outputs from the MSW classification process is zero for waste fractions such as BMSW and scraps going to a landfill or incinerator. In reality, the WM-company is likely to have to pay a tipping fee to the landfill operator for disposing of these waste fractions for which currently no market exists. The table also shows that the economic allocation percentages for outputs from FTS and steam cracking are roughly similar to the previously applied physical allocation numbers. As a result, it should be expected that reductions in environmental burdens will be mainly due to MSW classification, which is excluded from the MSW-based ethylene product considering market prices. This is shown in Figure 3-18 and Table 3-13.



Figure 3-18 Results of the sensitivity analysis showing the relative environmental impact of physical (phys) vs. economic (econ) allocation of the three carbon recycling systems under investigation. Impacts for economic allocation are shown relative to physical allocation (100%) for each impact category and system. \*TMR has been excluded for the Choren system due to a lack of non-aggregated data.

Carbon- recycling system		Battelle		M	TCI	Choren		
Impacts	Unit	Phys	Econ	Phys	Econ	Phys	Econ	
GWP	kg CO2 eq	1.959E+00	1.475E+00	1.786E+00	1.368E+00	1.990E+00	1.737E+00	
CED	MJ eq	3.035E+01	2.234E+01	2.745E+01	2.054E+01	2.779E+01	2.369E+01	
TMR	kg	3.696E+00	2.731E+00	3.259E+00	2.425E+00	2.243E+00	1.767E+00	
Acidification	H+ moles eq	7.499E-01	5.720E-01	6.900E-01	5.354E-01	6.095E-01	5.172E-01	
Smog	g NOx eq	9.172E-03	8.099E-03	8.746E-03	7.802E-03	7.864E-03	7.267E-03	
Water use	m3	1.069E-02	9.171E-03	1.029E-02	8.951E-03	1.261E-02	1.167E-02	

Table 3-13 Results of the sensitivity analysis showing the environmental impacts of physical (phys) vs. economic (econ) allocation of the three carbon recycling systems under investigation.

Environmental impacts decrease by about 7-26% when economic allocation is applied, depending on the impact category and system looked at. The most significant decreases in environmental burdens can be found for the Battelle and MTCI systems as these require large amounts of BMSW per kg ethylene, with energy-intensive MSW classification being excluded during economic allocation.

# E.7.2. Energy Substitution & Future Low Carbon Energy Mix

The choice of the energy mix is tested by modeling: 1.) An energy scenario in which energy inputs to the foreground system come from the U.S. average power grid but electricity generated by the carbon recycling systems and WM schemes (landfills and incinerators) offsets marginal<sup>45</sup> (non-baseload) power

<sup>&</sup>lt;sup>45</sup> Consisting of electricity from coal-fired power plants. The marginal power mix is included to be in line with the WARM model (EPA 2010a), used to model GHG emissions associated with landfilling (BAU), which assumes that energy from LFG recovery would offset electricity generation at a marginal, fossil-fuel-only environmental burdens rate. For simplicity, WARM currently uses the national weighted average of fossil-fuel plants as a proxy for the fuels displaced

('Marginal') following assumption from the EPA WARM model (EPA 2009a), and 2.) A future energy scenario in which power demands and offsets in the foreground system of the carbon recycling schemes and BAU scenarios comes from low-carbon sources ('*BLUEMap*') following envisioned energy shares from the IEA (IEA 2010) for the year 2050. Both scenarios are compared to the conventional case ('*Conv*'), in which energy inputs to the foreground system stem from the U.S. average power grid and energy offsets (from electricity coproduced by the FTS plants as well as by the landfills and incinerators) replace an equivalent amount of energy from the U.S. average power grid.

Results for the three carbon recycling schemes and their respective BAU scenarios are shown in the following figures and tables.

at the margin when electricity is displaced by electricity from landfills/incinerators. The fossil-fuel mix does not include nuclear power. Non-fossil sources are expected to meet baseload requirements because of the financial incentive to operate these at capacity.



Figure 3-19 Sensitivity with regards to the choice of the energy mix to processes of the foreground system. The figure shows the Battelle carbon recycling system and its respective BAU cases under various energy scenarios. Impacts are shown relative to the system with the highest environmental burdens (100%) for each category. The functional unit is 1 kg ethylene. Conv: Conventional fossil-based energy mix. Marginal: Marginal power mix. BLUEMap: Future low-carbon energy mix in the year 2050 according to (IEA 2010).



Figure 3-20 Sensitivity with regards to the choice of the energy mix to processes of the foreground system. The figure shows the MTCI carbon recycling system and its respective BAU cases under various energy scenarios. Impacts are shown relative to the system with the highest environmental burdens (100%) for each category. The functional unit is 1 kg ethylene. Conv: Conventional fossil-based energy mix. Marginal: marginal power mix. BLUEMap: Future low-carbon energy mix in the year 2050 according to (IEA 2010).



Figure 3-21 Sensitivity with regards to the choice of the energy mix to processes of the foreground system. The figure shows the Choren carbon recycling system and its respective BAU cases under various energy scenarios. Impacts are shown relative to the system with the highest environmental burdens (100%) for each category. The functional unit is 1 kg ethylene. Conv: Conventional fossil-based energy mix. Marginal: Marginal power mix. BLUEMap: Future low-carbon energy mix in the year 2050 according to models by the IEA (IEA 2010). \*TMR was excluded due to a lack of data for the Choren process.

Table 3-14 Sensitivity with regards to the choice of the energy mix to processes of the foreground system.

Impact category	Battelle (Conv)	Battelle (Marginal)	Battelle (BLUEMap)	Landfill (BAU)	Landfill (Marginal)	Landfill (BLUEMap)	Inciner. (BAU)	Inciner. (Marginal)	Inciner. (BLUEMap)
GWP 100a [kg CO2-eq]	1.96E+00	1.85E+00	1.72E+00	4.91E+00	4.12E+00	5.29E+00	- 5.03E+00	-1.03E+01	2.20E-01
CED [MJ-eq]	3.03E+01	3.01E+01	2.94E+01	8.21E+01	7.99E+01	8.49E+01	- 3.14E+01	-4.34E+01	-9.17E+00
TMR [kg]	3.70E+00	3.41E+00	3.47E+00	1.44E+01	1.19E+01	1.51E+01	5.92E-01	-1.32E+01	5.79E+00
Acidification [H+ moles-eq]	7.50E-01	7.18E-01	6.89E-01	1.55E+00	1.26E+00	1.71E+00	-4.44E-01	-2.00E+00	9.18E-01
Smog [g NOx-eq]	9.17E-03	8.90E-03	8.90E-03	8.32E-03	5.91E-03	8.94E-03	1.16E-03	-1.21E-02	7.07E-03
Water use [m3]	1.07E-02	1.06E-02	1.05E-02	1.66E-03	4.27E-04	2.26E-03	4.20E-02	3.52E-02	4.58E-02

The functional unit is 1 kg ethylene. Conv: Conventional fossil-based energy mix. Marginal: Marginal power mix. BLUEMap: Future lowcarbon energy mix in the year 2050 according to (IEA 2010).

Table 3-15 Sensitivity with regards to the choice of the energy mix to processes of the foreground system.

Impact category	MTCI (Conv)	MTCI (Marginal)	MTCI (BLUEMap)	Landfill (BAU)	Landfill (Marginal)	Landfill (BLUEMap)	Inciner. (BAU)	Inciner. (Marginal)	Inciner. (BLUEMap)
GWP 100a [kg CO2-eq]	1.79E+0 0	1.62E+00	1.66E+00	4.42E+00	3.74E+00	4.75E+00	- 4.20E+00	-8.81E+00	3.51E-01
CED [MJ-eq]	2.75E+0 1	2.71E+01	2.69E+01	8.13E+01	7.94E+01	8.37E+01	- 1.72E+01	-2.77E+01	2.05E+00
TMR [kg]	3.26E+0 0	2.82E+00	3.14E+00	1.30E+01	1.08E+01	1.37E+01	1.03E+00	-1.09E+01	5.55E+00
Acidification [H+ moles-eq]	6.90E-01	6.40E-01	6.58E-01	1.54E+00	1.30E+00	1.68E+00	-1.85E-01	-1.53E+00	9.93E-01
Smog [g NOx-eq]	8.75E-03	8.32E-03	8.60E-03	7.62E-03	5.53E-03	8.13E-03	1.40E-03	-1.01E-02	6.51E-03
Water use [m3]	1.03E-02	1.01E-02	1.02E-02	1.46E-03	3.90E-04	2.00E-03	3.65E-02	3.06E-02	3.98E-02

The functional unit is 1 kg ethylene. Conv: Conventional fossil-based energy mix. Marginal: marginal power mix. BLUEMap: Future lowcarbon energy mix in the year 2050 according to (IEA 2010).

Table 3-16 Sensitivity with regards to the choice of the energy mix to processes of the foreground system.

Impact category	Choren (Conv)	Choren (Marginal)	Choren (BLUEMap)	Landfill (BAU)	Landfill (Marginal)	Landfill (BLUEMap)	Inciner. (BAU)	Inciner. (Marginal)	lnciner. (BLUEMap)
GWP 100a [kg CO2-eq]	1.99E+00	1.99E+00	1.81E+00	3.09E+00	2.70E+00	3.26E+00	- 1.90E+00	-4.57E+00	7.11E-01
CED [MJ-eq]	2.78E+01	2.78E+01	2.70E+01	7.89E+01	7.78E+01	8.03E+01	2.19E+01	1.58E+01	3.30E+01
TMR* [kg]	-	-	-	9.18E+00	7.92E+00	9.58E+00	2.23E+00	-4.69E+00	4.88E+00
Acidification [H+ moles-eq]	6.09E-01	6.09E-01	5.64E-01	1.53E+00	1.39E+00	1.60E+00	5.27E-01	-2.53E-01	1.20E+00
Smog [g NOx-eq]	7.86E-03	7.86E-03	7.66E-03	5.68E-03	4.47E-03	5.92E-03	2.08E-03	-4.56E-03	4.98E-03
Water use [m3]	1.26E-02	1.26E-02	1.25E-02	9.09E-04	2.86E-04	1.27E-03	2.12E-02	1.78E-02	2.32E-02

The functional unit is 1 kg ethylene. Conv: Conventional fossil-based energy mix. Marginal: Marginal power mix. BLUEMap: Future lowcarbon energy mix in the year 2050 according to models by the IEA (IEA 2010). \*TMR was excluded due to a lack of data for the Choren process. Results of the assessment show that assuming *marginal* energy offsets leads to minimal changes in environmental burdens associated with the carbon recycling schemes<sup>46</sup>. This is due to only small amounts of power co-produced by the FTS unit. For WtE technologies (landfills and incinerators), replacing more carbon-intensive energy at the margins leads to lower environmental burdens. Incineration, due to high energy recovery, results in larger energy offsets than landfilling.

Under a future energy scenario (*BLUEMap*), environmental burdens associated with carbon recycling change only slightly due the fact that small amounts of power are required by the conversion systems (most of this for MSW classification), and power coproduced by the FTS unit is small. For the BAU scenarios, impacts significantly increase due to less carbon-intensive energy being replaced by conventional WtE systems. Avoided burdens associated with energy offsets in the future (Table 3-11) are lower than current power production (either U.S. average or marginal power) as the BLUE Map power mix use larger shares of renewables and coal-fired power plants applying CCS.

## E.7.3. Increased Conversion Efficiency

According to LCI data collected, the Battelle and MTCI systems have LHV conversion efficiencies of 27-31% from BMSW to FT-liquids. However, as shown in Table 3-8, pressurized and technically optimized gasification/FTS plants can have higher conversion efficiencies of up to 50-60%. We assume that further technological progress will allow the carbon recycling systems under investigation to reach LHV conversion

<sup>&</sup>lt;sup>46</sup> Environmental impacts of the Choren system's conventional and marginal energy scenario stay the same as no power is delivered to the grid and therefore no marginal energy offsets take place.

efficiencies of 50% from BMSW to FT-liquids in the near-to-medium term. Results of this analysis are shown below.



Figure 3-22 Influence of an increased LHV conversion efficiency (BMSW to FT-liquids) on environmental burdens associated with 1 kg ethylene production via the Battelle and MTCI routes. Environmental impacts are shown relative to the current system configurations (i.e. 26.85% LHV efficiency for Battelle and 31.25% LHV efficiency for MTCI). The Choren system has been excluded as it is already operated with an efficiency of roughly 53% (see Table 3-8).

Table 3-17 Influence of an increased LHV conversion efficiency (BMSW to FT-liquids) on environmental burdens associated with 1 kg ethylene production via the Battelle and MTCI system.

Impact category	Unit	Battelle 26.85% LHV	Battelle 50.00% LHV	MTCI 31.25% LHV	MTCI 50.00 % LHV
<b>GWP 100a</b>	kg CO2 eq	1.959E+00	1.654E+00	1.786E+00	1.571E+00
CED	MJ eq	3.035E+01	2.530E+01	2.745E+01	2.391E+01
TMR	kg	3.696E+00	2.676E+00	3.259E+00	2.533E+00
Acidification	H+ moles eq	7.499E-01	5.710E-01	6.900E-01	5.560E-01
Smog	g NOx eq	9.172E-03	6.744E-03	8.746E-03	6.876E-03
Water use	m3	1.069E-02	7.014E-03	1.029E-02	7.372E-03

The current system configurations are: 26.85% LHV efficiency for Battelle and 31.25% LHV efficiency for MTCl. We assume that in the near-to-medium term both systems may reach LHV efficiencies of 50%. The Choren system has been excluded as it is already operated with an efficiency of roughly 53% (see Table 3-8).

Being able to operate the carbon recycling systems with a higher conversion efficiency of 50.00% (LHV) instead of the currently 27-31% (LHV) would lead to a reduction in GWP and CED of roughly 26% for the Battelle and 23% for the MTCI systems, respectively. TMR would decrease from currently 3.7 to 2.7 kg/kg ethylene (27%) for the Battelle system and from 3.3 to 2.5 kg/kg ethylene (24%) for the MTCI system. The acidification potential decreases by approximately 24% from 0.75 to 0.57 H+ moles-eq (Battelle) and 19% from 0.69 to 0.56 H+ moles-eq. Smog decreases by about 26% for the Battelle and 21% for the MTCI configuration. The largest decrease in environmental burdens can be found for water use (34% for the Battelle and 28% for the MTCI configuration). However, as the higher conversion efficiency does not affect the FT-naphtha steam cracker (only the efficiency of converting BMSW into FT-liquids is modified), a rather energy-intensive process is not included. Given the higher conversion efficiency of 50% (LHV), about 10.2 kg and 10.3 kg BMSW are required per kg ethylene for the Battelle and MTCI configurations, respectively. This compares to currently 8.63 kg BMSW per kg ethylene for the Choren system with a conversion efficiency of roughly

53% (LHV). Finally, the following two figures show that even if uncertainty from the MC analysis is taken into account the carbon recycling systems with the higher conversion efficiency lead to lower impact scores when compared to current configurations.



Figure 3-23 Monte Carlo results of the comparison looking at Battelle 26.85% LHV vs. 50.00% LHV conversion efficiency. The number of outcomes in which 26.85% LHV (A) has a higher score than 50.00% (B) is shown per impact category. Results show that the differences shown in Table 3-17 are indeed significant (90 – 95% of the MC runs are favorable for the 50% LHV case).



Figure 3-24 Monte Carlo results of the comparison looking at MTCI 31.25% LHV vs. 50.00% LHV conversion efficiency. The number of outcomes in which 26.85% LHV (A) has a higher score than 50.00% (B) is shown per impact category. Results show that the differences shown in Table 3-17 are indeed significant (90 – 95% of the MC runs are favorable for the 50% LHV case, except for CED and GWP).

## E.7.4. Energy Inputs to the Steam Cracker

As shown in the results section, energy inputs (heat) to the steam cracker contribute significantly to the system-wide environmental impacts of all ethyleneproducing carbon recycling systems. As discussed in section D.5, energy inputs to the steam cracker may represent a conservative estimate and therefore varying energy inputs are discussed below.



Figure 3-25 Influence of varying energy inputs to the naphtha steam cracker of the Battelle system. Results are shown relative to the current configuration (100%) in which 25.914 MJ of thermal energy are required for the generation of 1 kg ethylene and various hydrocarbons (see LCI in Table 3-9). Inputs of thermal energy are varied between 15-30 MJ. The functional unit is 1 kg of ethylene. Only results for the Battelle system are shown as the trend is similar for all three carbon recycling systems.

Battelle	Unit	15 MJ	20 MJ	25.914 MJ*	30 MJ
IPCC GWP 100a	kg CO2 eq	1.397E+00	1.654E+00	1.959E+00	2.170E+00
CED	MJ eq	2.174E+01	2.568E+01	3.035E+01	3.357E+01
TMR	kg	3.024E+00	3.332E+00	3.696E+00	3.947E+00
Acidification	H+ moles eq	6.656E-01	7.043E-01	7.499E-01	7.815E-01
Smog	g NOx eq	8.504E-03	8.810E-03	9.172E-03	9.423E-03
Water use	m3	1.031E-02	1.048E-02	1.069E-02	1.084E-02
MTCI					· · · · · · · · · · · · · · · · · · ·
IPCC GWP 100a	kg CO2 eq	1.224E+00	1.481E+00	1.786E+00	1.997E+00
CED	MJ eq	1.884E+01	2.278E+01	2.745E+01	3.067E+01
TMR	kg	2.587E+00	2.895E+00	3.259E+00	3.511E+00
Acidification	H+ moles eq	6.057E-01	6.443E-01	6.900E-01	7.215E-01
Smog	g NOx eq	8.077E-03	8.383E-03	8.746E-03	8.996E-03
Water use	m3	9.906E-03	1.008E-02	1.029E-02	·1.044E-02
Choren**					
IPCC GWP 100a	kg CO2 eq	1.428E+00	1.685E+00	1.990E+00	2.201E+00
CED	MJ eq	1.918E+01	2.313E+01	2.779E+01	3.102E+01
Acidification	H+ moles eq	5.252E-01	5.638E-01	6.095E-01	6.410E-01
Smog	g NOx eq	7.196E-03	7.502E-03	7.864E-03	8.114E-03
Water use	m3	1.222E-02	1.240E-02	1.261E-02	1.275E-02

Table 3-18 Influence of varying energy inputs to the naphtha steam cracker.

Inputs of thermal energy are varied between 15-30 MJ. The functional unit is 1 kg of ethylene. \*Current configuration according to the life cycle inventory compiled in Table 3-9. \*\*TMR has been excluded due to a lack of data.

Slightly lower thermal energy inputs to the steam cracker of 5 MJ have the potential to lead to reductions in the order of 25% of GWP and CED, 7-10% for TMR and acidification, and 2-5% for smog and water use. Hence, system optimizations leading to lower steam requirements for this process have the potential to lead to reduced impacts, in particular with regards to GWP and CED.

# F.Life-Cycle Cost Analysis

*Capital cost figures* are taken from the following sources: waste classification (Broder et al. 1993), gasification (Niessen et al. 1996), gas cleaning equipment and oxygen plant (Hamelinck 2004), FTS system (Van Bibber et al. 2007), and steam cracker (Ren et al. 2009).

*Material & energy costs and revenues* were added to the inputs and outputs of the life cycle inventory. These include amongst others: US\$0.0681 per kWh for electrical power (USEIA 2011), BMSW tipping fee of US\$30 per MT (Valkenburg et al. 2008), US\$0.994 per kg FT-diesel<sup>47</sup>, US\$1.565 per kg propylene (CMAI 2011), US\$0.963 per kg hydrocarbons<sup>48</sup> (all in Jan 2011 US\$). If no information on operators was available, the labor cost was assumed to be 0.5% of TCI. Maintenance costs for plant equipment were calculated as 3% of TCI cost (unless more detailed data was available). Other cost estimates, including expenditures for insurance, administration, and contingencies, are taken from (Vogel et al. 2007). These are: Insurance 1% p.a. of TCI, Administration

<sup>&</sup>lt;sup>47</sup> www.eia.gov

<sup>&</sup>lt;sup>48</sup> Assuming that the price is equal to the market price of naphtha from www.icispricing.com

0.5% p.a. of TCI, Contingencies: 1% p.a. of TCI, Others: 0.75% p.a. of TCI. For biomass-based FTS systems previously investigated O&M costs are generally calculated as 3% - 4.5% of TCI (van Vliet et al 2009).

# **CHAPTER 4**

# LIFE CYCLE ASSESSMENT (LCA) OF CONSTRUCTION AND DEMOLITION (C&D) DERIVED BIOMASS GASIFICATION FOR ELECTRICITY PRODUCTION <sup>49</sup>

#### A. Abstract

With the goal to move society towards less reliance on fossil fuels and the mitigation of climate change, there is increasing investment and interest in the bioenergy sector. However, current bioenergy growth patterns may in the long term only be met through an expansion of global arable land at the expense of natural ecosystems and in competition with the food-sector. Increasing thermal energy recovery from solid waste reduces dependence on fossil- and bio-based energy production while enhancing landfill diversion. Using inventory data from pilot processes, this work assesses the cradle-togate environmental burdens of plasma gasification as a route capable of transforming construction & demolition derived biomass (CDDB) and forest residues into electricity. Results indicate that the environmental burdens associated with CDDB and forest residue

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gasification may be similar to conventional electricity generation. Land occupation is lowest when CDDB is used. Environmental impacts are to a large extent due to coal cogasified, coke used as gasifier bed material, and fuel oil co-combusted in the steam boiler. However, uncertainties associated with preliminary system designs may be large, particularly the heat loss associated with pilot scale data resulting in overall low efficiencies of energy conversion to electricity; a sensitivity analysis assesses these uncertainties in further detail.

Keywords: Life Cycle Assessment (LCA), Plasma Gasification, Waste Management, Construction & Demolition Waste, Forest Biomass, Electricity Production

#### **B.Introduction**

Renewable energy is expected to contribute towards sustainable development and reduce impacts in particular related to global climate change. By 2007, at least 64 countries around the world had set a national target for the share of renewables in their energy mix (Bringezu, Schütz, O'Brien, et al. 2009). In this context, biomass for energy and liquid fuels production is considered as an alternative to fossil-based energy systems by countries worldwide. In 2007, biomass provided about  $45 \pm 10$  EJ to global final energy consumption (out of a global total of 388 EJ per year), therefore being the largest renewable energy source used (IEA 2007). However, the majority of bioenergy is currently due to traditional biomass use such as cooking and heating, particularly in rural areas of the developing world (REN21 2011). In contrast, commercial energy production

from biomass for industry, biofuels, and power generation represents a lower but still significant share (some 7 EJ per year in 2000) (IEA 2007).

Biomass power contributed a total of 1.25%<sup>50</sup> to the global total power generation capacity of 4950 GW in 2010 (REN21 2011). In the United States, most biomass electricity comes from wood and agricultural residues as well as black liquor burned during cogeneration by industry. It is expected that total global primary energy demand in 2050 could reach between 800 and 1,400 EJ. The estimated global potential for bioenergy production is estimated to be between 200-400 EJ.

In view of current efforts to increase commercial bioenergy supplies, the availability of global arable land for non-food purposes requires special attention. The current growth in bioenergy production may in the long term only be met through an expansion of global arable land at the expense of natural ecosystems and in direct competition with the food-sector (Bringezu and Schütz 2008; Bringezu, Schütz, Arnold, et al. 2009; Bringezu, Schütz, O'Brien, et al. 2009). Against this background, the use of waste and production residues for bioenergy production is gaining increased interest as an alternative to the use of virgin greenwood biomass (Bringezu, Schütz, O'Brien, et al. 2009).

## **B.1. Waste as Bioenergy Feedstock**

Various types of organic waste including biodegradable waste and refuse-derived fuel from municipal waste streams, construction and demolition (C&D) waste, sewage

<sup>&</sup>lt;sup>50</sup> Does not include waste-to-energy capacity from municipal solid waste (MSW).

sludge, and industrial wastes are considered as potential feedstock for bioenergy and chemicals production (Brown et al. 2007, 2009; Juniper Consultancy Services 2001; Nuss et al. 2012).

In 2003 the United States generated an estimated 170 million short tons of C&D debris of which about 48% was recovered (EPA 2003). The wood fraction of C&D debris is a key component for recycling as a feedstock for thermochemical conversion. The amount of wood in C&D debris was found to average 31.5%, ranging from 20.2 to 45.3% in various states of the U.S. (DSM Environmental Services Inc. 2008). Pressure treated wood averaged 1.6% of all C&D waste, while high grade wood consisting of pallets and crates and other unpainted wood made up 11.5% of all C&D waste. Besides high grade wood, requiring little pre-treatment prior to recycling, also painted/stained wood (6.5%) engineered wood (8.1%), wood furniture (0.3%), and other wood (6.0%) may be used for energy recovery.

In the Northeastern (NE) United States, NEWMOA estimates that in 2006 approximately 12 million short tons of C&D waste were generated (NEWMOA 2009). Of this about 10 million tons was sent to landfills, with 70% of the total estimated C&D waste generated disposed as C&D waste and 13% used as alternative daily cover in landfills. Landfilling takes place despite the fact that several alternative options for the management of C&D derived wood exist or are being developed. These include the use of these wood fractions in biomass boilers, particle board manufacturing, gasification and pyrolysis plants, as well as cellulose ethanol production facilities (DSM Environmental Services Inc. 2008).

#### **B.2. Waste-to-Energy Systems**

Besides commonly used waste incinerators for energy generation, gasification and pyrolysis allow the production of a syngas that can be used for the generation of electricity or alternatively as a feedstock for the production of fuels and chemicals via various catalytic conversion routes (Nuss et al. 2012). The treatment of waste feedstock via gasification and pyrolysis is still a relatively novel waste management practice. However, both pyrolysis and gasification have been well regarded for their potential production of useful products from various types of organic waste, as well as for generating less air emissions and residues than conventional waste incineration technologies (Belgiorno et al. 2003; Klein 2002).

Among the various gasification technologies for solid waste treatment (see e.g. (Malkow 2004)), *plasma arc gasification* is seen as a commercially viable option (Young 2010). Plasma gasification is a high-temperature process in which the carbon-based materials of the organic waste stream are converted into syngas (CO and H2), and inorganics produce a glass-like vitrified slag. The high temperature needed to produce the plasma is provided by an electric arch in a torch using electricity. The plasma gasification reactor is typically operated between 3980 to 6980°C (Young 2010). In commercial operations, carbonaceous material such as coal or coke is added to the gasification feed (Juniper Consultancy Services 2008).

## **C. Experimental Section**

#### C.1. Goal and System Boundary

The goal of this study is to assess the system-wide environmental burdens of using plasma arc gasification in the NE United States for the production of renewable energy from feedstock mixes consisting of 1.) C&D derived biomass (CDDB), 2.) forest residues, and 3.) bituminous coal. Results are compared to conventional electricity production on the basis of a functional unit of 1 kWh of electricity at the factory gate. The research method used in this study is attributional life cycle assessment (LCA). Inventory data is obtained from direct communications with our company partner and various technical reports. Technologies included are currently existing processes such as pilot-plant plasma gasification, syngas clean-up systems, boiler and steam turbine, as well as conventional forest harvest and C&D waste sorting and processing systems. The modeled gasification and electricity generation process is located in the NE United States. An LCA model following the ISO 14040 and 14044 standards (ISO 2006a; b) is developed using SimaPro 7.3 software. Figure 4-1 shows the major stages of the product systems, which were investigated as unit processes.



Figure 4-1 System boundary of the plasma gasification system under study. T = Transport

The system under consideration includes sorting of the *C&D waste* in a materials recovery facility (MRF) to obtain the wood fraction, transportation to the plasma arc gasification plant, feed handling and gasification, syngas clean-up and subsequent electricity generation via combustion in a boiler/steam turbine. It is assumed that C&D debris is a waste that requires disposal or alternative uses and that waste collection take place because it is a regulatory requirement or economically feasible due to the value of recovered material (e.g. metals, wood, plastics). The environmental implications of the process that generated the C&D debris as well as waste collection were therefore excluded from the life-cycle analyzed. We assume that CDDB utilized in the NE United States would be diverted from landfills and hence credit the system with the avoided environmental burdens associated with waste wood landfilling (system expansion).

The life cycle of *forest biomass* includes tree cultivation & harvest, forest residues collection and transportation, feed handling and gasification, and subsequent electricity

generation via combustion in a boiler/steam turbine. It is assumed that primary wood residues from commercial logging operations are used as feedstock to the gasifier.

Among the major differences between C&D waste and NE forest residue utilization are wood growth and harvest, avoided landfilling, as well as the transport distance (using pilot plant data we assume that CDDB would be available within a 35 miles radius of the gasification power plant, while forest residues would come from within a 70 miles radius). The shorter transportation distance accounts for the fact that C&D waste processing stations would typically be located in closer proximity to conversion plants than forest biomass.

### C.2. Life-Cycle Inventory Data

The life cycle inventories (LCIs) compiled for C&D waste, forest biomass, and coal gasification are sourced from various reports and direct communications with our company partner. We use a combination of pilot plant and computer simulation data for the plasma gasification reactor (PGR), and data from confidential design reports as well as publicly available data to model the syngas cleanup system and subsequent electricity generation. Resource, material and energy inputs were linked to conventional LCI databases including U.S. LCI (NREL 2008) and Ecoinvent (Ecoinvent 2010) and all data fed into the LCA software SimaPro 7.3.

#### C.2.1.Forest Biomass

With the exception of short rotation forestry, forest biomass represents the prevalent source for wood-based fuels (Röser et al. 2008). In the NE U.S., the primary outputs of forest harvest are sawlogs, pulpwood, and bark as well as stumps and primary residues, typically left in the forests to decay (Oneil et al. 2010). For this study, we assume that primary residues (logging residues excluding stumps) are utilized as feedstock for plasma gasification. According to (Buchholz and S. Hamburg 2011), 77% of current forest biomass removals in the NE represents the merchantable fraction (i.e. sawlogs, pulpwood, and bark), while 4% of it is present in non-usable stumps (left in the forest), and the remainder (19%) represents primary residues such as tops and logging residues (currently left in the forest) that could be diverted to the plasma gasification plant. It should be noted that even though the use of primary wood residuals would not cause any consequential effects due to diversion and competition with other uses (e.g. pulp), it could still have adverse effects on the forest habitat (biodiversity) and soil nutrient fertility, especially if carried out on large scales. One way to counteract nutrient transport away from forests could be to recycle the wood ash from bioenergy plants, thereby compensating for nutrient loss (Röser et al. 2008). For our analysis, inventory data on traditional tree growth and harvest in the NE U.S. and related fuel and lubricant inputs comes from (NREL 2008; Oneil et al. 2010). Natural regeneration is assumed and life-cycle stages include stand establishment, tree harvest, transport of whole trees, delimbing, and loading of the wood onto a truck (NREL 2008). Atmospheric  $CO_2$ assimilation is based on the carbon content of wood fractions, while fuel and lubricant use is allocated based on the dry weight of product outputs. CORRIM (Oneil et al. 2010) simulated three different forest management scenarios for softwood and hardwood growth/harvest (low, medium, high). We use the medium management intensity level in our LCA model and assume primary residues consisting of equal amounts of hard- and softwood are diverted to the power plant. Land occupation estimates per cubic meter of wood are based on typical hardwood and softwood yields per year and rotation cycles published by CORRIM (Oneil et al. 2010). Ecoinvent data (Werner et al. 2007) is used to obtain estimates of land occupied by the building of roads for forest access. Land occupation in the NE forests equals 2.73 and 4.05 m<sup>3</sup>/ha yr for hardwood and softwood respectively. This is close to the U.S. forest average net annual increment (NAI)<sup>51</sup> of 3.64 m<sup>3</sup>/ha yr (W. B. Smith et al. 2009). Finally, forest residues are chipped in a mobile chipper prior to transportation to the conversion plant (Werner et al. 2007). The final product is 1 m<sup>3</sup> mixed softwood/hardwood chips from primary residues with a density of 480 kg/m<sup>3</sup> (dry weight). The average energy content of forest biomass (as received) is 17.31 MJ/kg. Water content varies between 12-26% and is taken from respective test reports of our industry partner.

## C.2.2. C&D Waste

The energy (20.3 kWh/metric ton mixed C&D waste) and fuel inputs (2.4 L diesel fuel/metric ton mixed C&D waste) associated with mixed <u>C&D waste</u> in a MRF are taken

<sup>&</sup>lt;sup>51</sup> NAI represents the average annual volume over a reference period of gross increment less natural losses and hence represents a good estimate for the required forest land area for biomass provision.

from (Levis 2008). Electricity inputs are modeled using fuel shares for the U.S. NE power grid (NEPOOL) (ISO New England 2010) and linking them to Ecoinvent (Ecoinvent 2010) and U.S. LCI (NREL 2008) unit processes. Fuel inputs are modeled using U.S. LCI data for diesel fuel. Allocation of fuel and energy inputs is based on the 'as received' weight of the components sorted (dirt/fines 25%, bricks 5%, concrete 10%, asphalt 6%, corrugated cardboard 7%, ferrous metals 3%, non-ferrous 2%, gypsum wallboard 8%, CDDB 22%, miscellaneous 12%). CDDB obtained is assumed to be pre-processed to a size that can be fed directly into the plasma gasification reactors (PGRs). The average energy content of CDDB (as received) is 10.65 MJ/kg. Water content varies between 16-30%. Feedstock transport is modeled using generic data for a diesel powered combination truck (NREL 2008).

### C.2.3. Plasma Gasification

Data for the unit processes of the plasma gasification power plant (Figure 4-2) are based on confidential test and design reports (AlterNRG 2009; Burak 2010; Darr et al. 2008; SNC-Lavalin 2008; Willis and Harris 2009).



Figure 4-2 Process step schematic of the plasma gasification power plant under consideration.

For the pilot <u>plasma gasification</u> plant, four separate refractory-lined plasma torch air blown gasifiers operating under high temperature and atmospheric pressure are to be used to thermally convert CDDB, forest residues, and coal into syngas. The commercial facility is envisaged to operate with six 600 kW plasma torches. Ash and other inorganic materials present in the feedstock are melted down and flow to the bottom of the PGR forming a slag. The slag (typically 1-5% by weight of feedstock input) exits the gasifier separately from the syngas and is removed from the process by the slag handling system. Gasifiers are designed to operate with metallurgical coke or anthracite. Because of the ash content of the coal, wood chips and coke mixture, flux material (limestone/sandstone) is required to maintain the proper slag basicity. We obtained energy and material balances for *pilot plant runs* in which forest residues and CDDB (100%), as well as combinations of coal with both feedstocks, was gasified from confidential test reports (Darr et al. 2008; R. B. Williams 2007). In addition, plasma heat and material balances for *commercial systems* based on computer model runs were provided (AlterNRG 2009; SNC-Lavalin 2008).

Table 4-1 Various gasifier feedstocks investigated.

Feedstock mix	Name	Type of Data
Forest biomass (100%)	I-Bio	Pilot plant
CDDB (100%)	2-CDDB	Pilot plant
Forest biomass (44%) / CDDB (56%)	5-Bio/CDDB	Pilot plant
CDDB (65%) / Forest Biomass (35%)	7-CDDB/Bio*	Computer simulation
Coal (38%) / Forest biomass (62%)	3-Coal/Bio	Pilot plant
Coal (33%) / CDDB (67%)	4-Coal/CDDB	Pilot plant
Coal (53%) / Forest Biomass (47%)	6-Coal/Bio*	Computer simulation

Percentages indicate percentage by mass of the feedstock. \*PGR based on computer simulation data.

The pilot plant is only about 1/5 the size of a commercial PGR with higher heat losses due to a limited amount of refractory lining and water wall cooling system. This is reflected in higher coke consumption and lower overall conversion efficiencies. Torch power inputs equal about 2.8% (as percentage of total energy input), while electricity requirements for feed processing (conveyor) and other auxiliary equipment are 0.017 kWh/kg feed (Burak 2010) and 0.100 kWh/kg feed<sup>52</sup> (Ducharme 2010), respectively. The PGR produces raw syngas (sent to subsequent syngas cleaning), heat (recovered via HRSG and used internally in the gasifier), slag (co-product), and small amounts of ash (landfilled). According to plant operators, slag produced can be used as roadbed aggregate or alternative daily cover in landfills. However, due to a lack of detailed data we assume that the beneficial use opportunities between slag co-produced are similar for

<sup>&</sup>lt;sup>52</sup> Includes electricity requirements to power an air separation unit for oxygen provision.

the different feedstocks and they are therefore removed from the assessment. Average transport distances to the NE region are 580 miles (coal from Pittsburgh), 350 miles (coke/anthracite from Pennsylvania), and 150 miles (limestone/sandstone).

#### C.2.4. Syngas Cleanup

Syngas cleanup consists of particulate removal using bag filters to remove fly ash from the raw syngas and two primary water scrubbers using water sprays to quench the syngas, condense particulate aerosols and help to remove fine solids and trace components not captured in the bag filter. Two final polishing wet scrubbers further condense aerosols and capture any residual acid gases, filterable and condensable particulate not captured in the primary system. It is assumed that due to quenching of the high-temperature syngas, roughly 0.400 kWh per kg waste are lost (Ducharme 2010). Solids removed are led into the slag handling system. Wastewater (0.031 kg per kg syngas) is discharged to a sump and treated in a conventional wastewater treatment plant. Mercury present in the feedstock is vaporized in the PGR. Activated carbon filters consisting of two static carbon filter beds in series are used to remove mercury from the syngas. Carbon filters, once mercury saturated, are disposed in a regulated hazardous waste landfill. Mercury emission rates come from respective test reports. Sulfur in the feedstock is mainly converted to hydrogen sulfide (H2S) during gasification. H2S is removed from the syngas stream via bio-desulphurization using the Shell Paques technology (Burak 2010; Greenhouse Gas Technology Center 2004a; b). Sodium hydroxide inputs equal roughly 0.020 kg and water inputs 0.610 kg per kg syngas. About 99.8% of all  $H_2S$  present in the raw syngas is removed during this step and elemental sulfur generated (Burak 2010). No specific data on the type of beneficial use of elemental sulfur was available and hence the co-product excluded from the assessment.

## C.2.5. Power Generation

Power generation takes place by combusting 90% cleaned syngas together with 10% No. 6 oil (by heat input) in a boiler, with steam generated powering a steam turbine for electricity production (Little and R. Green 2006). The boiler/steam turbine operates at an efficiency, expressed as useful energy output divided by total energy input, of 34.8%. Gross electricity generated at the steam turbine is used to supply internal power requirements (torch power, auxiliary equipment) first with excess available for external sale (net electricity). During the combustion of syngas and fuel oil in the boiler, flue gas is generated which is discharged via the stack of the power plant. Fossil and biogenic  $CO_2$  emissions associated with the generation of 1 kWh of electricity are calculated based on (Gillenwater 2005) using the following equation:

$$E = A_{f,m_i} \cdot F_{c,m} \cdot F_{ox} \cdot \left(\frac{44}{12}\right),$$

Where E = Mass emissions of CO<sub>2</sub> (kg),  $A_{f,m} = Mass$  of fuel consumed (kg),  $F_{c,m}$ = Carbon content of fuel on a mass basis (kg C/kg feedstock),  $F_{ox} = Oxidation$  factor to account for the fraction of carbon in the fuel that remains as soot or ash, and (44/12) = Ratio of the molecular weight of CO<sub>2</sub> to that of carbon. Fossil CO<sub>2</sub> emissions from coal (0.76 kg C/kg) (Darr et al. 2008), coke/anthracite (0.58-0.70 kg C/kg) (EPA 2007), and No.6 oil (0.85 kgC/kg) (EPA 2010b) were calculated assuming oxidation factors of 1. Biogenic  $CO_2$  is based on carbon contents from (Darr et al. 2008; Willis and Harris 2009). Syngas combustion leads to further air emissions including small amounts of particulate matter, hydrogen chloride, hydrogen fluoride, sulfur dioxide, and nitrogen oxides. Due to a lack of detailed emissions data it was decided to complement the LCI with data for the Plasco gasification process (Ducharme 2010).

#### C.2.6. System Expansion

Organic waste landfilled partially degrades under anaerobic conditions of a landfill and forms methane (a potent greenhouse gas) as well as leachate. Potential environmental benefits of waste wood diversion from landfills are modeled using data on typical waste wood disposal from (Doka 2009). According to this data about 0.065 kg  $CO_2$ -eq per kg wood are avoided if waste wood is diverted to other uses (Ecoinvent 2010). This is due to the low overall degradability (0 – 3.2%) of wood waste during 100 years (Doka 2009).

# C.2.7. Biogenic Carbon Accounting

In recent years, the carbon neutrality presumption of biomass feedstock in LCA has been challenged (E. Johnson 2009; Searchinger et al. 2009) as indirect emissions of land use change (Bringezu, Schütz, O'Brien, et al. 2009) and the dynamics of forest carbon flows over time (McKechnie et al. 2011; Walker 2010) are receiving increased

attention. For this study, collecting forest residues for bioenergy production results in short-term emissions of carbon stored in the feedstock as compared to long-term decomposition in the forest. The difference between current practices (decay of residuals left in the forest) and feedstock diversion to the gasification plant (syngas combustion) is the time-frame over which these emissions occur. Ideally this would be included using a forest-carbon model such as FORCARB2 (L. S. Heath et al. 2010). However, this is outside of the scope of the current assessment. Instead we give *implicit sequestration* credits, presuming a net flux of biogenic carbon of zero but report biogenic carbon stock changes (due to harvest) together with global warming potential (GWP) results. Similarly, CDDB is assumed to enter the plant without any prior environmental burdens and biogenic  $CO_2$  emissions are hence excluded. This approach can be justified as CDDB gasified is not reducing carbon stocks e.g. in a forest or on agricultural land (the initial reduction in carbon stocks is fully allocated to the waste's previous life).

#### **D. Results and Discussion**

#### **D.1. Power Plant Performance**

The net electricity generated per kg of feedstock is compared to literature data (*Appendix: Table 4-3*). Feedstock energy of woody biomass and coal used in this study is with 14 – 23 MJ/kg higher than typical energy contents of MSW. With the exception of route 6-Coal/Bio, electricity consumption (0.17 to 0.28 kWh/kg feed), syngas chemical

energy (8 to 14 MJ/kg feed) and net electricity generated (0.51 to 0.90 kWh/kg feed) is fairly similar to data reported elsewhere (Ducharme 2010; Herva et al. 2010; Young 2010). Differences for route 6 are due to high feedstock energy in combination with higher electrical conversion efficiencies (29%) of an anticipated commercial gasifier (using PGR simulation data). Electrical conversion efficiencies of pilot plants ranged between 13 – 20% (HHV), while for anticipated commercial plants this would be 25 – 29%. For regular solid waste gasification steam cycle plants the maximum net electrical efficiency is about 23% (Belgiorno et al. 2003). However, modified turbine designs such as integrated gasification combined cycle (IGCC) allow net electrical efficiencies of up to 30 - 40% if thorough syngas cleanup is carried out prior to IGGC (Belgiorno et al. 2003). This may be possible in a future design (see *sensitivity analysis*).

#### **D.2. Life Cycle Impact Assessment (LCIA)**

The LCIA was carried out using a combination of LCIA methods to evaluate midpoint impacts (

Table 4-2) including global warming potential (GWP) (IPCC 2007), fossil depletion, land occupation, ozone depletion (ODP), water use (ReCiPe World (H/A) v1.05 (Goedkoop et al. 2009)), and acidification, eutrophication, smog, and respiratory effects (TRACI v3.03 (Bare et al. 2002)). Single score endpoint impacts were assessed using the ReCiPe World (H/A) v1.05 method (*Appendix: Figure 4-22*). Detailed information on each impact category and the sensitivity analysis can be found in the *Appendix*.

				CDDB a	nd Forest Bio	omass	Coal and Forest			Conventional Power		
						_	B10	mass/CDDB-	rite da queda alcate		eneration-	
lı ca	npact itegory	Unit per kWh	1-Bio	2-CDDB	5- Bio/CDD B	7- Bio/CDDB *	3- Coal/Bio	4- Coal/CD DB	6- Coal/Bio*	U.S. NE mix	U.S. average mix	Hard coal power plant
G	WP	kg CO2 eq	5.137E-01	6.526E-01	5.726E-01	3.253E-01	1.902E+0 0	1.461E+0 0	1.088E+00	5.337E-01	7.754E-01	1.196E+00
GV (bi	WP biogenic)	kg CO2 eq	1.928E+0 0	2.318E+0 0	1.582E+0 0	1.083E+00	9.595E-01	7.033E-01	4.571E-01	2.709E-03	1.216E-02	4.429E-04
F d	ossil epletion	kg oil eq	1.804E-01	2.617E-01	1.841E-01	1.096E-01	5.230E-01	4.105E-01	2.951E-01	1.918E-01	2.164E-01	3.022E-01
L 01	and ccupation	m2a	5.720E+0 0	-6.837E- 04	2.807E+0 0	1.852E+00	3.320E+0 0	1.254E-02	1.583E+00	5.012E-03	1.571E-02	2.750E-02
0	DP	kg CFC- 11 eq	1.997E-08	1.720E-08	1.864E-08	1.490E-08	2.286E-08	1.694E-08	1.603E-08	1.405E-08	2.030E-08	5.313E-09
W	ater use	m3	3.201E-03	4.705E-03	3.880E-03	2.097E-03	4.250E-03	2.959E-03	1.696E-03	4.188E-04	2.149E-03	2.586E-03
Acid n Eutr catio Smo Resp effec	cidificatio	H+ moles eq	1.230E-01	1.898E-01	1.179E-01	6.947E-02	1.424E-01	1.093E-01	6.478E-02	2.128E-01	2.747E-01	3.935E-01
	utrophi- Ition	kg N eq	3.443E-04	-1.272E- 02	-5.332E- 03	-4.218E-03	2.805E-04	-4.859E- 03	1.458E-04	7.958E-05	1.426E-04	2.190E-04
	mog	g NOx eq	1.129E-03	1.293E-03	1.079E-03	6.662E-04	1.640E-03	1.163E-03	8.429E-04	8.050E-04	1.486E-03	2.572E-03
	espiratory Tects	kg PM2.5 eq	4.855E-04	7.776E-04	3.706E-04	2.239E-04	4.671E-04	3.613E-04	2.248E-04	1.000E-03	1.450E-03	2.151E-03
*		1		1 ( ) 1	4							

Table 4-2 LCIA results (per kWh) of plasma gasification routes utilizing various feedstocks and in comparison to conventional power generation.

\*PGR based on computer simulation data.

Results indicate that with regards to GWP ( $0.325 - 0.653 \text{ kg CO}_2\text{-eq/kWh}$ ), fossil depletion (0.110 - 0.262 kg oil-eq/kWh), acidification (6.95E-02 - 1.90E-01 H+ moles-eq/kWh), eutrophication (-1.27E-02 - 3.44E-02 kg N-eq/kWh), smog ( $6.66E-04 - 1.29E-03 \text{ kg NO}_x\text{-eq/kWh}$ ), and respiratory effects ( $2.24E-04 - 7.78E-04 \text{ kg PM}_{2.5}\text{-eq/kWh}$ ), the use of CDDB and forest residues as gasification feedstock for renewable electricity may result in environmental burdens slightly lower than electricity obtained from the NE or U.S. average power grid (

Table 4-2). Impacts are mainly due to current system configurations in which coke is used as gasifier bed material, fuel oil is co-combusted in the boiler, and extensive syngas cleanup is applied (see *Appendix*). On-site emissions are minimal due to the nature of the plasma gasifier which removes a large fraction of inorganics present in the waste feedstock as vitrified slag and applies extensive syngas cleanup prior to combustion, therefore reducing gas volume to be cleaned.

Co-gasification with coal (routes 3, 4, and 6) significantly increases impacts, in particular to GWP (1.08 to 1.90 kg  $CO_2$ -eq) and fossil depletion (0.295 to 0.52 kg oil-eq), due to coal acquisition and fossil feedstock carbon emitted on-site during syngas combustion. Applying carbon-capture-and-storage (CCS) or -reuse (CCR) may be capable of reducing system-wide GWP and should be investigated in a future study. Coal inputs range between 0.28 kg (route 6) to 0.43 kg (route 3) per kWh electricity generated. This compares to roughly 0.47 kg of coal per kWh for traditional coal-fired power plants (Ecoinvent 2010). In contrast to traditional combustion and gasification systems solely utilizing coal feedstock, the use of waste in a gasification-steam cycle boiler limits the overall plant electrical efficiency due to impurities present in the waste feedstock that have the potential to form sour gases (e.g. HCl,  $H_2S$ , etc.) at high temperatures potentially corroding tubes (Belgiorno et al. 2003).

The avoided burdens of diverting CDDB from landfills are captured via system expansion and reduce overall impacts. This is particularly pronounced for eutrophication which is drastically reduced if CDDB is diverted from landfills, mainly as a result of avoided nutrient leaching.

It should be noted that with a GWP of 0.325 to 0.653 kg CO<sub>2</sub>-eq per kWh (*Appendix: Figure 4-5*), biomass/CDDB gasification for electricity production results in much higher GHG emissions than data reported for other bio-power LCA studies. (G. Heath et al. 2011) reported a GWP meta-analysis of various bio-power systems (co-firing, combustion, gasification) and found 25<sup>th</sup> and 75<sup>th</sup> percentile ranges of life-cycle GHG emissions of 0.015 to 0.065 kg CO<sub>2</sub>-eq/kWh, respectively. Higher GWPs found in this study are primarily due to the current plasma gasification system configuration using fossil coke or anthracite as PGR bed material, oil co-combustion in the boiler to allow for a stable flame, and extensive cleanup using NaOH and other chemicals. This system configuration should not be seen as rigid since modified PGR systems may be able to operate with smaller amounts of coke, syngas cleanup may be further optimized in the future, and a different boiler design could allow the combustion of syngas without the need for fossil-based oil. In addition, electrical conversion efficiencies might be further improved using advanced turbine designs (see sensitivity analysis).
Land occupation (-6.84E-04 to 5.72E+00 m2a) is highest if forest residues are used as gasifier feedstock and lowest when CDDB is utilized (*Appendix: Figure 4-10*). This is due to upstream impacts associated with forest growth and harvest in the NE U.S. However, due to climatic conditions and natural regeneration assumed, land occupation results are region-specific for the NE U.S. Utilizing other biomass feedstock (e.g. short rotation crops on formerly degraded land) may be able to provide a suitable feedstock associated with lower land occupation.

Water use associated with gasification plants (1.70E-03 to 4.7E-03 m<sup>3</sup>/kWh) is higher than for conventional fossil-based systems (4.2E-04 to 2.6E-3 m<sup>3</sup>/kWh) due to extensive syngas cleanup (*Appendix: Figure 4-14*). This may be an obstacle for implementing those technologies in arid regions around the world.

Ozone depletion (ODP) (1.49E-08 – 2.28E-08 kg CFC-11-eq), associated with plasma gasification was found to be similar to NE and U.S. average power (1.41E-08 and 2.03E-08 kg CFC-11-eq) but higher than coal-fired power generation (0.53E-08 kg CFC-11-eq) (*Appendix: Figure 4-12*). ODP is mainly due to the unit processes of heavy fuel oil production for no. 6 oil provision co-combusted in the boiler, and sodium hydroxide provision for syngas cleaning (*Appendix: Figure 4-13*).

Using computer simulation data for the PGR (routes 6 and 7) shows how, in a full-scale commercial facility, environmental burdens may be further reduced due to lower heat losses associated with feedstock gasification. In such a case, utilizing bio-based feedstock in the plasma gasification system (route 7) may have the potential to lead to environmental burdens lower than U.S. average power in all impact categories, except

land occupation which due to forest residuals use is higher than for fossil-based power generation. Similarly, in a commercial plant coal used as co-gasification feedstock together with forest residuals (route 6) may allow energy provision that could compete with current coal-fired power plants in all impact categories excluding land occupation and ODP.

Finally, environmental impacts are compared using the *ReCiPe Endpoint method* (Goedkoop et al. 2009) to allow an easier comparison between the gasification routes investigated (*Appendix: Figure 4-22*). Results show that environmental impacts of electricity generated using forest residuals/CDDB-based (routes 1, 2, 5, and 7) are within the range of conventional electricity production. Lowest impacts are found for route 7-Bio/CDDB, envisioning a commercial plasma gasification reactor with higher electrical efficiencies than the pilot plant. A commercial plant may be capable to compete with electricity provided by the NE power grid in terms of its environmental impact per kWh. In contrast, using coal as co-gasification feed in a pilot PGR (routes 4 and 5) leads to higher impacts when compared to conventional energy provision including coal-fired power plants. Even if a commercial PGR is envisioned (route 6), environmental impacts are still within the range of current coal-fired power plants and significantly higher when compared to the NE energy mix (which uses large shares of natural gas and nuclear power) as well as the U.S. average power mix.

In general, environmental impact single scores are found to be lowest when CDDB is used as (co-) gasification feed (routes 2, 4, 5, and 7). This is mainly due to lower land occupation (when compared to forest residue use) and avoided landfilling, both resulting in less potential damage to ecosystems and preventing negative environmental burdens associated with conventional wood waste landfilling. However, depending on the *weighting set* chosen different results may be obtained. For the ReCiPe World H/A model a weighting set of 40% human health, 40% ecosystems, and 20% resources is used. Using the mixing triangle developed by Hofstetter et al (Hofstetter et al. 1999) shows that assuming different weighting factors, in which e.g. resource depletion is considered more important, could change results in favor of forest residuals (*Appendix: Figure 4-23*).

#### **D.3. Sensitivity Analysis**

Environmental impact categories investigated were found to be sensitive to varying assumptions with regards to coke/anthracite inputs, torch power, and turbine efficiencies (*Appendix: Figure 4-24, Table 4-7, Table 4-8*). Halving coke/anthracite inputs used as PGR bed material may result in a recognizable decrease in environmental burdens in particular with regards to fossil depletion potential (FD) (~30% reduction), respiratory effects and acidification (both ~20% reduction), as well as GWP and smog (both ~10% reduction). The lowest impacts are found if an increase in the turbine efficiency of up to 50% is assumed (this may be possible via IGCC, assuming thorough syngas cleanup prior to syngas being fed to the gas turbine). Assuming such a design, overall environmental impacts could be reduced by roughly 30% compared to current configurations. Applying economic allocation to the multi-output processes of 'waste sorting' and 'wood growth/harvest' would reduce upstream environmental burdens associated with these

processes as low-value feedstock (i.e. forest residuals currently left in the forest to decay and C&D wood currently landfilled) are utilized (*Appendix: Figure 4-25*). As a result, impacts with regards to land occupation might be drastically reduced for all routes utilizing forest residuals under economic allocation.

#### **E.Outlook and Recommendations**

Using forest residues and CDDB as feedstock for plasma gasification allows the generation of a syngas suitable for electricity production. Using CDDB as gasifier feedstock may represent a safe disposal alternative to current landfilling practices, would reduce the amount of waste landfilled, and decrease land occupation. However, in the current system configuration, life-cycle impacts of plasma gasification are largely influenced by the inputs of coke, anthracite (both gasifier bed material), and no. 6 oil (boiler), as well as water and chemicals inputs to the syngas cleanup system. In addition, low conversion efficiencies due to PGR heat losses and the use of a steam turbine, instead of more efficient gas turbine, limit the system-wide environmental performance. The gasification of other mixed waste feedstocks (e.g. MSW and industrial wastes) should be investigated in future studies. It should be noted that on-site emissions have been derived using a combination of existing data from our company partner and industry data from the literature on typical onsite emissions of MSW plasma gasification. These should therefore only be seen as a proxy of actual emissions which will vary depending on feedstock type, season, and syngas cleanup configuration. While plasma gasification seems to be competitive, in terms of environmental impacts, to fossil-based energy

production, in order to compete with other bio-based energy systems (G. Heath et al. 2011) plasma gasification has to significantly reduce GHG emissions, particularly associated with fossil inputs and syngas combustion. Capturing carbon (CCS or CCR) from the gas stream prior to combustion offers advantages (Herzog and Golomb 2004) and could help to reduce GWP-related impacts in future designs. However, the provision of a clean syngas also opens up future options of syngas utilization as feedstock for subsequent chemicals and fuels provision via various catalytic pathways (e.g. Fischer-Tropsch or Methanol-to-Olefins Synthesis) (Nuss et al. 2012). Plasma gasification should therefore also be investigated in coupled systems using subsequent syngas catalytic conversion into chemical feedstock and in comparison to fossil-based fuels and chemical feedstock provision.

## APPENDIX 53

# LIFE CYCLE ASSESSMENT (LCA) OF CONSTRUCTION AND DEMOLITION (C&D) DERIVED BIOMASS GASIFICATION FOR ELECTRICITY PRODUCTION

#### A.Abstract

The supporting information includes detailed information on process performance and the impact categories of global warming potential (GWP), fossil depletion, land occupation, ozone depletion potential (ODP), water use, acidification, eutrophication, smog, and respiratory effects carried out as part of the life-cycle impacts assessment (LCIA). In addition, information on weighting within ReCiPe (Goedkoop et al. 2009) is provided and result on the sensitivity analysis given.

## **B.** Power Plant Performance

The net electricity generated per kg of feedstock is compared to literature data (Table 4-3).

<sup>&</sup>lt;sup>53</sup> The appendix has been submitted as supporting information with the paper: Nuss, P., Gardner, K.H., and Jambeck, J.R. (In review). "Life Cycle Assessment of Construction and Demolition (C&D) Derived Biomass Gasification for Electricity production." *Environ. Sci. Technol.* 

Feedstock	Feedstock energy [MJ/kg]	Electricity consumption [kWh/kg feed]	Syngas [MJ/kg feed]	Net electricity [kWh/kg feed]	Source	HHV Conversion efficiency [%]	Comments
1-Bio	16.54	0.22	11.83	0.90	Our study	19.69	
2-CDDB	13.68	0.20	7.92	0.51	Our study	13.31	Steam turbine, 34.8%
5-Bio/CDDB	16.52	0.22	9.79	0.68	Our study	14.76	conv. eff.
7-CDDB/Bio*	12.84	0.17	9.69	0.90	Our study	25.36	
3-Coal/Bio	23.18	0.28	11.51	0.82	Our study	12.71	
4-Coal/CDDB	19.79	0.22	14.22	0.90	Our study	16.33	steam turbine, 34.8% conv. eff.
6-Coal/Bio*	23.25	0.25	21.03	1.85	Our study	28.70	
RDF	-	1.06	14.46	-	(Herva et al. 2010)	-	-
RDF	-	0.58	-	0.90	(Herva et al. 2010)	-	-
Wood	-	1.00	15.28	0.93	(Herva et al. 2010)	-	-
Wood	-	0.33	-	-	(Herva et al. 2010)	-	-
MSW	10.08	0.26	5.82	0.45	(Ducharme 2010)	16.07	InEn Tec; Gas turbine, 50% conv. eff.
MSW/coke	11.29	0.32	8.10	0.62	(Ducharme 2010)	19.67	AlterNRG; Gas turbine, 50% conv. eff.
MSW/tires	11.13	0.42	8.01	0.50	(Ducharme 2010)	16.18	Europlasma, Gas turbine, 50% conv. eff.
MSW	10.08	0.28	-	0.53	(Ducharme 2010)	18.93	Plasco, Gas turbine, 50% conv. eff.
MSW	11.34	-	•	0.82	(Young 2010)	25.90	-

Table 4-3 Performance comparison of the plasma gasification energy production routes investigated.

\*Anticipated commercial Plasma Gasification Reactor (PGR) performance based on computer simulation results. HHV: Higher Heating Value.

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#### C. Midpoint Impact Categories

The life cycle impact assessment was carried out using a combination of commonly used LCIA methods, including IPCC  $2007^{54}$  (IPCC 2007), TRACI v3.03<sup>55</sup> (Bare et al. 2002), and the ReCiPe World (H/A) v1.05<sup>56</sup> (Goedkoop et al. 2009) method, to evaluate the potential impacts of global warming potential (GWP), fossil depletion, land occupation, ozone depletion, water use, and the potential contributions of elementary flows to acidification, eutrophication, smog, and respiratory effects.

<u>Global warming potential (GWP)</u> indicates the amount of greenhouse gases emitted over the course of the life cycle. Results are expressed relative to carbon dioxide in  $kg CO_2$ -eq. We use characterization factors from the IPCC for a 100-year timeframe (IPCC 2007).

<u>Fossil depletion</u> captures fossil raw materials used throughout the life cycle. The midpoint characterization factor is based on the higher heating value of all non-renewable resources used and is expressed in *kg oil-eq*, relative to crude oil feedstock with an energy content of 42 MJ per kg (Goedkoop et al. 2009).

Land occupation: is accounted for using the ReCiPe characterization factors (Goedkoop et al. 2009) for forest land areas (termed agricultural land in ReCiPe). The impact category quantifies the area in m2 and year required for the provision of 1 kWh of electricity at the factory gate.

<sup>&</sup>lt;sup>54</sup> Intergovernmental Panel on Climate Change (IPCC)

<sup>&</sup>lt;sup>55</sup> Tool for the Reduction and Assessment of Chemical and other Environmental Impacts (TRACI)

<sup>&</sup>lt;sup>56</sup> http://www.lcia-recipe.net/

<u>Ozone depletion potential (ODP)</u> is a relative measure of the ozone depletion capacity of substances emitted. The substance CFC-11 (trichlorofluoromethane) is used as a reference and hence results expressed as kg CFC-11-eq (Goedkoop et al. 2009).

Water use: quantifies the total freshwater consumed by the conversion systems. It is expressed as the volume of water consumed (m3) and based on the characterization factors of the ReciPe (H) v1.04 method (Goedkoop et al. 2009). Water use may be a limitation for the implementation of novel systems in arid regions and developing countries.

Acidification, eutrophication, smog, and respiratory effects: were accounted for using TRACI, a stand-alone computer program developed by the U.S. Environmental Protection Agency (Bare et al. 2002). TRACI uses region-specific characterization factors for North America. Acidification impacts are expressed as H+ moles-eq, eutrophication as kg N-eq, smog as g NOx-eq, and respiratory effects in terms of kg PM2.5-eq.

An overview of system-wide environmental impacts for the seven different plasma gasification routes utilizing either biomass feedstock and CDDB, or combinations of coal and biomass/CDDB are shown in Figure 4-3 and Figure 4-4. The functional unit in all comparisons in 1 kWh electricity generated at the factory gate.

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Figure 4-3 Relative environmental burdens of plasma gasification routes utilizing <u>forest</u> <u>biomass and/or CDDB</u> and in comparison to conventional power mixes. The functional unit is 1 kWh of electricity at the factory gate. \*Gasification data for routes 1, 2 and 5 comes from a pilot plant while route 7 represents data from computer simulations. \*\*Land occupation includes forest and urban land occupation.



Figure 4-4 Relative system-wide environmental burdens of plasma gasification routes utilizing <u>coal with forest biomass and/or CDDB</u> and in comparison to conventional power mixes. The functional unit is 1 kWh of electricity at the factory gate. \*Gasification data for routes 3 and 4 comes from a pilot plant while route 6 represents data from computer simulations. \*\*Land occupation includes forest and urban land occupation.

## C.1. Global Warming Potential (GWP)

Impacts to global warming are shown in Figure 4-5. The functional unit for comparison is 1 kWh of electricity at the factory gate.



Figure 4-5 Global warming potential (GWP) associated with plasma gasification and conventional electricity production. The functional unit for comparison is 1 kWh of electricity at the factory gate. The dotted lines indicate groups of gasifier feedstocks (i.e. CDDB/biomass and coal/CDDB/biomass) vs. conventional power generation. \*Approximation of commercial gasification reactor.

The figure shows the contributions of different unit processes of the foreground system to GWP. GHG emissions range between 0.33 and 1.90 kg CO<sub>2</sub>-eq per kWh of electricity generated for plasma gasification and 0.53 to 1.20 kg CO<sub>2</sub>-eq for conventional electricity generation. As expected, the LCI models using inventory data for commercial plasma gasification reactors (PGRs) (i.e. route 6 and 7) lead to the lowest GWP. This is due to a higher HHV conversion efficiency as heat loss is significantly smaller than in the pilot plant reactor and therefore more of the feedstock energy content is available as syngas chemical energy for subsequent power generation. For all plasma gasification routes, carbon stored in fossil feedstock, including coal, anthracite, coke, as well as no.6

oil, is co-combusted in the steam boiler and thereby emitted back to the atmosphere contributing to GWP. Fossil CO<sub>2</sub> emissions are highest for routes 3, 4 and 6 since coal is co-gasified and hence a significant fraction of carbon present in the syngas is of fossil origin only. For coal co-gasification, GWP from syngas combustion is due to fossil CO<sub>2</sub> from coal (78%), coke (15%), and no. 6 fuel (7%). Impacts from gasification ('raw syngas') are mainly due to inputs of anthracite and coke (gasifier bed materials) as well as feedstock transport and up-stream burdens associated with coal mining and processing, fuel production, etc. GWP from syngas cleaning is to a large extent due to sodium hydroxide inputs used for H<sub>2</sub>S removal from the raw syngas. On the other hand, avoided landfilling leads to a small reduction of overall environmental burdens (indicated as negative burdens) for routes in which CDDB is used as (co-)gasification feedstock. However, due to the fact that wood waste landfilled only partially degrades under anaerobic conditions, methane emissions are rather small (0.065 kg CO2-eq are avoided per kg CDDB) and hence reductions in GWP minimal.

When compared to conventional electricity generation, plasma gasification of biogenic feedstock seems beneficial in comparison to the U.S. average power grid and coal-fired power plants. Only route 7, representing a commercial PGR, scores with 0.325 kg CO<sub>2</sub>-eq/kWh lower than an equivalent amount of energy provided from the Northeastern (NE) grid (0.534 kg CO<sub>2</sub>-eq). Utilizing coal as co-gasification feed leads to a higher GWP than conventional power generation. An exception is route 6 which scores with 1.09 kg CO<sub>2</sub>-eq/kWh slightly below the 1.20 kg CO<sub>2</sub>-eq/kWh found for coal-fired power plants. Figure 4-6 shows a Sankey diagram for the production of 1 kWh (3.6 MJ)

of electricity from CDDB (route 2). Only processes contributing 3.3% or more to the overall GWP are shown (cut-off criterion).



Figure 4-6 Sankey diagram showing the GWP [kg  $CO_2$ -eq] associated with the generation of 1kWh electricity from CDDB (Route 2). Only processes contributing 3.36% or more to GWP are shown. The most carbon-intensive processes in terms of fossil GWP are the combustion of syngas in the boiler/steam turbine during which fossil carbon from anthracite bed material and no. 6 oil co-combusted is emitted to the atmosphere. This is followed by the production of sodium hydroxide used during gas cleaning, and anthracite used as bed material in the gasifier. Avoided CDDB landfilling reduces GWP by roughly 18.3 % to 0.653 kg  $CO_2$ -eq/kWh.

Emissions of *biogenic CO2* are mainly a result of forest residues and CDDB combustion in the boiler. Carbon present in primary residues utilized in the plasma gasifier is emitted back to the atmosphere faster than it would have if the biomass feedstock were left in the forest to decay naturally. Carbon present in the CDDB feedstock is assumed not to lead to carbon stock changes in the natural environment. However, if C&D waste wood, e.g. originating from construction, is seen to constitute a carbon-stock decrease, the biogenic carbon emissions would need to be accounted for. From a policy-makers perspective CO2 currently emitted to the atmosphere might be of greatest interest as in the short term both biogenic and fossil carbon contribute to GWP. Since differences in opinion with regards to carbon accounting will lead to different outcomes we simply show the impact of counting biogenic CO2 towards GWP (Figure 4-7 and Table 4-4).



Figure 4-7. Global warming potential of the various plasma gasification routes for electricity generation. The figure shows that biogenic carbon dioxide emissions lead to significantly higher GWP associated with the plasma gasification routes utilizing biogenic feedstock (CDDB and forest residuals). \*Commercial gasification reactor.

Table 4-4 Global warming potential of the various plasma gasification routes for electricity generation.

Impact category [kg CO2-eq/kWh]	1-Bio	2- CDDB	5-Bio/ CDDB	7-Bio/ CDDB*	3- Coal/ Bio	4- Coal/ CDDB	6- Coal/ Bio*	U.S. NE mix	U.S. averag e mix	Hard coal mix
Fossil CO <sub>2</sub>	0.514	0.653	0.573	0.325	1.902	1.461	1.088	0.534	0.775	1.196
Biogenic CO <sub>2</sub>	1.928	2.318	1.582	1.083	0.960	0.703	0.457	0.003	0.012	0.000
<b>GWP TOTAL</b>	2.442	2.971	2.154	1.409	2.862	2.164	1.545	0.536	0.788	1.196

\*Commercial gasification reactor.

The results show that accounting for biogenic carbon leads to plasma gasification routes having much higher impacts to GWP than their conventional counterparts.

#### **C.2. Fossil Depletion Potential**

Figure 4-8 shows the fossil depletion potential associated with plasma gasification and conventional electricity production.



Figure 4-8 Fossil depletion potential associated with plasma gasification and conventional electricity production. The functional unit for comparison is 1 kWh of electricity at the factory gate. The dotted lines indicate groups of gasifier feedstocks (i.e. CDDB/biomass and coal/CDDB/biomass) vs. conventional power generation. \*Approximation of commercial gasification reactor.

As shown in the figure, the bulk of fossil depletion potential (FD) is due to 'raw syngas' generation during which fossil raw materials including coal, coke, and anthracite are utilized. Besides fossil raw material acquisition for the provision of feedstock and bed material, transportation fuel for feedstock shipping to the gasification plant (located in the NE US) contributes to FD. Impacts from syngas cleaning are mainly due to chemicals (e.g. sodium hydroxide) used for pollutant removals. The use of no. 6 oil during the 'syngas combustion' step results in additional contributions to FD. During the life-cycles using CDDB as feedstock, waste diversion avoids fossil raw materials use associated with landfills operation. However, due to the small amounts of CDDB diverted the effect on total FD is small.

The use of CDDB and/or biomass leads to FD impacts of roughly 0.110 to 0.262 kg oil-eq per kWh, while the use of combinations of coal with biogenic feedstock requires between 0.295 to 0.523 kg oil-eq per kWh electricity at the factory gate. Similar to previous results, the use of commercial PGR data reduces the impact in comparison to pilot plant runs. When compared to conventional power generation, the use of CDDB and/or biomass may have the potential to result in FD impacts similar to current fossil-based power production. However, using coal as co-gasification feed results in impacts similar to or higher than coal-fired power plants.

Finally, Figure 4-9 shows unit processes contributing most significantly to FD of the route 3 life-cycle utilizing coal and forest residues as an example.



Figure 4-9 Inventory contributions to the fossil depletion potential of 1 kWh of electricity via route 3 (Coal/Bio). Coal used in the gasifier contributes the bulk to the environmental impacts to FD.

## **C.3. Land Occupation**

Land occupation due to land requirements for feedstock growth and harvest

(forest residues), infrastructure for plants, roads etc. is shown in Figure 4-10.



Figure 4-10 Land occupation (forest and urban) associated with plasma gasification and conventional electricity production. The functional unit for comparison is 1 kWh of electricity at the factory gate. The dotted lines indicate groups of gasifier feedstocks (i.e. CDDB/biomass and coal/CDDB/biomass) vs. conventional power generation. \*Approximation of commercial gasification reactor.

The use of forest residues as gasification feedstock leads to land occupation in the range of 1.58 to 5.72 m2a per kWh, while this is avoided if CDDB is utilized. Land occupation for routes 2 and 4 (no forest residue use) is similar to or less than land occupied for conventional energy provision. Negative impacts to land occupation are due to avoided landfill space. Figure 4-11 shows the unit processes contributing to land occupation when only forest residues are used as feedstock for plasma gasification.



Figure 4-11 Sankey diagram showing land occupation (forest and urban) [m2a] associated with the generation of 1kWh electricity from forest residuals (Route 1). Only processes contributing 0.5% or more are shown. The growth and harvest of forest biomass (softwood and hardwood) contributes more than 99% to total land occupation. The figure also shows that in comparison to forest land occupation the infrastructure requirements e.g. due to roads, plants, etc. is almost negligible.

#### **C.4. Ozone Depletion Potential (ODP)**



Figure 4-12 Ozone depletion potential (ODP) associated with plasma gasification and conventional electricity production. The functional unit for comparison is 1 kWh of electricity at the factory gate. The dotted lines indicate groups of gasifier feedstocks (i.e. CDDB/biomass and coal/CDDB/biomass) vs. conventional power generation. \*Approximation of commercial gasification reactor.

Ozone depletion potential (ODP) is due to chlorofluorocarbons (CFCs) and halons released from unit processes such as heavy fuel oil production for no. 6 oil co-combusted in the boiler, and sodium hydroxide production for syngas cleaning. In addition, the use of diesel fuel in a mobile chopper for wood chips production from forest residues contributes to ODP. Avoided landfilling reduces ODP for routes, 2, 4, 5, and 7. ODP was found to be relatively similar for the different life cycles. Figure 4-13 shows the Sankey diagram for the route 2 life-cycle.



Figure 4-13 Sankey diagram showing ODP [kg CFC-11-eq] associated with the generation of 1kWh electricity from CDDB (Route 2). Only processes contributing 10% or more are shown. Most of the environmental burdens are due to heavy fuel oil production for the provision of no.6 oil. This is followed by sodium hydroxide generation. Avoided landfilling reduces ODP by roughly 23% to a total impact of 1.72E-08 kg CFC-11-eq.



Figure 4-14 Water use [m3] associated with plasma gasification and conventional electricity production. The functional unit for comparison is 1 kWh of electricity at the factory gate. The dotted lines indicate groups of gasifier feedstocks (i.e. CDDB/biomass and coal/CDDB/biomass) vs. conventional power generation. \*Approximation of commercial gasification reactor.

Water use is mainly due to water requirements during syngas cleaning, i.e. use in scrubbers and quenching, as well as for sodium hydroxide makeup. The figure also shows that forest biomass provision contributes only a small share to total water use. The reason for this is that forestry practices in the NE United States rely to a large extent on natural regeneration cycles in which artificial irrigation is not required (Oneil et al. 2010). With 2.1 to 4.7 kg water per kWh of electricity for CDDB/biomass-based routes and 1.7 to 4.3

kg/kWh for coal-based routes, energy production via plasma gasification results in slightly higher water usage than conventional electricity provision from the U.S. power grid (2.1 kg) and coal-fired power plants (2.6 kg). Power from the NE grid scores lowest with only about 0.42 kg water per kWh of electricity generated. Most of the water utilized is process water used directly at the plant for syngas cleaning. This is followed by decarbonized water used within systems up the supply chain. Figure 4-15 shows some of the unit processes contributing to water use for the route 3 life cycle utilizing coal and forest residuals as an example.



Figure 4-15 Relative contributions of unit processes to water use of the route 3 life cycle utilizing coal and forest biomass. The functional unit is 1 kWh of electricity at the factory gate. A cut-off of 1% has been applied.

## **C.6.** Acidification

Acidification is due to the emissions of e.g.  $SO_2$ ,  $SO_X$ ,  $NO_X$ ,  $NH_4^+$  and other substances during the energy-production life cycle. System-wide acidification potentials for the various systems are shown below.



Figure 4-16 Acidification potential [kg H+ moles-eq] associated with plasma gasification and conventional electricity production. The functional unit for comparison is 1 kWh of electricity at the factory gate. The dotted lines indicate groups of gasifier feedstocks (i.e. CDDB/biomass and coal/CDDB/biomass) vs. conventional power generation. \*Approximation of commercial gasification reactor.

As shown in the figure, impacts to acidification range between 0.07 and 0.19 kg  $H^+$  moles-eq for biomass/CDDB-based routes and 0.06 to 0.142 kg  $H^+$  moles-eq when combinations of coal and biomass/CDDB are used as plasma gasifier feedstock. This

compares to 0.39 kg  $H^+$  moles-eq for conventional coal-fired power plants and 0.28 and 0.21 kg  $H^+$  moles-eq for US average power and the US NE power mix, respectively. Impacts to acidification in the plasma gasification life-cycles are due to a combination of unit processes. Representative for the various life-cycles, Table 4-5 shows a summary of processes contributing most significantly to acidification of the route 2-CDDB life-cycle.

Process	Used by	Total	MRF	Land- filling	Raw Syngas	Syngas Cleaning	Syngas Combus tion
Total of all processes	-	1.90E-01	9.32E-03	-1.06E-02	9.32E-02	5.97E-02	3.82E-02
Anthracite coal, combusted in industrial boiler NREL /RNA	Gasifier bed (foreground)	7.66E-02	x	x	7.66E-02	x	x
2-Electricity, CDDB	Syngas combustion (foreground)	2.82E-02	x	x	x	x	2.82E-02
Hard coal, burned in power plant/RFC with US electricity U	US Electricity mix (background)	1.61E-02	8.44E-04	-5.18E-04	1.09E-04	1.53E-02	4.24E-04
Hard coal, burned in power plant/SERC with US electricity U	US Electricity mix (background)	1.23E-02	6.41E-04	-3.95E-04	8.34E-05	1.16E-02	3.24E-04
Natural gas, at production/RNA with US electricity U	US Electricity mix (background)	9.99E-03	5.20E-05	-3.37E-04	7.15E-05	9.93E-03	2.76E-04
Transport, single unit truck, diesel powered NREL/US	Trans- portation	9.10E-03	x	x	9.10E-03	x	x
Natural gas, processed, at plant NREL /US	US Electricity mix (background)	5.92E-03	5.16E-03	x	7.60E-04	x	x
Natural gas, sour, burned in production flare/MJ/GLO with US electricity U	US Electricity mix (background)	3.96E-03	7.22E-06	-4.62E-04	4.05E-05	4.23E-04	3.95E-03
Diesel, burned in building machine/GLO with US electricity U	Various processes (landfilling, NaOH production, etc.) (background)	-3.35E-03	1.58E-05	-4.13E-03	3.12E-04	3.19E-04	1.33E-04
Remaining processes	-	3.09E-02	2.60E-03	-4.76E-03	6.18E-03	2.21E-02	4.81E-03

Table 4-5 Process contributions to acidification of the 2-CDDB lifecycle.

All numbers are in kg H+ moles-eq and given per functional unit of 1 kWh at the factory gate.

The use of anthracite as gasifier bed material in route 2 contributes with 0.077 kg H+ moles-eq a large share (42%) to total acidification. This is due to emissions mainly of  $SO_2$  and  $NO_X$  during raw material acquisition. In addition, the use and burning of coal (and to a smaller extent natural gas) for energy provision to the US power mixes, used in a variety of background processes such as NaOH production, energy inputs to the MRF (waste sorting), landfilling, etc., contributes to acidification. Furthermore, transportation of feedstock (CDDB, anthracite, flux material) to the power plant, during which diesel is combusted, impacts acidification. Figure 4-17 shows the inventory contributions to the route 2 life-cycle. According to this, emissions of  $SO_2$  and  $NO_x$  are responsible for more than 95% of overall acidification.



Figure 4-17 Inventory contributions to acidification of the 2-CDDB lifecycle. All numbers are in kg H+ moles-eq and given per functional unit of 1 kWh at the factory gate.

#### **C.7. Eutrophication**

Impacts to eutrophication are dominated by offsets from avoided landfilling as shown in Figure 4-18. Eutrophication is due to emissions (landfill leaching) of nitrate, phosphate, ammonia, COD<sup>57</sup>, BOD<sup>58</sup> and others substances.



Figure 4-18 Eutrophication potential [kg N-eq] associated with plasma gasification and conventional electricity production. The functional unit for comparison is 1 kWh of electricity at the factory gate. The dotted lines indicate groups of gasifier feedstocks (i.e. CDDB/biomass and coal/CDDB/biomass) vs. conventional power generation. \*Approximation of commercial gasification reactor.

For all routes utilizing CDDB as feedstock eutrophication associated with anticipated and avoided landfill leaching reduces the environmental burden significantly. As a result, routes no. 2, 4, 5, and 7 (all diverting CDDB from the landfill) lead to an eutrophication reduction of between -2.81E-3 (route 3) and -12.70E-3 kg N-eq (route 2).

<sup>&</sup>lt;sup>57</sup> Chemical Oxygen Demand (COD)

<sup>&</sup>lt;sup>58</sup> Biological Oxygen Demand (BOD)

The reason for the low eutrophication associated with route 2 is that solely CDDB is used. In comparison, conventional fossil-based routes lead to slightly positive eutrophication potentials of 2.19E-4 kg N-eq for coal-fired power plants and 1.43E-4 or 7.96E-5 kg N-eq for US average power and US NE power, respectively. Landfill leaching is dependent on local conditions such as rainfall, landfill maintenance and operating conditions and will therefore vary depending on the dataset used.

## C.8. Smog

Smog potential ranges between 6.66E-4 (7-Bio/CDDB) and 1.64E-3 kg NO<sub>x</sub>-eq (3-Coal/Bio) per kWh electricity generated (see Figure 4-19). This compares to 8.05E-4 to 2.57E-3 for conventional energy production.



Figure 4-19 Smog potential [g NOx-eq] associated with plasma gasification and conventional electricity production. The functional unit for comparison is 1 kWh of electricity at the factory gate. The dotted lines indicate groups of gasifier feedstocks (i.e. CDDB/biomass and coal/CDDB/biomass) vs. conventional power generation. \*Approximation of commercial gasification reactor.

The unit processes contributing most significantly to 'raw syngas' production include coal and coke/anthracite provision as well as road transportation of the gasifier feedstocks. Impacts from 'syngas cleaning' are due to emissions associated with electricity inputs to the NaOH production process (NaOH is used for  $H_2S$  removal). Landfilling slightly reduces the smog potential during the route 2, 4, 5 and 7 life-cycles. Wood chips production requires the use of diesel fuel for loader operations, skidding and the mobile chopper all contributing to smog potential.

The only *on-site* smog emissions are due to 'syngas combustion' during which cleaned syngas is combusted in the steam boiler, thereby releasing air pollutants to the atmosphere. With the exception of route 2-CDDB (low conversion efficiency), impacts

from syngas combustion range between 6.044E-5 (4% of total smog, 3-Coal/Bio) and 2.65E-4 kg NO<sub>x</sub>-eq (25% of total smog, 5-Bio/CDDB) per kWh generated. It should be noted that on-site emissions have been derived using a combination of existing data from our company partner and industry data from the literature on typical onsite emissions of MSW plasma gasification (data for the latter comes from Plasco as shown in (Ducharme 2010)). In reality, emissions from syngas combustion will to a large extent be influenced by the:

- Type and heterogeneity of the gasifier feedstock (which itself depends on consumer habits, season, etc.)
- Design of the syngas cleanup system
- Subsequent electricity generating system (steam cycle vs. gas turbine<sup>59</sup>).

Finally, Figure 4-20 shows a Sankey diagram for route 7. It shows the different contributions of unit processes to smog and offsets associated with landfill diversion.

<sup>&</sup>lt;sup>59</sup> A gas turbine will require a much cleaner syngas than if a boiler and steam turbine is used.



Figure 4-20 Sankey diagram showing smog [kg NOx-eq] associated with the generation of 1kWh electricity from forest residuals and CDDB (Route 7). Only processes contributing 6.8% or more are shown. Most of the environmental burdens are due to wood chips production, feedstock transport, and on-site emissions during syngas combustion. Avoided landfilling reduces smog by roughly 10.3% to a total impact of 6.662E-4 kg NOx-eq. It should be noted that due to a lack of data some of the on-site emissions were derived using industry data on MSW plasma gasification as a proxy. In reality, on-site emissions will depend to a large extent on the type and heterogeneity of feedstock, syngas cleanup system and boiler system.

### **C.9. Respiratory Effects**

Finally, the last impact category investigated is respiratory effects consisting of nitrogen oxides ( $NO_x$ ), particulate matter (PM), sulfur dioxides ( $SO_2$ ) and others (see Figure 4-21).



Figure 4-21 Respiratory effects [kg PM2.5-eq] associated with plasma gasification and conventional electricity production. The functional unit for comparison is 1 kWh of electricity at the factory gate. The dotted lines indicate groups of gasifier feedstocks (i.e. CDDB/biomass and coal/CDDB/biomass) vs. conventional power generation. \*Approximation of commercial gasification reactor.

Respiratory effects were found to be smaller for the plasma gasification lifecycles when compared to conventional fossil-based energy provision systems. Impacts are due to feedstock inputs (coal, coke/anthracite, wood chips) to the gasifier (raw syngas), electricity requirements for NaOH production (syngas cleaning), and heavy fuel oil co-combusted with syngas (syngas combustion) in the boiler/steam turbine. Respiratory effects for plasma gasification routes were found to be between 2.25E-4 to 7.78E-4 kg PM2.5-eq/kWh, while those of conventional energy production ranged between 1.00E-3 and 2.15E-3 kg PM2.5-eq/kWh.

#### **D.Endpoint Impacts Weighting (ReCiPE)**

Feedstock gasified at the plasma gasification power station is compared to conventional energy provision using the ReCiPe World H/A Endpoint method (Figure 4-22). In ReCiPe, eighteen midpoint indicators are transformed into three endpoint indicators including 1.) Damage to human health, 2.) Damage to ecosystems, and 3.) Damage to resource availability.



Figure 4-22 ReCiPe (World H/A) single-score results (per kWh) of plasma gasification routes utilizing various feedstocks and in comparison to conventional power generation. \*PGRs using computer simulation data.

Depending on the *weighting set* chosen for the single score comparison different results may be obtained. For the ReCiPe World H/A model a weighting set of 40% human health, 40% ecosystems, and 20% resources is used. Using the mixing triangle developed by Hofstetter et al (Hofstetter et al. 1999) Figure 4-23 graphically depicts the outcome of product comparisons for various weighting sets.


Figure 4-23 Mixing triangle according to Hofstetter et al. (Hofstetter et al. 1999) comparing the use of CDDB and forest residuals on the basis of 1 kWh for single score impact categories 1.) Human Health, 2.) Ecosystems, and 3.) Resources. The ReCiPe World H/A method uses an average weighting set of 40% (human health), 40% (ecosystems), and 20% (resources). According to this, route 2, using CDDB as gasifier feed, would lead to a lower environmental impact that route 2 using forest residuals.

Assuming different weighting factors, in which e.g. resource depletion is considered more important, could change results in favor of forest residuals use. It should however also be noted that varying conversion efficiencies (e.g. 20% for 1-Bio vs. only 13% for 2-CDDB) impact the outcome of the LCA. In an optimized commercial plant, electrical conversion efficiencies are likely to be rather constant for various feedstocks gasified.

#### **E.Sensitivity Analysis**

The sensitivity of LCA results to assumptions made is tested by varying some of the input parameters (Table 4-6). This includes electricity inputs into the plasma torch as well as anthracite/coke inputs used as gasifier bed material. In addition, assumptions with regards to the steam turbine efficiency are tested.

Process	Current assumption	Alternative assumption	Note			
Energy inputs to plasma torch	2.8% (as percentage of total energy input)	1.7-5.0% (as percentage of total energy input)	Current assumption based on (AlterNRG 2009). Varying energy inputs reported in literature.			
Inputs of anthracite/coke to gasifier	~8% (by weight of feed)	4-8% (by weight of feed)	Pilot plant requires larger amounts of anthracite/coke. Commercial facility is expected to operate with about 4% (by weight of feed).			
Electrical conversion efficiency	34.8%	30-50%	Advanced combined cycle gas turbine could presumably provide higher conversion efficiency of up to 50-60% (Belgiorno et al. 2003)			
Waste Sorting (Choice of allocation*)	Mass allocation (22% of waste sorting allocated to plasma gasification LCA)	Economic allocation (waste sorting excluded from the analysis)	Concerned with waste inputs used as feedstock for plasma gasification.			
Wood growth/ harvest (Choice of allocation*)	Mass allocation (19% of wood growth/harvest allocated to plasma gasification LCA)	Economic allocation (wood growth/ harvest excluded from the analysis)	Concerned with biomass inputs used as feedstock for plasma gasification.			

Table 4-6 Parameters considered in the sensitivity analysis.

\*For mass allocation, the weight-% allocated towards the plasma gasification life-cycle is concerned with the dry weight of C&D wood recovered from the waste stream, or forest residues obtained from commercial forestry activities. For economic allocation it is assumed that the price of C&D wood is zero and all the environmental burdens of waste sorting are allocated to higher value products such as metals, plastics, and minerals recovered from the mixed waste stream. Similar to this, the price of forest residues (a by-product of saw log and pulpwood production currently left in the forest to decay) is assumed to be zero applying economic allocation and hence all upstream environmental burdens are allocated solely to saw log and pulpwood production. The sensitivity of varying <u>torch power</u> and <u>coke/anthracite</u> inputs as well as <u>turbine efficiencies</u> was tested. In the current pilot plant design the torch power equals 2.8% (as percentage of total energy input) while coke/anthracite inputs are approximately 8% by weight of the feed input. However, in a commercial plant it may be possible to further reduce the torch power inputs and coke/anthracite used as PGR bed material is predicted to vary around only 4% by weight of the feed input. This is due to lower heat losses associated with a larger PGR reactor design. Furthermore, in the current design the conversion efficiency of the steam turbine was modeled to equal 34.8%. Given that the installation of an advanced combined cycle gas turbine (IGCC) in the plasma gasification system may be able to reach higher efficiencies of up to 60% (Belgiorno et al. 2003), the turbine efficiency is varied between 30-50%. Results of the sensitivity analysis are shown below as an example for the 2-CDDB life-cycle.



Figure 4-24 Sensitivity analysis for the 2-CDDB life-cycle with regards to anthracite and torch power inputs as well as varying turbine efficiencies for electricity generation. All environmental impacts are shown relative to the current pilot plant design (100%) within each impact category. The functional unit is 1 kWh at the factory gate. \*Current pilot plant design: Plasma torch power = 2.8% (as percentage of total energy input), Coke/Anthracite input = 8% by weight of feed input, Steam turbine conversion efficiency = 34.77%.

As shown in Figure 4-24, halving anthracite inputs used as PGR bed material leads to a recognizable decrease in environmental burdens in particular in fossil depletion potential (FD) (~30%), respiratory effects and acidification (both ~20%), as well as GWP and smog (both ~10%). Varying plasma torch power inputs impact the overall conversion efficiency of the system therefore either reducing (1.7% of total energy input) or increasing (5.0%) environmental burdens. The lowest impacts are found if an increase in

the turbine efficiency of up to 50% is assumed (e.g. via IGCC). This may have the potential to reduce overall environmental impacts by roughly 30% compared to the pilot plant configuration. Table 4-7 shows absolute results for the 2-CDDB life cycle.

Impact category	Unit	Pilot Plant*	Anthracite (4%)	Plasma Torch 1.7%	Plasma Torch 5.0%	Turbine Eff 30%	Turbine Eff 40%	Turbine Eff 50%
GWP 100a	kg CO2 eq	6.53E-01	5.95E-01	6.03E-01	7.82E-01	7.56E-01	5.67E-01	4.54E-01
Fossil depletion	kg oil eq	2.62E-01	1.84E-01	2.42E-01	3.14E-01	3.03E-01	2.28E-01	1.82E-01
Land occupation	m2a	-6.84E-04	-7.88E-04	-6.32E-04	-8.19E-04	-7.92E-04	-5.95E-04	-4.75E-04
ODP	kg CFC- 11 eq	1.72E-08	1.71E-08	1.59E-08	2.06E-08	1.99E-08	1.50E-08	1.20E-08
Water use	m3	4.71E-03	4.69E-03	4.35E-03	5.64E-03	5.45E-03	4.09E-03	3.27E-03
Acidific.	H+ moles eq	1.90E-01	1.51E-01	1.75E-01	2.27E-01	2.20E-01	1.65E-01	1.32E-01
Eutrophic.	kg N eq	-1.27E-02	-1.29E-02	-1.18E-02	-1.52E-02	-1.47E-02	-1.11E-02	-8.85E-03
Smog	g NOx eq	1.29E-03	1.16E-03	1.19E-03	1.55E-03	1.50E-03	1.12E-03	8.99E-04
Respiratory effects	kg PM2.5 eq	7.78E-04	6.16E-04	7.18E-04	9.31E-04	9.01E-04	6.76E-04	5.41E-04

Table 4-7 Sensitivity analysis for the 2-CDDB life-cycle with regards to anthracite and torch power inputs as well as varying turbine efficiencies for electricity generation. The functional unit is 1 kWh at the factory gate. \*Current pilot plant design: Plasma torch power = 2.8% (as percentage of total energy input), Coke/Anthracite input = 8% by weight of feed input, Steam turbine conversion efficiency = 34.77%.

For life cycles 6 and 7 computer simulation data envisioning a commercial PGR was used. In this reactor the inputs of power to the plasma torch and the use of coke as bed material have already been optimized. Hence, we only vary assumptions with regards to the turbine efficiency assuming that in a modified design the overall electrical conversion efficiency may be increased due to the use of a gas turbine/IGCC. Table 4-8 shows results of the analysis for the 7-Bio/CDDB life-cycle.

Table 4-8. Sensitivity analysis for the 7-Bio/CDDB life-cycle with regards to varying turbine efficiencies for electricity generation (all other inputs have already been optimized due to the use of computer simulation model data for the PGR).

Impact category	Unit	Current configuration*	Turbine Eff 30%	Turbine Eff 40%	Turbine Eff 50%
GWP 100a	kg CO2 eq	3.25E-01	3.77E-01	2.83E-01	2.26E-01
Fossil depletion	kg oil eq	1.10E-01	1.27E-01	9.53E-02	7.62E-02
Land occupation	m2a	1.85E+00	2.15E+00	1.61E+00	1.29E+00
ODP	kg CFC-11 eq	1.49E-08	1.73E-08	1.29E-08	1.04E-08
Water use	m3	2.10E-03	2.43E-03	1.82E-03	1.46E-03
Acidification	H+ moles eq	6.95E-02	8.05E-02	6.04E-02	4.83E-02
Eutrophication	kg N eq	-4.22E-03	-4.89E-03	-3.67E-03	-2.93E-03
Smog	g NOx eq	6.66E-04	7.72E-04	5.79E-04	4.63E-04
Respiratory effects	kg PM2.5 eq	2.24E-04	2.59E-04	1.95E-04	1.56E-04

The functional unit is 1 kWh at the factory gate. \*Current configuration using computer simulation data for the PGR and assuming a steam turbine conversion efficiency of 34.77%.

As shown in the table, the use of a turbine with 50% efficiency could reduce environmental impacts of the overall plasma gasification power plant by roughly 30%. In such a design GWP would be with 0.226 kg CO<sub>2</sub>-eq per kWh about half that of the NE power grid and one third that of conventional average power generation in the U.S.

The <u>choice of allocation</u> (mass vs. economic) associated with the multi-output unit processes 'waste sorting' and 'wood growth/harvest' is investigated by comparing results of the current LCA model using mass allocation to an economic allocation case in which the upstream environmental burdens of both unit processes are excluded from the analysis. This may be justified because of the low economic value of forest residues (in the NE U.S. these are currently left in the forest to decay) as well as C&D derived wood (the majority of C&D wood in the NE U.S. is currently landfilled). Figure 4-25 shows results of the sensitivity analysis for all bio-based test phases.



Figure 4-25 Economic vs. mass allocation for the bio-based life-cycles. It is assumed that if economic allocation is applied the upstream environmental burdens of C&D waste sorting and wood/growth and harvest are excluded due to the fact that both forest residues and C&D wood represent low-value waste fractions utilized by the plasma gasification system.

The figure shows that for the majority of impact categories the choice of economic allocation only leads to slightly reduced impacts. One exception is land occupation which is drastically reduced if economic allocation is assumed, as upstream land occupation associated with forest growth and harvest is excluded.

## **CHAPTER 5**

# LIFE CYCLE ASSESSMENT (LCA) OF POLYITACONIC ACID (PIA) PRODUCTION FROM U.S. NORTHEAST SOFTWOOD BIOMASS <sup>60</sup>

#### A. Abstract

Shifting the resource base for chemical and energy production from fossil feedstocks to renewable raw materials is seen by many as one of the key strategies towards sustainable development. The objective of this study is to assess the environmental burdens of producing polyitaconic acid (PIA), a water soluble polymer derived from itaconic acid identified by the U.S. Department of Energy as one of the top 12 value added chemicals, from U.S. Northeastern (NE) softwood biomass. Results are compared to corn-derived PIA and fossil-based poly acrylic acid (PAA) on the basis of 1 kg of polymer at the factory gate.

This study uses attributional life cycle assessment to quantify global warming potential (GWP), fossil energy demand (CED), acidification, eutrophication, water use, and land occupation of the polymer production routes. This includes feedstock growth

<sup>&</sup>lt;sup>60</sup> This chapter has been submitted as: Nuss, P. and Gardner, K. H. (In review). "Attributional Life Cycle Assessment (ALCA) of Polyitaconic Acid Production from U.S. Northeast Softwood Biomass." *Int. J. LCA.* 

and harvest, sugar extraction, fermentation, itaconic acid recovery, and subsequent polymerization.

Results indicate that the use of softwood-based PIA may be advantageous in terms of GWP (1.32 kg CO<sub>2</sub>-eq), CED (15.0 MJ-eq), and acidification (0.38 H+ moles eq) when compared to both, the integrated corn-biorefinery (per kg PIA: 2.19 kg CO<sub>2</sub>-eq, 24.8 MJ-eq, and 0.73 H+ moles eq) and fossil-based PAA production (per kg PAA: 2.74 kg CO<sub>2</sub>-eq, 70.6 MJ-eq, and 0.45 H+ moles eq), respectively. When looking at impacts to eutrophication and water use, the use of softwood leads with 0.00970 kg N eq and 0.00751 m3 water to less potential impacts compared to its corn-based counterpart (0.0164 kg N eq and 0.0123 m3 water), but to higher impacts when compared to fossilbased PAA (0.000425 kg N eq and 0.00478 m3 water). Land occupation, to a large extent due to lower yields and longer growth cycles associated with softwood growth in the NE, is highest for softwood-derived PIA (8.41 m2a) and lowest for fossil-based PAA (0.024 m2a). Environmental impacts are mainly the results of up-stream environmental burdens associated with on-site electricity use, inputs of activated carbon and sodium hydroxide, as well as water use during sugar extraction and fermentation. Assumptions with regards to allocation, activated carbon inputs, and electricity mixes to processes of the foreground system are tested in a sensitivity analysis.

Wood-derived PIA production may be an interesting alternative to current fossilbased pathways and could contribute to a future bio-based economy. However, currently land occupation and water use are high when compared to traditional PAA production. The use of short rotation plantations and optimization with regards to water requirements and reuse could be investigated to further lower system-wide impacts.

*Keywords:* Life cycle assessment (LCA), polyitaconic acid, biopolymers, integrated biorefinery, softwood biomass, corn feedstock, polyacrylic acid, stream-integrated process.

#### **B.Introduction**

The last decade has seen a strong political and technical focus on using biomass feedstock to produce bio-fuels and energy. Much less attention has been given to biomass as a feedstock for chemicals production (Dodds and Gross 2007). However, while the economy of energy can be based on various alternative technologies utilizing e.g. wind, sun and water, the materials economy of substances will increasingly depend on the use of renewable biomass as well as the reutilization of existing material stocks within the technosphere. Using biomass to generate electricity and process heat is likely to be a bridge-technology before other renewable energy becomes economically viable on a widespread basis. It is expected that as the era of a chemical industry based on non-renewable oil, gas and coal will gradually come to an end over the course of the next 50 to 75 years, industrial production of platform chemicals from biomass feedstock will become of growing interest (B. Kamm et al. 2006).

In 2004, itaconic acid (IA) was identified by the DOE-EERE<sup>61</sup> as one of the top 12 value added chemicals potentially available from biomass (Werpy and G Petersen 2004). After polymerization, bio-based polyitaconic acid (PIA) is functionally an alternative to fossil-derived polyacrylic acid (PAA), a well-established petrochemical with a current global production of more than 2 million tons per year (Itaconix, LLC 2009). PIA is a water soluble polymer with a wide range of applications including superabsorbents, anti-scaling agents in water treatments, co-builders in detergents, and dispersants for minerals in coatings (Itaconix, LLC 2009). PIA is currently produced from corn-derived IA by fermentation. The use of woody biomass from the Northeastern (NE) United States (U.S.) as feedstock for PIA production is investigated by Itaconix LLC<sup>62</sup> via a *stream-integrated approach* in which extracted hemicellulose serves as feedstock for the biorefinery, while the partially macerated wood and lignin can be used in other existing processes such as pulp & paper plants for conventional pulp and bioenergy<sup>63</sup> production (Durant 2011; Itaconix, LLC 2009).

The seven states of the NE U.S.<sup>64</sup> have abundant forest resources and an established forest management sector (Benjamin et al. 2009). Currently, the region has an average accessible forestland cover of 70.6%, ranging from a high of 88.4% in Maine to a low of 52.8% in Rhode Island (Benjamin et al. 2009; US Forest Service 2010). The total

<sup>&</sup>lt;sup>61</sup> U.S. Department of Energy (DOE) - Energy Efficiency and Renewable Energy (EERE)

<sup>62</sup> www.itaconix.com

<sup>&</sup>lt;sup>63</sup> Following current practices, hemicellulose and lignin would be used as 'black liquor' for bioenergy production on site the pulp & paper plant.

<sup>&</sup>lt;sup>64</sup> Connecticut, Maine, Massachusetts, New Hampshire, New York, Rhode Island, and Vermont

accessible forestland area totals 49.9 million acres. For new biorefineries to work in a competitive manner, the degree to which existing feedstock use overlaps with biomass requirements for new biochemical production pathways needs to be examined. The stream integrated approach proposed may offer advantages in terms of feedstock competition. However, it is unclear if the use of wood-derived feedstock for PIA production is beneficial from an environmental perspective.

#### **C.Methods**

Against this background, the purpose of this paper is to assess the cradle-to-gate environmental burdens associated with softwood-derived PIA production and how these compare to conventional production practices. The study uses *attributional life cycle assessment (ALCA)* in combination with commonly used impact assessment (LCIA) methods to evaluate global warming potential (GWP) and fossil/nuclear cumulative energy demand (Goedkoop et al. 2008), acidification and eutrophication (Bare et al. 2002), as well as water use and land occupation (Goedkoop et al. 2009). SimaPro 7.3 is used to build the LCA model and carry out the impact assessment.

#### C.1. Functional unit and system boundary

The synthesis pathway assessed in this report produces *Itaconix<sup>TM</sup> Dispersant* DSP2K (poly (sodium itaconate)), a low molecular weight linear polyitaconic acid partially neutralized with sodium salt (Itaconix, LLC 2010). In the present study, for simplicity the product is assumed to be functionally equivalent to fossil-based poly

(sodium acrylate) which is obtained by polymerizing acrylic acid partially neutralized with sodium salt. We assume that PIA (DSP2K) based on either woody biomass or corn feedstock has similar or better properties in comparison to its fossil-based counterpart and would *functionally substitute* poly (sodium acrylate). Hence, a *functional unit* of 1 kg of dry polymer is used. For the remainder of this chapter we will use the abbreviation *PIA* for the poly (sodium itaconate) product and *PAA* for the poly (sodium acrylate) product.

Figure 5-1 shows the major stages of the product systems, which are investigated as unit processes.



Figure 5-1 System boundary of the PIA production route studied.

The system under consideration includes wood cultivation and harvest, transport to the biorefinery, hemicellulose (xylane) extraction, fermentation, recovery of the IA, and polymerization into PIA. The hemicellulose extraction process yield valuable byproducts including Kraft pulp which can be sent to conventional pulp & paper mills. Fermentation yields mycelium as by-product which could serve as protein-rich animal feed after drying. For multi-output processes mass allocation is applied. The impact of economic allocation on results is tested in a sensitivity analysis.

Results are compared to PIA produced from corn-based IA. The life-cycle includes corn cultivation and harvest and transportation to the biorefinery, corn wet milling, and fermentation into IA followed by polymerization using the Itaconix process. Furthermore, the life-cycle of functionally equivalent PAA consists of fossil raw materials acquisition, acrylic acid production via the acroelein process (two-step propylene oxidation), polymerization and neutralization using sodium hydroxide (NaOH).

#### C.2. Biogenic Carbon Accounting

We assume that biogenic  $CO_2$  emitted during PIA production (e.g. from fermentation) and subsequent biodegradation is eventually equal to the carbon assimilated during growth, thus presuming a net flux of biogenic carbon of zero (*implicit sequestration credits*). The reasoning behind this is that PIA used e.g. in detergents and dispersants would presumably have a short life-time (days to months) before biodegradation takes place and hence carbon would not be captured over longer time periods (i.e. years as in durable goods). However, carbon storage will depend on the type of final product and consumer habits. If PIA is used in long-lived products such as plastics) carbon sequestration may be accounted for. Furthermore, carbon dynamics associated with forest growth (see e.g. (E. Johnson 2009; McKechnie et al. 2011; Walker 2010)) are excluded from the analysis.

## **D.Life Cycle Inventory**

Data for the biorefinery unit processes (Figure 5-1) is based on *confidential process data* from Itaconix and its partners (Durant 2011). Inputs of materials, energy, and resource use are modeled using data from the ecoinvent (Ecoinvent 2010) and U.S. LCI database (NREL 2008). The modeled biorefinery located in the U.S. NE region with biomass feedstock (softwood or corn) available within a 70 miles radius<sup>65</sup>. The NE power grid distribution (NEPOOL) (ISO New England 2010) is used for electricity inputs to the unit processes. We test the sensitivity of results with regards to varying power grid inputs to biorefinery unit processes, i.e. NEPOOL (ISO New England 2010), U.S. average (Ecoinvent 2010), and hydropower (Bauer et al. 2007; Ecoinvent 2010). The following sections explain process steps and assumptions in more detail. Due to the confidentiality of some of the foreground data obtained directly from Itaconix (Durant 2011), quantitative numbers are only partly provided.

# **D.1. Softwood-based PIA**

The softwood-derived PIA production system is divided into five major steps including 1.) Softwood cultivation and harvest; 2.) Transport of softwood to the xylane

<sup>&</sup>lt;sup>65</sup> In reality, the corn-based biorefinery would likely be located in the Midwestern U.S. in close proximity to the corn-belt. However, since the goal & scope of this paper is to compare the biorefinery process using wood to the use of corn, we choose similar electricity mixes for all processes of the foreground system.

extraction plant followed by wood processing and conversion into fermentable carbohydrates (xylose); 3.) Conversion of carbohydrates into IA via fermentation; 4.) Recovery of the acid as sodium itaconate; and 5.) Conversion of sodium itaconate into poly (sodium itaconate) (DSP2K) polymer.

#### D.1.1. Softwood growth & harvest

Significant differences between countries and even regions within the United States (e.g. U.S. NE vs. Inland Northwest (INW)) exist in terms of site preparation activities, stand establishment, and fertilization (Oneil et al. 2010). For instance, for the NE no slash reduction activities are mandated for wildfire risk reduction, natural forest regeneration is assumed to be sufficient on all sites with no need for fertilization. In contrast, forest management activities in the Northwestern (NW) United States generally include seedling production for regrowth, commercial thinning and fertilization. We use the datasets for softwood growth and harvest and subsequent conversion towards saw-and pulplogs from (NREL 2008; Oneil et al. 2010)<sup>66</sup>. We consider pulpwood from forest harvest as feedstock for the integrated biorefinery. Wood chipping takes place by feeding the softwood logs at forest road into a mobile chopper (Ecoinvent 2010). With 4.05 m3/ha yr (Oneil et al. 2010), forest land occupation associated with NE softwood provision compares to an U.S. country average net annual increment (NAI)<sup>67</sup> of 3.64 m3/ha yr (W. B. Smith et al. 2009). Data on urban land occupation associated with forest

<sup>&</sup>lt;sup>66</sup> The process 'Pulpwood, softwood, US NE-NC' is used.

<sup>&</sup>lt;sup>67</sup> NAI represents the average annual volume over a reference period of gross increment less natural losses and hence represents a good estimate for the required forest land area for biomass provision.

roads comes from (Werner et al. 2007). *Allocation* of environmental burdens is *based on* the *weight* of pulpwood and by-products (i.e. sawlogs and bark).

# D.1.2. Xylane extraction

Softwood chips are transported to the conversion plant from within a 70 miles radius using a truck (NREL 2008). Wood chips are continuously fed to the extraction column and hemicelluloses xylane polymers extracted using steam. Steam is provided to the process from a natural gas (36.6 MJ/kg PIA) fired steam boiler located on site (Durant 2011). Electricity (0.56 kWh/kg PIA) is required to power auxiliary process equipment (e.g. conveyor belt, water pumps) (Durant 2011). The extraction process yields fermentable carbohydrates (7% dry weight), lignin (1%), and pulp-grade wood (92%). Lignin and partially macerated (pulp-grade) wood are separated from the liquid extract. Activated carbon (AC) (obtained from charcoal (Ecoinvent 2010) and activated using excess steam from the hemicellulose extraction process) and ionic exchange columns are used to remove potential fermentation inhibitors (e.g. acid soluble lignin or furfural) from the liquid extract before it is diverted to the fermenter. A total of 67.1 kg softwood chips at a moisture content of 55% are required for the production of 1 kg PIA and byproducts <sup>68</sup> (before allocation). The diversion of (hemicellulose-derived) fermentable carbohydrates is accounted for by mass as well as economic allocation based on the economic heating value of each product output (Appendix: Section C.1). Based on a heating value of 13.6 MJ/kg for hemicellulose and 25 MJ/kg for lignin (Amidon 2006),

<sup>&</sup>lt;sup>68</sup> This includes pulp-grade wood, lignin, and mycelium.

and a heating value of 15 MJ/kg and market price of \$35 per tonne for pulp-grade wood (Durant 2011), the economic value of hemicellulose and lignin equals \$32 per tonne and \$59 per tonne, respectively.

### D.1.3. Fermentation

IA is produced by fermentation of xylose and other extracted carbohydrates received from the wood extraction unit (referred to only as xylose throughout this paper). The process combines xylose and other media, adds a microbial inoculum as well as micronutrients to fuel the fermentation process, and produces crude IA (Durant 2011). Electricity (1.16 kWh/kg PIA) is used to power an air compressor and separation unit (ASU), adjusting fermentation conditions and controlling foam built-up and micronutrients added (Durant 2011). The outputs of the fermentation process are a clarified broth containing *IA* as well as *mycelium* from fungus growth. The culture broth is filtered to remove mycelia and other suspended solids. *Allocation* of environmental burdens is *based on* the dry *weight* of the product outputs. We investigate the impact of *economic allocation* (sensitivity analysis) using a current average market price of \$1.63/kg<sup>69</sup> for IA (Itaconix, LLC 2009) and \$0.80/kg for mycelium assumed to replace high-protein soybean animal feed (USDA 2011) (*Appendix: Section C.1*).

<sup>&</sup>lt;sup>69</sup> This price is simply based on currently existing commercial IA production pathways and does not imply the production cost or target price for Itaconix LLC.

## D.1.4. Recovery

After fermentation, IA is extracted from the clarified and filtered broth solution. Regeneration of the extractant solution (0.13 kWh/kg PIA) is required (Itaconix, LLC 2009). NaOH pellets (0.26 kg/kg PIA) are added to adjust the pH of the solution and AC added to further purify the broth before concentration using an evaporator takes place (Durant 2011). The main output of the recovery process step is an evaporated solution containing *sodium itaconate (NaIA)*. Due to a lack of data on the detailed composition of *wastewater* generated, we use typical numbers from commercial polylactic acid (PLA) production according to (Althaus, Werner, et al. 2007; E. T. H. Vink et al. 2003; E. Vink et al. 2007).

## D.1.5. Polymerization

Polymerization is based on a method developed by Itaconix that eliminates the need for post polymerization purification. An initiator is added to the itaconate solution to start the polymerization reaction (Durant 2011). The conversion efficiency is high and the resulting polymer is further packaged as a granulated material.

#### **D.2.** Corn-based PIA

Corn-based PIA represents a potential competitive alternative to wood-derived PIA (*Appendix: Figure 5-13*). The LCI for corn-based PIA is based on a combination of data describing 1.) Corn production in the United States) (Ecoinvent 2010; Jungbluth, Chudacoff, et al. 2007); 2.) Glucose production via corn wet milling (Akiyama et al.

2003; Gerngross 1999; Khoo et al. 2010; US EPA 2011b); and 3.) Fermentation, IA recovery, and polymerization (Durant 2011).

The PIA production plant is located on site the wet mill and hence no transportation of glucose is required. About 1.46 kg corn is required to produce 1 kg of glucose (Akiyama et al. 2003). By-products of the process are 0.378 kg corn meal & feed as well as 0.063 kg corn oil (allocation is based on mass). The energy requirements are estimated to be 4.9 MJ/kg glucose. The transportation distance of corn to the biorefinery is assumed to equal 49.5 miles one way (corn basket with a radius of 70 miles) via a combination truck using U.S. average fuel (NREL 2008). The input of dry glucose to the fermentation process is 1.71 kg per kg of PIA produced (Durant 2011). Yields, resource and energy inputs as well as emissions associated with fermentation, recovery, and polymerization are assumed to be similar to wood-based PIA production (see previous section). We account for water requirements for subsequent fermentation. In order to be in line with eco-profiles on bio-polymers published elsewhere (E. T. H. Vink et al. 2003, 2010; E. Vink et al. 2007) enzyme use (0.1%) by weight of corn input) is included in the assessment. However, due to a lack of LCI data on industrial enzyme production for integrated corn refineries, it was decided to use data on fungal glucoamylase production (Nielsen et al. 2006) as a proxy for environmental burdens.

#### **D.3. Fossil-based PAA**

PAA is produced from fossil-derived acrylic acid via polymerization using a radical initiator. Acrylic acid is produced via a two-step process from propylene. Due to

limited process data publically available on industrial PAA production, we use data from ecoinvent (Althaus, Hischier, et al. 2007) on acrylic acid production. In order to allow a fair comparison with PIA produced via the Itaconix process, we account for NaOH inputs required for the generation of sodium acrylate. We assume that the polymerization step of PIA and PAA production is not significantly different and use data on yields, energy requirements and amount of initiator used from (Durant 2011). Polymerization takes place in water using roughly 1 kg of process water per kg of polymer generated (Durant 2011). The final product is 1 kg of dry *poly (sodium acrylate)* at the factory gate.

# **D.4. Background Processes**

Background processes include electricity supplied from the U.S. Northeastern power grid and the U.S. average grid as well as hydropower used in an alternative scenario in which the PIA production plant is located nearby to a hydropower plant.

Table 5-1 shows the shares of different fuel types used in the NE region (*NEPOOL*) according to a 2010 report by ISO New England<sup>70</sup> (ISO New England 2010). We assume average distribution losses of 9.58% for the provision of electricity.

<sup>&</sup>lt;sup>70</sup> http://www.iso-ne.com/

Fuel type	Input [kWh]	Input [%]
Natural Gas	0.4646	42.4
Nuclear Power	0.3320	30.3
Coal	0.1348	12.3
Hydro	0.0767	7
Renewables	0.0668	6.1
Pumped Storage	0.0131	1.2
Oil	0.0077	0.7
TOTAL	1.0958*	100

Table 5-1 Electricity inputs to the NE electricity grid required to provide 1 kWh of electricity (ISO New England 2010).

The life cycle inventory for the background process "U.S. average electricity" and "hydropower" is based on data published by (Ecoinvent 2010). Detailed information on hydropower production can be found in (Bauer et al. 2007; Ecoinvent 2010). The process includes shares of electricity produced by run-of-river (84%) and reservoir hydropower plants in non-alpine regions (16%).

Table 5-2 shows the carbon intensity and contributions to other impact categories

of the power grid data used for the foreground system of the biorefinery plants.

Electricity inputs to the NE electricity grid required to provide 1 kWh of electricity (ISO New England 2010). Unit processes to the U.S. LCI database and Ecoinvent were used to provide the shares of electricity inputs. \*Accounts for average line losses of 9.58%.

Impact category	Unit	U.S. Average	U.S. Northeast	U.S. Hydropower	
		[per kWh]	[per kWh]	[per kWh]	
IPCC GWP 100a	kg CO2 eq	7.714E-01	5.372E-01	4.915E-03	
CED (Non-renewable, fossil)	MJ eq	9.035E+00	8.095E+00	3.793E-02	
CED (Non-renewable, nuclear)	MJ eq	3.325E+00	1.049E-01	4.182E-03	
CED (Non-renewable, biomass)	MJ eq	1.847E-06	2.644E-06	4.520E-08	
CED (Renewable, biomass)	MJ eq	1.359E-01	2.387E-02	2.464E-04	
CED (Renewable, wind, solar, geothermal)	MJ eq	1.564E-02	5.613E-02	1.886E-05	
CED (Renewable, water)	MJ eq	2.989E-01	4.626E-01	3.791E+00	
Acidification	H+ moles eq	2.725E-01	2.141E-01	9.952E-04	
Eutrophication	kg N eq	2.957E-03	8.023E-05	1.780E-06	
Water use	m3	2.176E-03	4.286E-04	4.323E-05	
Land occupation	m2a	1.549E-02	5.082E-03	1.505E-04	

Table 5-2 System-wide environmental burdens of delivering 1 kWh of electricity from the various energy carriers (power grids) to the conversion plant.

Data comes from NEPOOL, Ecoinvent and USEI.

# **E.Results and Discussion**

# E.1. Life Cycle Impact Assessment

## E.1.1. Midpoint Impact Categories

Figure 5-2 shows the results of the comparative LCA for the impact categories of global warming potential (GWP), fossil and nuclear cumulative energy demand (CED), acidification, eutrophication, water use, and land occupation (incl. forest, agricultural, urban).



Figure 5-2 Results of the comparative LCA showing relative contributions to each of the six impact categories investigated. The production pathway with the highest environmental impact is shown as 100% and impacts of the other two routes shown relative to this. The functional unit for comparison is 1kg of dry polymer at the factory gate. Land occupation for the wood-derived polymer (PIA, wood) is mainly due to the use of forest land (NE pulpwood used as PIA feedstock; woody biomass used for activated carbon (AC) provision), while com-based PIA requires both agricultural (corn feedstock) and forest land area (AC provision). The share of urban land occupation is negligible.

Table 5-3 shows quantitative results of the comparative LCA looking each at 1 kg softwood- and corn-derived PIA as well as fossil-based PAA. Relative contributions of

process steps to each impact category are shown in Figure 5-3.

Table 5-3 Results of the comparative LCA for all six impact categories investigated.

Impact category	Unit	PIA, Wood	PIA, Corn	PAA, Fossil-based
IPCC GWP 100a	kg CO2 eq		2.193E+00	1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1
CED (fossil/nuclear)	MJ eq	1.499E+01	2.480E+01	+ 1°
Acidification	H+ moles eq	3.781E-01		4.525E-01
Eutrophication	kg N eq	9.659E-03		4.253E-04
Water use	m3	7.506E-03		4.784E-03
Land occupation	m2a		2.457E+00	2.435E-02

The functional unit for comparison is 1 kg of polymer at the factory gate. The route with the lowest impact is shown in green color, while the route with the highest impact is shown in red. Land occupation for the wood-derived polymer is mainly due to the use of forest land (NE pulpwood used as PIA feedstock; woody biomass used for activated carbon (AC) provision), while corn-based PIA requires both agricultural (corn feedstock) and forest land area (AC provision). The share of urban land occupation is negligible.



Figure 5-3 Relative contributions of subsystems to the environmental impacts of PIA production from corn and softwood. The functional unit is 1 kg of PIA at the factory gate. GWP: global warming potential; CED: cumulative energy demand (fossil and nuclear); AP: acidification potential; EP: eutrophication potential; WU: water use; LO: Land occupation. LO for the wood-derived polymer is mainly due to the use of forest land (NE pulpwood used as PIA feedstock; woody biomass used for activated carbon (AC) provision during IA recovery), while corn-based PIA requires both agricultural (corn feedstock) and forest land area (AC provision). The share of urban land occupation, mainly due to forest roads, is negligible.

Results indicate that with 1.32 kg CO<sub>2</sub>-eq per kg product output, the wood-based polymer has a significantly lower *GWP* than both corn-based PIA (2.19 kg CO<sub>2</sub>-eq) as well as fossil-based PAA (2.73 kg CO<sub>2</sub>-eq) (Figure 5-4).



Figure 5-4 Global warming potential (GWP) of the PIA polymers (1 kg at the factory gate) from wood and corn feedstock compared to the production of an equivalent amount of PAA from fossil-feedstock.

Polymerization, with its high conversion efficiency, contributes only a minimal share to overall GWP, while IA recovery from the broth solution leads to roughly 0.92 kg  $CO_2$ -eq for both biobased polymers (wood and corn). The reason for this is mainly the use of NaOH, used for neutralization and formation of NaIA, and activated carbon (AC) added before evaporation takes place. In addition, electricity required during fermentation and IA recovery contributes to GWP. The major difference between the wood- and cornbased production routes in terms of  $CO_2$  emissions are the different contributions of

feedstock growth/harvest as well as sugar extraction. Applying mass allocation, only 7.2% of all upstream burdens (i.e. softwood growth/harvest and fermentable sugar extraction) are counted towards the production of wood-based PIA. This is a result of the *stream integrated approach* undertaken in which hemicellulose from softwood chips is extracted with the partially macerated wood serving as feedstock for conventional Kraft pulping in a pulp & paper mill, therefore being a usable by-product. In contrast, for cornderived PIA 69.4% of all upstream burdens associated with glucose production from corn wet milling are allocated towards the polymer life-cycle. Hence, the contributions to overall GWP are significantly higher than for softwood-derived PIA. This trend is visible throughout the various impact categories.

With 15.0 MJ-eq, the wood-based route requires significantly less primary and secondary *energy* from fossil and nuclear sources<sup>71</sup> than corn-based PIA (24.8 MJ-eq) and their fossil-based counterpart (70.6 MJ-eq) (Figure 5-5).

<sup>&</sup>lt;sup>71</sup> The CED indicator encompasses non-renewable fossil (i.e. coal, oil, etc.) and nuclear (i.e. uranium) energy demand.



Figure 5-5 Cumulative energy demand (CED – fossil/nuclear) for the three polymer production routes.

Trends observed are similar to GWP. The reason is that the energy content of the biomass feedstock (softwood or corn) is not captured in the indicator. However, even if we account for CED including renewables such as biomass and others, the environmental burdens are still lower than for PIA from corn and fossil-based PAA (Figure 5-6).



Figure 5-6 Cumulative energy demand (CED) for non-renewables and renewables comparing 1 kg polymer output from the different production routes. Even if the energy content of the non-renewable biomass is taken into account, CED is still lowest for the softwood-based PIA production route.

Acidification associated with corn-based PIA production is with 0.73 H<sup>+</sup> moles-eq almost twice as high as for wood-based PIA (0.38 H<sup>+</sup> moles-eq) and fossil-based PAA (0.45 H<sup>+</sup> moles-eq) (Figure 5-7).



Figure 5-7 Acidification impacts associated with the production of 1 kg polymer at the factory gate.

The unit processes leading to the highest share in acidification burdens for the corn-based route are electricity inputs to corn wet milling and glucose fermentation process (51%) as well as NaOH inputs to the recovery step (27%) and corn production (22%), during which e.g. ammonia and nitrous oxides from fertilization are emitted to the environment.

*Eutrophication* is highest for the corn-based production route (0.0164 kg N-eq) and lowest for fossil-based PAA (0.00043 kg N-eq) (Figure 5-8).



Figure 5-8 Eutrophication impacts associated with the production of 1 kg polymer at the factory gate.

The reason for this is that during IA recovery large amounts of wastewater are generating causing eutrophication during conventional treatment (Ecoinvent 2010). Due to a lack of site specific wastewater composition data we use numbers for commercial PLA production (Althaus, Werner, et al. 2007) and therefore the eutrophication potential should only be seen as a first indicator. For corn-based PIA, corn growth and harvest lead to additional eutrophication impacts (0.00669 kg N-eq), mainly due to commercial fertilizer use. In contrast, according to (Oneil et al. 2010) softwood production in the U.S. NE generally does not use any fertilizer during the growth phase<sup>72</sup> and therefore eutrophication impacts are minimal. However, this assumption will differ depending on forest management practices and whether future demands can be supplied from naturally regenerating forests in the NE.

<sup>&</sup>lt;sup>72</sup> According to (Oneil et al. 2010), fertilization is used by a few large private landowners but it is overall not a common practice in the NE region.

Impacts to *water use* are caused mainly by water use during feedstock growth (irrigation) and inputs to the biorefinery, in particular to the unit processes of fermentable sugar extraction and fermentation (Figure 5-9).



Figure 5-9 Water use associated with the production of 1 kg polymer at the factory gate.

With 0.0123 m<sup>3</sup> per kg PIA, water use seems highest for the corn-based route, followed by 0.00751 m<sup>3</sup>/kg for wood-based PIA and 0.00478 m<sup>3</sup>/kg for conventional PAA. Corn feedstock irrigation results in 0.0047 m<sup>3</sup> of water use, while NE softwood stems from naturally grown forests not requiring artificial irrigation. Corn wet milling and fermentation lead to an additional demand of 0.0017 and 0.0051 m<sup>3</sup> water, respectively. For the softwood-based polymer, impacts to water use are a result mostly of water used for xylane extraction and fermentation, wastewater treatment, and AC production (background process). The production of wood-derived PIA leads to the occupation of 8.3 m2a forest and 0.092 m2a urban land area. *Land occupation* is to 77% due to softwood tree growth, which have relatively long rotation cycles (65 years) and lower yields per hectare and year when compared to corn feedstock. This is due to the fact that NE forest biomass comes from naturally regenerating forests (i.e. no short rotation plantations are used). The use of pulpwood from naturally regenerating forest in the NE United States has to be distinguished from the use of feedstock grown on agricultural land (e.g. corn feedstock) specifically for the purpose of use in a biorefinery. Both may result in different pressures on ecosystems per unit of land area occupied. The remaining 23% of land occupation are due to land requirements for AC provision (produced from hardwood residuals) used during IA recovery. Urban land occupation is small (by a factor of 100) when compared to forest land and is mainly due to the use of forest road for vehicles to access forest area for maintenance and final harvest (Werner et al. 2007). No infrastructure area requirements for the biorefinery plant are accounted for.



Figure 5-10 Land occupation (incl. forest, agricultural, urban) associated with the production of 1 kg of polymer at the factory gate. Land occupation for the wood-derived polymer is mainly due to the use of forest land (NE pulpwood used as PIA feedstock; woody biomass used for activated carbon (AC) provision), while corn-based PIA requires both agricultural (corn feedstock growth and harvest) and forest land area (AC provision). Urban land occupation, due to e.g. forest roads and other infrastructure, is negligible.

With 2.4 m2a of land used (roughly 25% of this is due to agricultural land occupation for corn production, and 75% due to charcoal (AC) production from forest lands), the use of corn seems beneficial from a land occupation standpoint even if the wood biorefinery is integrated with a pulp and paper facility recovering macerated softwood after extraction for use in a pulp & paper plant. However, the diversion of pulp-softwood towards PIA production on scales that would allow continued sustainable forestry practices (i.e. harvest in NE forests equals natural regeneration) would also help to preserve NE forest lands. Hence, the occupation of naturally regenerating forest land may have positive aspects that are only captured by extending the analysis to investigate related ecosystem pressures per m2 of land occupied, and by including land

transformation in the analysis (see e.g. (Koellner and Scholz 2007). This remains to be done in a future study. In contrast to both biobased polymers, fossil-based PAA production (a highly optimized process) results in significantly less land occupation (0.0244 m2a/kg) since mostly fossil feedstock (crude oil for the generation of propylene) is required.

## E.1.2. Endpoint Impacts (ReCiPe)

Finally, polymers produced from NE softwood are compared to corn feedstock and conventional fossil-based PAA production using the ReCiPe (H/A) endpoint method (Goedkoop et al. 2009). In ReCiPe, eighteen midpoint indicators are transformed into three endpoint indicators including 1.) Damage to human health; 2.) Damage to ecosystems; and 3.) Damage to resource availability. The motivation to calculate and show the endpoint indicators is that the large number of midpoint indicators is often difficult to interpret.

Results assuming mass allocation show that the softwood-based production route scores significantly better when compared to PIA obtained from corn as well as PAA from fossil resources (Figure 5-11). Figure 5-12 shows the single impact score for the softwood based Itaconix process per unit process (i.e. feedstock growth/harvest, sugar extraction, fermentation, recovery, and polymerization).



Figure 5-11 Comparison of the system-wide environmental impacts associated with the generation of 1 kg of PIA from softwood and corn feedstock and in comparison to conventional fossil-based PAA production. Environmental impacts were calculated using the ReCiPe endpoint method (World H/A). It should be noted that calculation of a single score leads to higher uncertainty when compared to midpoint indicators. Results shown in this figure should therefore be used in connection with results of TRACI as well as CED and land occupation (see previous chapters).


Figure 5-12 Single impact score using ReCiPe for each unit process of the foreground system of softwood-based PIA production. The functional unit is 1 kg of PIA at the factory gate. As expected, polymerization with its high conversion efficiency leads to almost no environmental impact, while recovery and fermentation contribute significantly to climate change and fossil depletion. The process of feedstock (i.e. softwood) growth and harvest has the largest impact to land use as discussed earlier. Environmental impacts were calculated using the ReCiPe endpoint method (World H/A). It should be noted that calculation of a single score leads to higher uncertainty when compared to midpoint indicators. Results shown in this figure should therefore be used in connection with results of TRACI as well as CED and land occupation (see previous chapters).

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#### E.2. Sensitivity Analysis

Applying *economic allocation* instead of mass allocation to the biorefinery model (sugar extraction and fermentation) leads to a slight increase of environmental burdens for both PIA production routes (*Appendix: Section C.1*). The reason is that IA obtained from fermentation has a higher market price when compared to mycelium (assumed to serve as high-protein animal feed). In contrast to this, applying economic values to the products of the sugar extraction process does not significantly change allocation percentages. However, even if economic allocation is applied to both unit processes (sugar extraction and fermentation), impacts to GWP (1.46 kg CO<sub>2</sub>-eq), CED (17.3 MJ-eq), and acidification (0.433 H+-moles-eq) for the softwood-based route are still significantly lower when compared to corn-based PIA (2.66 kg CO<sub>2</sub>-eq, 31.0 MJ-eq, 0.914 H+ moles-eq) and fossil-based PAA (2.74 kg CO<sub>2</sub>-eq, 70.6 MJ-eq, 0.423 H+ moles-eq) production. Nevertheless, with regards to eutrophication, water use, and land occupation, fossil based PAA production still results in the lowest system-wide environmental impacts.

Replacing the NEPOOL power mix with *hydropower* reduces GWP for the softwood-based polymer to 0.96 kg  $CO_2$ -eq, CED to 9.4 MJ-eq, and acidification to 0.232 H+ moles-eq. (*Appendix: Section C.2*) Impacts to eutrophication, water use, and land occupation are mainly a result of direct inputs and/or emissions to the biorefinery system including feedstock growth/harvest. As a result, these impacts are less influenced by choices made with regards to the electricity inputs. Overall, using hydropower has the potential to reduce impacts to all categories (reductions in eutrophication and land

occupation are minimal), while using U.S. average power would lead to the highest impacts to all categories. For corn-based PIA, a similar tendency is observed.

Activated carbon (AC) used for the removal of chemical substances from the broth solution (sugar extraction and fermentation) was assumed to be disposed after use (i.e. burned on site without energy recovery)<sup>73</sup>. In reality, AC in stacks may be reactivated using heat (e.g. provided by burning by-products from extraction on site). In addition, the amount of AC is likely to vary depending on the final process design and concentration of chemical substances to be removed. Varying the amounts of AC from the baseline configuration (100%) to half (50%) and twice (200%) the amount of AC, respectively, most significantly impacts GWP as well as land occupation (*Appendix: Section C.3*). For example, reducing overall AC use by half could lead to a reduction of roughly 17% in GWP for the softwood-based biorefinery (1.32 to 1.09 kg CO<sub>2</sub>-eq) and 10% for the corn-based route (2.19 to 1.98 kg CO<sub>2</sub>-eq). Similarly, land occupation could be reduced by 11% from 8.3 to 7.4 m2a/kg for softwood-based PIA and 38% from 2.4 to 1.5 m2a/kg for corn-based PIA, respectively.

#### **F.Conclusions**

#### F.1. Recommendations

Among the main contributors to both wood and corn-based PIA production are: 1.) Electricity used during fermentation and itaconic acid (IA) recovery, 2.) Activated

<sup>&</sup>lt;sup>73</sup> Carbon dioxide from AC disposal is of biogenic origin, hence not accounted for in the GWP indicator.

carbon (AC) used during IA recovery and sugar extraction (only for wood-based PIA), 3.) NaOH used during recovery, 4.) Water use during sugar extraction and fermentation, and 5.) Land occupation associated with softwood, corn, and AC inputs. As a result, life cycle wide systems performance is directly affected by variations in these inputs.

For example, the choice of a less carbon intensive electricity mix (e.g. from hydropower) on site the biorefinery could be combined with increased energy efficiency measures, i.e. optimizing the electricity inputs in particular to air compression (fermentation) and evaporation/concentration (IA recovery). Furthermore, lowering ACinputs to the unit processes of extraction and recovery (e.g. by regeneration on site) may contribute to reduce impacts in particular to GWP and land occupation. The use of NaOH most significantly contributes to GWP, CED, and acidification. This is due to upstream burdens associated with NaCl production and power use in the diaphragm membrane and cell electrolysis processes. NaOH is used up by the process and therefore needs to be continuously replaced. System-wide impacts may be reduced by testing other neutralizing agents with lower life-cycle wide burdens to obtain new polymer products. Water use for sugar extraction and subsequent fermentation may be reduced by investigating on-site effluent treatment and enhanced recirculation (e.g. of distilled water obtained during evaporation). High water use may be an obstacle for PIA production in arid regions. Finally, considering other waste feedstocks such as softwood derived from construction and demolition could help to reduce impacts to land occupation.

#### F.2. Limitations of the LCA

*Cradle-to-gate perspective:* Our assessment does not include the use and disposal phase of the polymer product. PIA and PAA polymers can be used in a variety of applications (e.g. superabsorbents, anti-scaling agents in water treatment, co-builders in detergents, etc.) and all of these products will have different use-phases, life-times and disposal scenarios. By choosing a *cradle-to-gate* perspective these environmental impacts are not included in our assessment.

Attributional LCA: Results of the LCA describe the environmentally relevant flows using *current inventory data* and market prices for economic allocation. However, many of the assumptions made in this assessment might change over time. For example, market prices used for economic allocation fluctuate on a daily basis, in particular with regards to global prices for e.g. corn and other food crops. The LCA model assumes that results are stable over time and resistant to changes in other parts of the economy. This type of analysis does therefore not take into account that due to a decision supported by the LCA, production patterns might be changed in the future.

Forest growth carbon dynamics: A distinction is made between fossil and biogenic sources of carbon emitted to the atmosphere. Biomass feedstock for PIA production is assumed to be carbon neutral. However, there is a recent controversy among the scientific community with regards to the carbon neutrality of biomass (E. Johnson 2009). Literature published on the subject suggests to report carbon-stock changes and to include the effect of time in any sustainability analysis (E. Johnson 2009; McKechnie et al. 2011; Walker 2010). Especially in temperate forests, in which the harvest cycle can range from 60 to 100 or more year, carbon stock replacement can take many decades. We did not account for carbon dynamics associated with forest growth as this was outside the goal & scope of the assessment.

Geographical scope of the assessment: The study has been carried out for the NE U.S. using region-specific data for softwood growth, the NEPOOL power mix, and U.S. specific LCIA characterization factors for acidification and eutrophication (Bare et al. 2002). Forest growth and harvest practices in the NE U.S. are distinctively different from other regions. For example, no artificial fertilization of the trees is undertaken and rotation age and harvest yields were found to be significantly different than for other regions. Therefore, results of the study may not directly be applied to other regions of the U.S. or the world.

Lab- and pilot-scale data: Finally, the life-cycle inventory compiled for this assessment is based on preliminary energy and mass balances from Itaconix and its partners (Durant 2011). Data comes from lab- and pilot plant test runs and was assumed to be scalable to larger facilities. In reality, some of the data used in our assessment might change for a commercial facility. In addition, the process might be further optimized in the future according to experience gained during commercial operation.

#### **G.** Acknowledgements

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## APPENDIX 74

# LIFE CYCLE ASSESSMENT (LCA) OF POLYITACONIC ACID (PIA) PRODUCTION FROM U.S. NORTHEAST SOFTWOOD BIOMASS

## A. Abstract

This document contains additional detailed information with regards to the sensitivity analysis carried out as part of this project.

## **B.Inventory Analysis**

Figure 5-13 shows the process step for corn based PIA production.

<sup>&</sup>lt;sup>74</sup> The appendix has been submitted as supporting information with the paper: Nuss, P. and Gardner, K. H. (In review). "Attributional Life Cycle Assessment (ALCA) of Polyitaconic Acid Production from U.S. Northeast Softwood Biomass." *Int. J. LCA*.



Figure 5-13 Process flow chart for the production of corn-based PIA polymers.

## **C. Sensitivity Analysis**

We perform a sensitivity analysis in order to see the influence of the most important assumptions on the results of the LCA. This includes:

• The influence of mass vs. economic allocation for outputs of the sugar extraction unit process;

- The influence of mass vs. economic allocation for mycelium by-produced during fermentation
- Stability of the results with regards to varying electricity mixes (i.e. NEPOOL vs. U.S. average vs. Hydropower)
- Varying inputs of activated carbon (AC) to the xylane extraction and itaconic acid (IA) recovery process

## C.1. Mass vs. Economic Allocation

The choice of mass vs. economic allocation for product outputs from the *xylane extraction* process (a multi-output process), during which fermentable carbohydrates, pulp-grade wood, and lignin are produced, plays an important role for the results of the LCA. Mass allocation accounts for the fact that hemicellulose for sugar extraction represents only a small fraction (by mass) of the softwood, while the remainder (excluding a small lignin fraction) is assumed to be sent as feedstock to a conventional pulp & paper plant after extraction took place. Mass allocation in this sense accounts for the stream integrated approach pursued by Itaconix in which it is assumed that the biorefinery is co-located (or in close proximity) to a pulping plant accepting the partially macerated softwood as feed.

However, from an economic standpoint it may make sense to look at the economic value of each product output (i.e. the hemicellulose for sugar extraction, pulpgrade wood and lignin). According to current practices hemicellulose and lignin would be burned on-site the pulp & paper plant. Economic allocation should therefore be based on the economic heating value of each product output.

Based on a heating value of 13.6 MJ/kg for hemicellulose and 25 MJ/kg for lignin (Amidon 2006), and a heating value of 15 MJ/kg and market price of \$35 per tonne for pulp-grade wood (Durant 2011), the economic value of hemicellulose and lignin equals \$32 per tonne and \$59 per tonne, respectively.

Table 5-4 Average market prices as of January 2011 and corresponding allocation factors for the products of the Itaconix extraction process.

[\$/tonne]	Source	Allocation [%] based on mass	Allocation [%] based on economic value
31.98	(Amidon 2006)	7.20	6.53
35.27	(Durant 2011)	91.99	92.12
58.78	(Amidon 2006)	0.81	1.35
	<b>[\$/tonne]</b> 31.98 35.27 58.78	[\$/tonne]         Source           31.98         (Amidon 2006)           35.27         (Durant 2011)           58.78         (Amidon 2006)	[\$/tonne]         Source         Allocation [%] based on mass           31.98         (Amidon 2006)         7.20           35.27         (Durant 2011)         91.99           58.78         (Amidon 2006)         0.81

<sup>1</sup>Based on a heating value of 13.6 MJ/kg for hemicellulose. <sup>2</sup>Based on a price for pulp-grade wood chips of \$32/short ton and a heating value of 15 MJ/kg. <sup>3</sup>Based on a heating value of 25 MJ/kg for lignin.

As shown in Table 5-4, applying economic allocation does not result in a significant difference in allocation percentages.

Similar to this, instead of mass allocation the use of economic allocation can be applied to the outputs of the *fermentation process*, i.e. itaconic acid and mycelium. Mycelium, being rich in protein, has the potential to replace conventional animal feed such as high-protein soybeans, meat & bone meal, or fish meal. We assume that the mycelia by-product would replace high-protein soybean animal feed.

Table 5-5 Average market prices as of January 2011 and corresponding allocation factors for the products of the Itaconix fermentation process.

Market prices of fermentation products	[\$/tonne]	Source	Allocation [%] based on mass	Allocation [%] based on economic value
Itaconic acid <sup>1</sup>	1625.00	(Itaconix, LLC 2009)	46.77	64.07
Mycelia <sup>2</sup>	800.57	(USDA 2011)	53.23	35.93

<sup>1</sup>Based on a price of \$1.35-1.90/kg for itaconic acid (Itaconix, LLC 2009). <sup>2</sup>Based on a price for highprotein soybean animal feed of \$0.36/lb (USDA 2011) which is assumed to represent animal feed to be replaced.

The market price of itaconic acid, ranging between \$1.35-1.90/kg (Itaconix, LLC 2009), is higher than animal feed and therefore more of the environmentally relevant mass and energy flows are counted towards the PIA product (Table 5-5).

A comparison of the impact of mass vs. economic allocation on all environmental impact categories of the *softwood-based PIA polymer* is shown in Figure 5-4.

Quantitative results of the allocation comparison are shown in Table 5-6.



Figure 5-14 Sensitivity of the results of the softwood-based LCA to changes in allocation assumptions (i.e. mass vs. economic allocation for outputs of the sugar extraction and fermentation unit processes). The first bar shows results based solely on allocation by mass (see results shown in the previous sections for a detailed contribution analysis). In order to put results into perspective, the grey bar represents results of the LCA looking at conventional fossil-based PAA production. Mass stands for mass allocation, econ for allocation based on the economic value of the unit process outputs.

Table 5-6 Sensitivity of the results of the softwood-based LCA to changes in allocation assumptions (i.e. mass vs. economic allocation for outputs of the sugar extraction and fermentation unit processes).

Impact category	Unit	mass/mass (Baseline)	econ/mass	mass/econ	econ/econ	PAA
GWP 100a	kg CO2 eq	1.321E+00	1.311E+00	1.471E+00	1.457E+00	2.738E+00
CED (fossil/nuclear)	MJ eq	1.499E+01	1.474E+01	1.764E+01	1.730E+01	7.058E+01
Acidification	H+ moles eq	3.781E-01	3.738E-01	4.388E-01	4.329E-01	4.525E-01
Eutrophication	kg N eq	9.659E-03	9.654E-03	9.698E-03	9.691E-03	4.253E-04
Water use	m3	7.506E-03	7.445E-03	9.967E-03	9.882E-03	4.784E-03
Land occupation	m2a	8.412E+00	7.801E+00	1.084E+01	1.001E+01	2.435E-02

Mass/Mass: Allocation for products/byproducts of both unit processes is based on mass; Econ/Mass: Economic allocation is applied to outputs of the sugar extraction process, while mass allocation is applied to the outputs of fermentation; Mass/Econ: Mass allocation is applied to sugar extraction and economic allocation to fermentation; Econ/Econ: Economic allocation is applied to all products/byproducts.

Results of the analysis indicate that applying economic allocation to the extraction process results only in a minimal decrease in environmental burdens. This decrease is most distinct for impacts related to land occupation as upstream environmental burdens from softwood growth and harvest are allocated more towards pulpwood and lignin product outputs if economic heating values are taken as basis for allocation.

Allocating energy and material flows associated with outputs of the fermentation process by economic value leads to higher system-wide environmental burdens per kg of polymer produced for all impact categories when compared to the initial analysis in which only mass allocation was applied. The increase is largest for impacts to acidification (0.38 (mass/mass) vs. 0.44 H+ moles-eq (mass/econ)), water use (0.00751 (mass/mass) vs. 0.00997 m3 (mass/econ)), and land occupation (8.4 (mass/mass) vs. 10.8 m2a (mass/econ)). This is due to the fact that more of the upstream environmental burdens of sugar extraction and softwood harvest & growth are now allocated to the PIA polymer.

Applying economic allocation to both multi-output processes (sugar extraction and fermentation) results in only a slight increase in environmental burdens to GWP (1.46 kg CO<sub>2</sub>-eq), CED (17.3 MJ-eq), and eutrophication (0.00696 kg N-eq), while increases in impacts to acidification (0.043 H+ moles-eq), water use (0.0099 m3), and land occupation (10.1 m2a) are more pronounced. Under such a scenario impacts to acidification may be almost as high as acidification impacts from conventional fossilbased PAA production (0.452 H+ moles-eq). Eutrophication, water use, and land occupation are still higher for wood-based PIA production when compared to conventional PAA production. However, fossil CED and GWP associated with PIA production are still significantly lower when compared to its fossil-based counterpart. Relative contributions to each impact category of the softwood-based PIA lifecycle applying economic allocation to multi-output processes are shown in Figure 5-15.



Figure 5-15 Relative contributions to each impact category of the softwood-based PIA lifecycle applying economic allocation (instead of mass allocation) to all multi-output processes (i.e. sugar extraction and fermentation).

The choice of allocation also affects results of the *corn-based PIA polymer*. The use of market prices for allocation of impacts to products from corn-wet milling (i.e. glucose, corn meal & feed, and corn-oil) does not significantly change overall results (Table 5-7). The reason is that the allocation percentages for corn-wet milling only change slightly with market prices.

Market prices of corn wet milling products	[\$/tonne]	Source	Allocation [%] based on mass	Allocation [%] based on economic value
Glucose (dextrose) from corn <sup>1</sup>	330.70	(Durant 2011)	69.40	69.07
Corn meal & feed	252.03	(Moreau et al. 2010)	26.23	19.90
Corn oil	838.42	(Moreau et al. 2010)	4.37	11.03
	1 5 /11			

Table 5-7 Average market prices (January 2011) and corresponding allocation factors for the products of corn wet milling.

Based on a dextrose price of \$0.15/lb.

On the other hand, applying economic allocation to the fermentation process leads to a 10-25% increase in environmental impacts to all impact categories of the corn-based PIA production route (Table 5-5). Detailed results of this sensitivity analysis are shown in Figure 5-16 and Table 5-8.



Figure 5-16 Sensitivity of the results of the corn-based LCA to changes in allocation assumptions (i.e. mass vs. economic allocation for outputs of the corn wet milling and fermentation unit processes). The first bar shows results based solely on allocation by mass (see results shown in the previous sections for a detailed contribution analysis). In order to put results into perspective, the grey bar represents results of the LCA looking at conventional fossil-based PAA production. Mass stands for mass allocation, econ for allocation based on the economic value of the unit process outputs.

Table 5-8 Sensitivity of the results of the corn-based LCA to changes in allocation assumptions (i.e. mass vs. economic allocation for outputs of the corn wet milling and fermentation unit processes).

Impact category	Unit	mass/mass (Baseline)	econ/mass	mass/econ	econ/econ	РАА
GWP 100a	kg CO2 eq	2.193E+00	2.189E+00	2.665E+00	2.659E+00	2.738E+00
CED (fossil/nuclear)	MJ eq	2.480E+01	2.474E+01	3.107E+01	3.099E+01	7.058E+01
Acidification	H+ moles eq	7.264E-01	7.245E-01	9.160E-01	9.135E-01	4.525E-01
Eutrophication	kg N eq	1.642E-02	1.639E-02	1.896E-02	1.892E-02	4.253E-04
Water use	m3	1.229E-02	1.226E-02	1.652E-02	1.648E-02	4.784E-03
Land occupation	m2a	2.457E+00	2.454E+00	2.685E+00	2.681E+00	2.435E-02

Mass/Mass: Allocation for products/byproducts of both unit processes is based on mass; Econ/Mass: Economic allocation is applied to outputs of the corn wet milling process, while mass allocation is applied to the outputs of fermentation; Mass/Econ: Mass allocation is applied to corn wet milling and economic allocation to fermentation; Econ/Econ: Economic allocation is applied to all products/byproducts.

Relative contributions to each impact category of the softwood-based PIA lifecycle applying economic allocation to multi-output processes are shown in Figure 5-17.



Figure 5-17 Relative contributions to each impact category of the corn-based PIA lifecycle applying economic allocation (instead of mass allocation) to all multi-output processes (i.e. corn wet milling (sugar extraction) and fermentation).

In *conclusion*, applying economic allocation to the unit processes of sugar extraction and fermentation leads to slightly higher environmental burdens associated with the production of 1kg of PIA at the factory gate when compared to mass allocation. However, even if allocation is based to 100% on market prices (econ/econ), impacts to GWP, CED, and acidification associated with softwood-based PIA production are still lower than production of an equivalent amount of polymer via fossil-based or corn-based routes. Nevertheless, with regards to eutrophication, water use, and land occupation fossil based PAA production still results in the lowest system-wide environmental impacts. A summary is shown in the following table.

Table 5-9 System-wide environmental burdens associated with the production of 1 kg PIA from softwood and corn applying economic allocation to all multi-output processes and in comparison to fossil-based PAA.

Impact category	Unit	Softwood- based PIA (econ/econ)	Corn-based PIA (econ/econ)	Fossil-based PAA	
IPCC GWP 100a	kg CO2 eq	医肠管视镜 化链门 计	2.659E+00		
CED (fossil/nuclear)	MJ eq	ા અંદરકાર્ટ સંશો	3.099E+01		
Acidification	H+ moles eq	4. 17611 M		4.525E-01	
Eutrophication	kg N eq	9.691E-03		4431:01	
Water use	m3	9.882E-03			
Land occupation	m2a		2.681E+00	· ===\$1=\$1/	

The route with the lowest impact is shown in green color, while the route with the highest impact is shown in red.

#### C.2. Choice of Energy Mix

The sensitivity of the choice of direct electricity inputs to processes of the biorefinery (i.e. sugar extraction, fermentation, recovery, and polymerization) is investigated by comparing results of the LCA study, using U.S. Northeast (NE) power grid inputs, to two different scenarios using either the U.S. average grid or 100% hydropower. These account for the fact that the biorefinery might be located in another part of the United States, or might use a renewable energy mix to power its processes in the near future<sup>75</sup>. Results of the analysis for *softwood-based PIA* are shown in the following figure. Quantitative results are shown in the following table.

<sup>&</sup>lt;sup>75</sup> Itaconix indicated that the commercial plant may be located in close proximity to a hydropower dam which could supply electricity inputs to the biorefinery system.



Figure 5-18 Results of the sensitivity analysis for softwood-based PIA investigating the choice of electricity inputs on the LCA results. Allocation is based completely on the mass. Results for fossil-based PAA (grey bars) are shown for comparison.

Impact category	Unit	US Northeast (Baseline)	US average	Hydropower	РАА
IPCC GWP 100a	kg CO2 eq	1.321E+00	1.482E+00	9.568E-01	2.738E+00
CED (fossil/nuclear)	MJ eq	1.499E+01	1.784E+01	9.402E+00	7.058E+01
Acidification	H+ moles eq	3.781E-01	4.182E-01	2.322E-01	4.525E-01
Eutrophication	kg N eq	9.659E-03	1.163E-02	9.605E-03	4.253E-04
Water use	m3	7.506E-03	8.704E-03	7.242E-03	4.784E-03
Land occupation	m2a	8.412E+00	8.419E+00	8.409E+00	2.435E-02

Table 5-10 Results of the sensitivity analysis for softwood-based PIA investigating the choice of electricity inputs on the LCA results.

The functional unit is 1kg of polymer at the factory gate. Allocation is based on mass. Results for fossilbased PAA are shown for comparison.

Results of the analysis show that the use of hydropower, having the lowest carbon intensity, reduces GWP to 0.96 kg  $CO_2$ -eq and CED to 9.4 MJ-eq. Overall, the use of hydropower has the potential to reduce impacts to all categories (reductions in environmental burdens to eutrophication and land occupation are minimal), while using

U.S. average power would lead to the highest impacts to all categories. However, this is least pronounced land occupation impacts as the majority of land takes place during feedstock growth which is relatively independent from energy use. Eutrophication impacts of the softwood-based LCA are mainly influenced by off-site treatment of effluent originating from biorefinery process which is relatively independent of electricity mix choices made.

Figure 5-19 and Table 5-11 show results of the sensitivity analysis for the *corn*based life cycle.



Figure 5-19 Results of the sensitivity analysis for corn-based PIA investigating the choice of electricity inputs on the LCA results. The functional unit is 1kg of polymer at the factory gate. Allocation is based completely on the mass. Results for fossil-based PAA are shown for comparison.

Impact category	Unit	US Northeast	US average	Hydropower	PAA
IPCC GWP 100a	kg CO2 eq	2.193E+00	2.605E+00	1.272E+00	2.738E+00
CED (fossil/nuclear)	MJ eq	2.480E+01	3.210E+01	1.059E+01	7.058E+01
Acidification	H+ moles eq	7.264E-01	8.290E-01	3.549E-01	4.525E-01
Eutrophication	kg N eq	1.642E-02	2.147E-02	1.629E-02	4.253E-04
Water use	m3	1.229E-02	1.536E-02	1.173E-02	4.784E-03
Land occupation	m2a	2.457E+00	2.475E+00	2.449E+00	2.4350E-02

Table 5-11 Results of the sensitivity analysis for corn-based PIA investigating the choice of electricity inputs on the LCA results.

The functional unit is 1kg of polymer at the factory gate. Allocation is based completely on the mass. Results for fossil-based PAA are shown for comparison.

Results shown indicate a similar tendency with the highest impacts being found for the scenario using energy from the U.S. average power grid. For the impact category of GPW, the use of U.S. average power (2.61 kg  $CO_2$ -eq) leads to an environmental impact only slightly lower than fossil-based PAA production (2.74 kg  $CO_2$ -eq).

In *conclusion*, the choice of a less carbon intensive electricity mix (e.g. from hydropower) has the potential to lead to significantly lower impacts in particular on GWP and CED for both softwood- and corn-based biorefineries. However, the impacts to eutrophication, water use, and land occupation are mainly a result of direct inputs and/or emissions to the biorefinery system including feedstock growth/harvest. As a result, these impacts are less influenced by choices made with regards to the electricity inputs.

## **C.3. Varying Inputs of Activated Carbon**

Activated carbon (AC) used for the removal of chemical substances from the broth solution (sugar extraction and fermentation) was varied between 50% and 200% of

the baseline value<sup>76</sup> (100%). Results for both softwood- and corn-based PIA are shown in the following figures and tables.



Figure 5-20 Sensitivity of varying amounts of activated carbon (AC) on the environmental impact results for *softwood-based PIA*. The functional unit is 1 kg of PIA at the factory gate.

Table 5-12 Sensitivity of varying amounts of activated carbon (AC) on the environmental impact results for softwood-based PIA.

Impact category	Unit	AC (50%)	AC (100%) Baseline	AC (200%)	РАА
IPCC GWP 100a	kg CO2 eq	1.094E+00	1.321E+00	1.776E+00	2.738E+00
CED (fossil/nuclear)	MJ eq	1.450E+01	1.499E+01	1.596E+01	7.058E+01
Acidification	H+ moles eq	3.647E-01	3.781E-01	4.050E-01	4.525E-01
Eutrophication	kg N eq	9.639E-03	9.659E-03	9.698E-03	4.253E-04
Water use	m3	7.341E-03	7.506E-03	7.838E-03	4.784E-03
Land occupation	m2a	7.457E+00	8.412E+00	1.032E+01	2.435E-02

<sup>&</sup>lt;sup>76</sup> The total quantity of AC used is not given due to confidentiality of the data.



Figure 5-21 Sensitivity of varying amounts of activated carbon (AC) on the environmental impact results for *corn-based PIA*. The functional unit is 1 kg of PIA at the factory gate.

Impact category	Unit	AC (2%)	AC (4%) Baseline	AC (8%)	РАА
IPCC GWP 100a	kg CO2 eq	1.975E+00	2.193E+00	2.630E+00	2.738E+00
CED (fossil/nuclear)	MJ eq	2.433E+01	2.480E+01	2.573E+01	7.058E+01
Acidification	H+ moles eq	7.135E-01	7.264E-01	7.522E-01	4.525E-01
Eutrophication	kg N eq	1.640E-02	1.642E-02	1.646E-02	4.253E-04
Water use	m3	1.213E-02	1.229E-02	1.261E-02	4.784E-03
Land occupation	m2a	1.539E+00	2.457E+00	4.294E+00	2.435E-02

Table 5-13 Sensitivity of varying amounts of activated carbon (AC) on the environmental impact results for corn-based PIA.

Results show that the impact categories most significantly being influenced by varying amounts of AC used are GWP as well as land occupation. Optimizing the amounts of AC used during sugar extraction (only softwood-based process) and recovery (both softwood and corn-based biorefineries) therefore has the potential to lead to reduced impacts with regards to these impact categories. For example, halving overall AC use from (from 100% (baseline) to 50%) could lead to a reduction of roughly 17% in GWP for the softwood-based biorefinery (1.32 to 1.09 kg  $CO_2$ -eq) and 10% for the cornbased life cycle (2.19 to 1.98 kg  $CO_2$ -eq). Similarly, halving AC inputs would decrease system-wide land occupation by 11% from 8.4 to 7.5 m2a/kg softwood-based PIA and by 37% from 2.5 to 1.5 m2a/kg corn-based PIA, respectively.

## **CHAPTER 6**

#### CONCLUSIONS AND FUTURE RESEARCH

## A. Conclusions

#### A.1. Results

This dissertation assessed various technologies capable of utilizing organic waste and forestry residuals for the generation of chemical feedstock. Using life-cycle assessment, environmental hot-spots within the supply chain could be highlighted and results compared to currently operating fossil-based production systems and conventional waste management schemes.

The generation of a clean syngas from various mixed waste streams, either for subsequent chemical synthesis (carbon recycling) or power production (plasma gasification), seems to be possible. Sugar extraction from softwood in an integrated biorefinery and subsequent fermentation into PIA has been successfully proven in first lab- and pilot-tests and is currently repeated at larger scale by Itaconix LLC.

Carbon recycling systems, in which organic waste is recycled into naphtha for chemical feedstock production, was found to have the potential to lead to an overall reduction in environmental burdens when compared to conventional waste landfilling. In the future, power offset by waste-to-energy systems may not be as carbon- and resourceintensive (per kWh electricity generated) as today, and carbon recycling schemes (which may operate at higher conversion efficiencies as indicated in the sensitivity analysis in chapter 3) may then also become increasingly attractive from an environmental perspective to conventional incineration systems. While production cost of Fischer-Tropsch derived chemicals seems not yet competitive to fossil-based chemicals provision<sup>77</sup>, future price increases in global oil prices as well as changes in waste tipping fees, and efficiency gains on site of the waste conversion systems, may alter the economics and allow carbon recycling routes to reach a price competitive to fossil-based production routes.

Plasma gasification in waste-to-energy applications was introduced several decades ago with the aim to enhance landfill diversion while providing renewable energy. However, a lack of transparent information, with regards to the technologies' potential system-wide environmental burdens and process performance, have so far served as obstacle towards commercialization. This study aimed to bridge this gap by using pilot scale data in a LCA model to estimate environmental impacts associated with waste plasma gasification for electricity production, identify the environmental 'hot-spots' along the supply chain, and make recommendations for future improvement. While environmental impacts were found to be roughly similar to conventional fossil-based power systems (per kWh of electricity generated), process optimization with respect to

<sup>&</sup>lt;sup>77</sup> The ethylene route from natural gas is a highly optimized route that has been operating and been improved for several decades.

coal co-gasified, coke used as gasifier bed material, and fuel oil co-combusted in the steam boiler, would allow to significantly lower the system-wide environmental burdens. In addition, by producing a clean syngas, plasma gasification may be further advanced in the future to allow fuels, chemicals, and polymer provision via various catalytic pathways.

*Biochemical production of PIA* polymers from hemicellulose extracted from softwood via a stream integrated approach (with the partially macerated wood and lignin being used in other existing processes such as pulp & paper plants for conventional pulp and bioenergy production) represents an innovative approach of potential interest for biorefineries in the U.S. Northeast and a future bio-economy in general. Using lab- and pilot scale data, the assessment indicated lower global warming potential, energy demand, and acidification, for the wood-based PIA polymer, when compared to corn-based PIA and fossil-based PAA. However, water use associated with wood-derived PIA was found to be higher than fossil-based PAA production and land occupation is highest for the woodderived polymer.

All three designs, i.e. gasification/FTS, plasma gasification/steam turbine, and fermentation/PIA production, represent a combination of existing technologies that are commercially available today (though not in an integrated fashion) and data from pilot plants and process models. Uncertainties associated with our results, derived from a combination of existing uncertainty information, semi-quantitative approaches, and sensitivity analysis, are large and may differ from those obtained in more detailed engineering design studies. Hence, absolute results of our study should be used with

caution and many uncertainties will remain until the actual operation of similar integrated systems. Nevertheless, our study was able to indicate the sources of large uncertainties, parameters of greatest sensitivity, and identify the subsystems of the life-cycles responsible for the highest environmental burdens (and costs for FTS systems).

The systems investigated are capable of recycling a certain fraction of the feedstock carbon (organic waste/forest biomass) for chemical feedstock provision and/or electricity recovery. Chemicals obtained by gasification/FTS and fermentation/PIA production may serve as feedstock for subsequent polymer provision. In the current system configuration, the carbon efficiency<sup>78</sup> towards chemical feedstock is roughly 24% (38% at higher LHV conversion efficiencies) for FTS/gasification (MTCI system) (Figure 6-1). Numbers for PIA provision are not shown due to data confidentiality. Syngas generated during plasma gasification could serve as intermediate towards chemical feedstock provision. However, in our study energy recovery via syngas combustion and using a steam turbine takes place during which feedstock carbon is released back to the atmosphere. Carbon efficiencies in particular with regards to thermochemical conversion systems may be further increased e.g. by applying pressurized gasification systems and minimizing heat and carbon losses from the gasification reactors and FTS systems (see chapters 4 and 5 for detailed discussions).

<sup>&</sup>lt;sup>78</sup> Carbon conversion efficiency = Chemical Feedstock [Kg<sub>carbon out</sub>]/Waste Feedstock [kg<sub>carbon in</sub>] x 100%.



Figure 6-1 Indicative carbon balance for the three systems investigated. The FTS system (Chapter 3 - MTCI system shown) is capable of capturing roughly 24% of the feedstock carbon as FT liquids (38% at higher conversion efficiencies (50 % LHV)). Due to data confidentiality, no numbers are shown for PIA production (Chapter 5 – pulpwood-based route shown). In its current system configuration, plasma gasification (Chapter 4 – Route 7-Bio/CDDB shown) is designed to recover energy from fossil and biogenic feedstock, hence immediately releasing the carbon back to the atmosphere.

#### A.2. Contributions

This project involved extensive data collection and development of life-cycle assessment models and cost analysis to investigate the potentials of emerging technologies to utilize organic waste feedstock or forest byproducts for the generation of chemical feedstock<sup>79</sup>. The unique contributions of this work include:

- Detailed environmental assessments and transparent inventory data of carbon recycling routes that may foster understanding with regards to their potential contributions to sustainable resource use and enhanced materials recycling in the future (Chapter 2 and 3);
- Identifying key capital and operating costs, as well as significant sensitivities of the ethylene production cost via carbon recycling schemes, that may help to speed the commercialization of similar systems (Chapter 3);
- First system-wide environmental assessment of 1.) Plasma gasification as a wasteto-energy alternative in the United States, and 2.) PIA production from NE softwood biomass as alternative to corn-derived and fossil-based polymers, and transparent and detailed information on hot-spots along the supply chain, sensitivities of results, and potentials for improvement for both systems (Chapter 4 and 5);

These contributions are hoped to provide valuable information to: 1.) Waste managers and decision makers working on waste and biomass policies, 2.) Businesses and

<sup>&</sup>lt;sup>79</sup> Syngas generated by the plasma gasification system (Chapter 4) is seen as 'chemical feedstock' in this context, as it would, besides power production in a steam turbine, allow the generation of platform chemicals via catalytic pathways similar to other thermochemical systems assessed in this dissertation.

companies considering an investment in the biomass/waste management sector, 3.) Petrochemical industry, and 4.) Resource managers and the academic community.

Transparent information on the environmental consequences associated with conversion technologies is important to gain interest from stakeholders interested in the system-wide potentials and limitations of similar systems. Currently, investing in thermochemical conversion systems in the United States is bound to a high risk and the payback on investment has not been attractive enough. This is to a large extent due to uncertainties on the regulatory side (i.e. the permitting process) (J. Binder and Higgins 2008). Questions that arise in this context are for example what air emissions regulations will apply to thermochemical systems and how to categorize them (e.g. as recycling facility, thermo conversion facility, etc.), and whether those technologies should be eligible for diversion credits and/or renewable and alternative energy credits which could foster implementation<sup>80</sup>.

Furthermore, as fossil-resources are expected to become increasingly scarce and difficult to access over the course of the next decade, results of this study could be of interest to the petrochemical industry. Performance data on the different steps from the organic feedstock to the final polymer could provide valuable information on the prospects of these technologies for replacing conventional fossil-based processes.

Finally, with regards to academia, life cycle inventory data collected during this study could add to the existing LCI databases, which so far only provide limited

<sup>&</sup>lt;sup>80</sup> For example, C&D derived biomass planned to be used in Waste to Energy (WtE) plants in Massachusetts is currently not recognized as eligible biomass under the state's cap & trade regulations and consequently companies will have to take the risk and act as the frontrunner in applying for the eligibility of the feedstock.

information on many steps of the proposed conversion pathways. The research could provide a basis for discussion on the feasibility and economics of these technologies and could trigger further discussion around the topic of future sustainable waste management and biomass use and how this fits into a portfolio of other measures striving towards increased resource efficiency and recycling.

#### **B. Future Research**

The following projects have not been carried out as part of this dissertation but they present logical future work to obtain a more comprehensive and holistic picture of the conversion systems. There are numerable additional projects that could build upon the results of the dissertation, only a few selected are presented here.

#### **B.1. Advanced Sustainability Assessment**

While attributional life cycle assessment (ALCA), used in this dissertation, provides first insights into the system-wide environmental burdens associated with the investigated conversion systems, to more fully understand their potential to contribute to sustainable development, advanced system-based metrics and modeling tools considering the three pillars of sustainability (environmental, social, economic) and interrelationships of variables, indicators and metrics over time should be applied.

In fact, results from the ALCA studies do not account for indirect impacts, e.g. occurring due to market forces (product substitution) outside the system boundaries (Ekvall 2000). In addition, life cycle thinking (i.e. environmental LCA, life cycle costing

(LCC), social LCA (SLCA) (Jørgensen et al. 2007)) represent steady-state methods only providing snapshots of impacts along the supply chain based on historical data (Halog and Manik 2011). However, for results to be most useful to decision and policy makers dynamic interrelationships over time are of importance.

Hence, a follow up study might include the development of an integrated systems model applying consequential LCA (CLCA) (Earles and Halog 2011; Ekvall 2000) and system dynamics (SD) (Halog and Manik 2011; Sterman 2001) for advanced sustainability assessment of carbon recycling routes in the United States.

#### **B.2. Additional Carbon Recycling Routes**

This dissertation only focused on a few selected polymer production routes. This was due to the limited inventory data and time available. However, theoretically both conversion platforms (thermochemical and biochemical) allow the production of a variety of base chemicals and subsequent plastic polymers from organic waste (Figure 6-2).



Figure 6-2 Schematic flow diagram showing the production pathways from organic waste into various plastics polymers. Platform chemicals are shown in shaded boxes and the associated polymer end products are shown in yellow boxes. PHAs (Polyhydroxyalkanoates) are produced directly via fermentation of carbon substrate within the microorganism and the compound is therefore shown as platform chemical and polymer end product (grey and yellow). PE: Polyethylene; PP: Polypropylene; PIA: Polyitaconic Acid; PLA: Polylactic Acid; PTT: Poly(trimethylene terephtalate); PURs: Polyurethanes; PBT: Poly(butylenes terephtalate); PBS: Poly(butylenes succinate); FDCA: 2,5-furandicarboxylic acid.

With more inventory data becoming available on each of those routes in the future, environmental assessment using LCA could be undertaken.

#### **B.3.** Policy Analysis: Barriers to Implementation

The hurdles for implementing emerging waste and biomass conversion technologies in the United States are diverse. In general, low landfill costs and sufficient space for landfilling (compared to EU and Japan) reduces pressures on the states to look for sustainable alternatives that would enhance landfill diversion. The lack of fiscal market distorters such as landfill and CO<sub>2</sub> taxes on the federal level result in less willingness to switch from fossil-based feedstocks for electricity, fuels and chemicals production to renewables including biomass and waste. Using interviews, statistics, and related literature, this study could present some of the main drivers and barriers to the introduction of conversion technologies into the market.

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