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Hydrothermal Gasification of Waste Biomass: Process Design and Life Cycle Asessment

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A process evaluation methodology is presented that incorporates flowsheet mass and energy balance modeling, heat and power integration, and life cycle assessment. Environmental impacts are determined by characterizing and weighting (using CO_2 equivalents, Eco-indicator 99, and Eco-scarcity) the flowsheet and inventory modeling results. The methodology is applied to a waste biomass to synthetic natural gas (SNG) conversion process involving a catalytic hydrothermal gasification step. Several scenarios are constructed for different Swiss biomass feedstocks and different scales depending on logistical choices: large-scale (155 MW_{SNG}) and small-scale (5.2 MW_{SNG}) scenarios for a manure feedstock and one scenario (35.6 MW_{SNG}) for a wood feedstock. Process modeling shows that 62% of the manure's lower heating value (LHV) is converted to SNG and 71% of wood's LHV is converted to SNG. Life cycle modeling shows that, for all processes, about 10% of fossil energy use is imbedded in the produced renewable SNG. Converting manure and replacing it, as a fertilizer, with the process mineral byproduct leads to reduced N_2O emissions and an improved environmental performance such as global warming potential: $-0.6\ kg_{CO_2eq}/MJ_{SNG}$ vs $-0.02\ kg_{CO_2eq}/MJ_{SNG}$ for wood scenarios.

1. Introduction

Historical and anticipated increases in primary energy consumption have amplified concerns about the environmental consequences of continued fossil fuel extraction and use. As a result, research in biofuels produced from energy crops and, more recently, ligno-cellulosic based biofuels produced from waste or residual biomass have aimed at substituting such fuels with so-called "carbon-neutral" fuels. Studies have questioned the sustainability and energetic efficiency of producing first generation biofuels (see, e.g., *1, 2*) and highlighted the advantages of second generation biofuels (*2*). Therefore, it is essential to develop methods capable of thoroughly assessing the environmental consequence of implementing a given process and of proposing methods to reduce negative consequences. The use of these methods is especially important in early evaluations of proposed sustainable fuel production systems. This article presents a methodology capable of assessing and reducing environmental effects during early design stages and illustrates its use for renewable SNG production in the Swiss context.

1.1. Conceptual Process Design and Life Cycle Assessment. Conceptual process design has traditionally been used to optimize process configurations—i.e., a list of interconnected equipment with defined sizes and operating conditions—using cost and efficiencies as objectives (*3*). Life cycle assessment (LCA) allows evaluation of the environmental performance of a process and its entire life cycle, but has mainly been used to compare process options. Only a few authors report the integration of LCA in process design methods. For example, Keoleian (*4*) and Nielsen and Wenzel (*5*) present stepwise methodologies for environmentally integrated process design. These authors indicate, directly or through examples, that energy efficiency and energy use constitute important aspects of environmental impact. Nevertheless, a specific methodology to address these issues is not presented. Process design and optimization using life cycle assessment are treated by Alexander et al. (*6*) and Azapagic (*7*). These authors suggest the need for aggregation of economic and environmental objectives during the optimization procedure. However, Azapagic acknowledges that such aggregation is often "controversial" (*7*). The present work avoids aggregation by optimizing the process on an economic basis, while also comparing these choices on an environmental basis. Specific applications of life cycle assessment to waste biomass to fuel conversion processes is also documented (*8–10*). None of these studies include heat and power integration even though heat recovery is identified as "essential to system efficiency and performance" (*8*). In addition, these studies do not address industrial ecology possibilities associated with biomass conversion when addressing the life cycle. Indeed, the partial revalorization of waste biomass into a fertilizer can have important environmental effects.

1.2. Hydrothermal Gasification of Waste Biomass. The hydrothermal gasification process evaluated upgrades biomass to SNG as an energy carrier while avoiding common issues of biomass conversion, which are linked to its energy intensive production and high moisture content. Given that methane is a gas essentially insoluble in water, the need to separate it from water using a distillation process is completely avoided, unlike the separation of ethanol from a water-rich fermentation product stream in the corn-grain to ethanol conversion process (*6*). A catalytic hydrothermal gasification process was developed at the Paul Scherrer Institut (PSI) in Villigen, Switzerland that allows for the production of methane from woody biomass (*11–13*). This process is carried out in an aqueous system at conditions near or above the critical point of water: 647 K (374 °C) and 22.1 MPa. This process also avoids having to dry the starting product, thereby increasing energy efficiency, especially for wet biomass.

Additional research is being done to adapt this process for a manure feedstock. In salt-containing feedstocks such as manure, salts must be separated prior to catalytic

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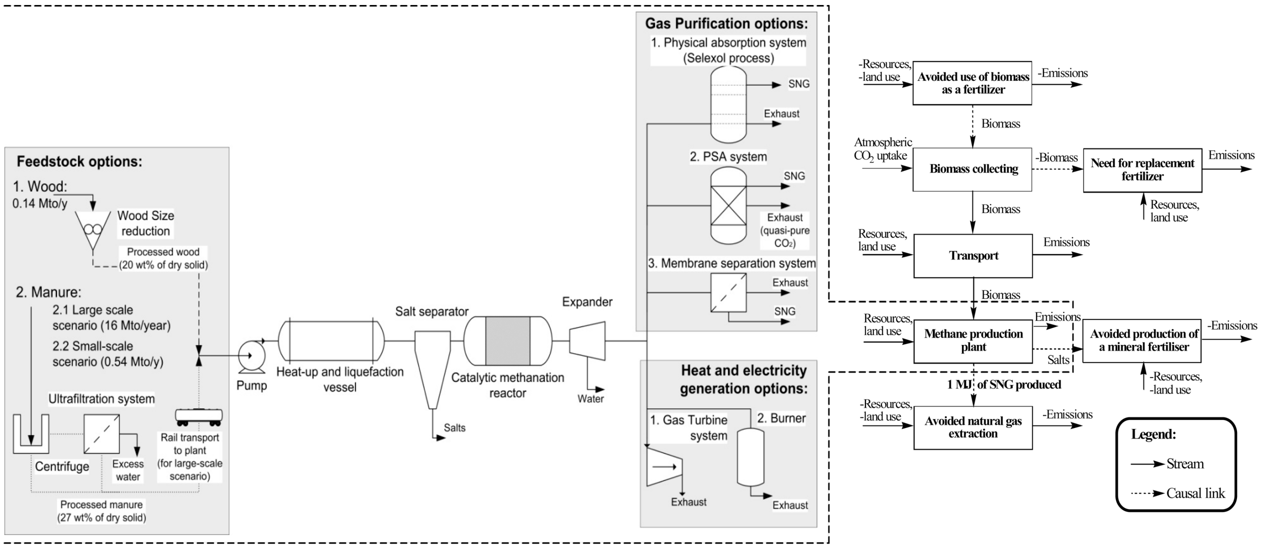


FIGURE 1. Overview of the flowsheet (left section) and life cycle inventory flow model (right section) used for the simulated biomass to methane conversion processes in this study. The flowsheet model constitutes one of the stages of the life cycle inventory model.

methanation to achieve long catalyst lifetimes. These salts can potentially be revalorized as a fertilizer. We selected both wood waste and manure for further study as they represent residual biomass and, thus, avoid an energy intensive agricultural production process.

2. Methodology

A multiscale modeling approach is taken using flowsheet models, energy integration and cost evaluation. These models constitute the core of a life cycle inventory model. This methodology includes several important aspects of sustainable energy and sustainable development in process design: energy efficiency, industrial ecology, and life cycle assessment.

2.1. Process Modeling and Heat Integration. Thermo-economic process evaluation follows the method presented by Gassner et al. (*14*). The Aspen plus 2004.1 flowsheet modeling software was developed by AspenTech (*15*) to simulate processes and their different options. The process minimum energy requirement (MER) is determined with combined heat and power integration (*16*) by extracting process stream data from the flowsheet modeling software. Once properly integrated, the optimal process is chosen based on economic criteria. Information transfer is accomplished using the OSMOSE (*17*) framework software, which was developed by the Industrial Energy Systems Laboratory of the Swiss Federal Institute of Technology in Lausanne.

Two scales of biomass conversion systems are analyzed based on the availability and delivery possibilities of the biomass feedstock in Switzerland (more details are available in the Supporting Information). A large-scale scenario assumes central processing and an average transport distance of 83 km by rail and 5 km by tractor and covers the majority of Switzerland's accessible agricultural density (according to manure availability modeling: see Supporting Information for more details). A small-scale scenario involving only tractor transport over a maximum distance of 10 km yields a feedstock of 0.54 million tons of wet manure per year. A wood scenario, by analogy with the study on conventional wood gasification and methanation by Felder et al. (*9*), assumes truck transport over 25 km, delivering wood chips with a 50 wt% humidity content to a plant and treating 136,220 tons of these wet chips every year.

The feedstock enters the process as illustrated by the left section of Figure 1. It must be processed to obtain a slurry of particles of about 0.2 mm diameter, with 20 wt% of heterogeneous solid (considered the upper limit for satisfactory pumping). For manure, this corresponds to a dry solid content of 27 wt% because a fraction of the dry solid dissolves in water. The resulting slurry is then pressurized to 30 MPa and heated to above the critical temperature of water. This heat-up leads to the decomposition of the larger organic molecules present in the biomass, the precipitation of inorganic salts (removed in the salt separator), and, in the methanation reactor, with the presence of a ruthenium catalyst, to the formation of a gaseous mixture of about 50 vol% methane, 50 vol% carbon dioxide, and a small amount of hydrogen (*11*). The salts must be separated prior to the catalytic stage because otherwise the catalyst will become quickly deactivated (*12, 18*). Following the catalytic hydrothermal gasification phase, the crude gas is split between a gas treatment stage and a heat and electricity generation stage. The former will ensure purification of the crude gas to SNG, acceptable for delivery to the Swiss grid (i.e., 50 bar with a minimum content of 95% methane (*19*)), and the latter generates the heat needed for the process plus additional power. We considered three possible gas treatment options: physical absorption in polyethylene glycol dimethyl ether (DMPEG), pressure swing adsorption (PSA), and membrane separation. Other gas purification options such as chemical absorption and cryogenic distillation were disregarded because of difficulties with high CO_2 partial pressures and high cost, respectively. As for heat and electricity generation, two options were considered: gas turbine or burner. A Rankine steam cycle was added for conversion of waste heat into additional electricity. However, for the smaller-scale manure conversion process, it was, a priori, considered not worthwhile to invest in a Rankine steam cycle for the revalorization of waste heat due to the small scale of this process variant.

2.2. Life Cycle Assessment. The process model described above forms the core of a larger model, accounting for the entire life cycle of the process from the harvesting of the biomass to the delivery of SNG to the Swiss natural gas grid. Linking process modeling to LCA allows for the systematic calculation of environmental impact when changing the process design. This life cycle inventory model uses data from the Ecoinvent database whenever possible (*20*). The inventory result is then used to calculate the life cycle's imbedded fossil energy and its environmental impact. This assessment is performed according to the guidelines given by the International Office of Standardization (ISO) for life cycle assessment (LCA) (*21*).

The goal of this LCA study is to identify the environmental hot-spots of the process and to evaluate its environmental performance with respect to its direct competitors. The competing technology for manure to methane conversion is assumed to be anaerobic digestion as described by Edelmann and Schleiss (*22*). The direct competitor for wood to methane conversion is considered to be conventional wood gasification and methanation as analyzed by Felder et al. (*9*). The right section in Figure 1 illustrates the system and its boundaries used for modeling all the biomass to methane conversion processes life cycle.

Since one goal of this study is to compare different routes for bio-SNG production, SNG is chosen as the final product. The use phase is disregarded, since SNG use is independent of the production process. The functional unit for this study is therefore *1 MJ of SNG brought to the Swiss network*.

The life cycle inventory (LCI) model outputs a list of environmental impact parameters (resource use, emissions, etc.) specific to the studied scenario. Life cycle impact assessment (LCIA) translates these results into environmental consequences to make them more comprehensible. This is done using three methods: the characterization of the environmental load into global warming potential according to IPCC 2001 (*23*), and weighting according to two methods: Eco-indicator 99 (*24*), and Eco-scarcity (*25*). The Eco-indicator method contains three submethods, which calculate an environmental impact according to a defined set of cultural values: Individualist (short-term perspective, human health is considered the major issue), Egalitarian (long-term perspective, ecosystem quality is weighted heavily and resource depletion is considered), and Hierarchist (in Ecoinvent: this corresponds to the average of the two other categories). The characterizing global warming potential (GWP) is chosen because it is a parameter of interest regarding biofuel production. The two weighting methods are chosen because they are well-known methods among LCA practitioners and for the applicability of Eco-scarcity to Switzerland. Indices from the Ecoinvent database (*20*) for these three methods are used.

3. Results and Discussion

3.1. Process Modeling. *Heat Integration.* Because part of the SNG is used for satisfying the heating load of the process, heat recovery has a major impact on process efficiency and environmental performance. The process composite curves, shown in Figure 2, are constructed by calculating the total process heat load at a given temperature. Pinch point analysis is used to compute maximum heat recovery. It defines the minimum energy requirement (MER), which is mainly a result of the heat-up phase prior to catalytic gasification and occurs close to 600 K (327 °C) for each process scenario (represented for the large-scale manure scenario in Figure 2). It coincides with the beginning of the endothermic biomass decomposition reaction. This creates an overall minimum heating requirement above 600 K (327 °C), which is met by cooling combustion fumes exiting the turbine at about 800 K (527 °C) or leaving a burner at about 2200 K (1927 °C) to the pinch point temperature. This heat exchange with combustion gases is represented on the composite curves in Figure 2 by the decrease of the process composite curve between 2200 K (1927 °C) and 600 K (327 °C). Process cooling requirements are due to the need to cool the different process streams to ambient conditions (298 K or 25 °C for liquids and 423 K or 150 °C for gases). Hot streams also include the combustion fumes used for heat generation, which are cooled from the pinch-point temperature down to the stack temperature (423 K or 150 °C). High waste heat temperatures enable mechanical work production prior to rejecting this heat to river water, which is heated from 285 to 289 K (12 to 16 °C). Revalorization of waste heat to electricity is done by integrating a steam Rankine cycle.

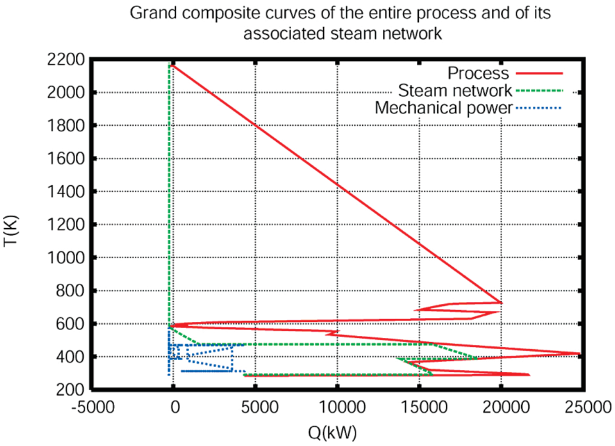


FIGURE 2. Integrated grand composite curve of the process (including its utilities) with its associated steam network (*16*) for the large-scale manure process using a burner for heat generation. The full line represents the grand composite curve of the process, the dashed line represents the steam network composite curve, and the dotted turbine schematics represent mechanical power production by the different streams of steam bleeding from the turbine (the basis of this schematic represents the amount of heat converted to power and the height of the schematic shows the temperature interval at which this conversion occurs). Each section of the grand composite curves is corrected by a minimum approach temperature difference depending on the nature of the phase. This temperature difference is set to 8 K for gaseous streams, 4 K for liquid streams, 2 K for evaporating or condensing streams, and 25 K for reacting streams.

Figure 3 demonstrates that the turbine option requires significantly more crude gas to fulfill the minimum heating requirements than the burner option, which reduces the net amount of produced methane. This is because the temperature of the turbine's exit gases is significantly lower than the temperature of the burner's flue gases. When a turbine is used, some of the combustion energy is used for producing mechanical and electrical work, and excess air at the turbine's inlet is used to keep its temperature below damaging levels. Comparing the energy balances for different starting products, one observes that the wood scenario produces more SNG in the case of the burner scenario and about the same amount of SNG in the case of the turbine scenario. In the first case, more organic material is available for wood conversion compared with manure, which has a higher inorganic content. Therefore, more SNG is produced with wood conversion despite its higher heating requirement (due to the higher water content in the feed). In the turbine scenario, the more important use of SNG for heating equalizes the two processes.

Thermo-Economic Analysis of Different Process Options. The three gas separation processes show very similar capital costs. In addition, they have virtually no influence on the process MER and all show a methane recovery ratio between 92 and 93%, which means that they do not strongly influence the amount of produced SNG and the sales revenue. These three options offer Wobbe indices that conform to Swiss regulations (14.5 kWh/Nm^3 for physical absorption, 14.0 kWh/Nm^3 for PSA, and 13.3 kWh/Nm^3 for membrane separation, which are all between 13.3 and 15.7 kWh/Nm^3 (*19*)). However, the resulting water content is of about 393 mg/Nm^3 for PSA and 72 mg/Nm^3 for membrane separation, both of which are above Swiss regulations (50 mg/Nm^3 (*19*)). Therefore, an end-of-pipe drying operation must be added. This is not the case for physical absorption in DMPEG, which

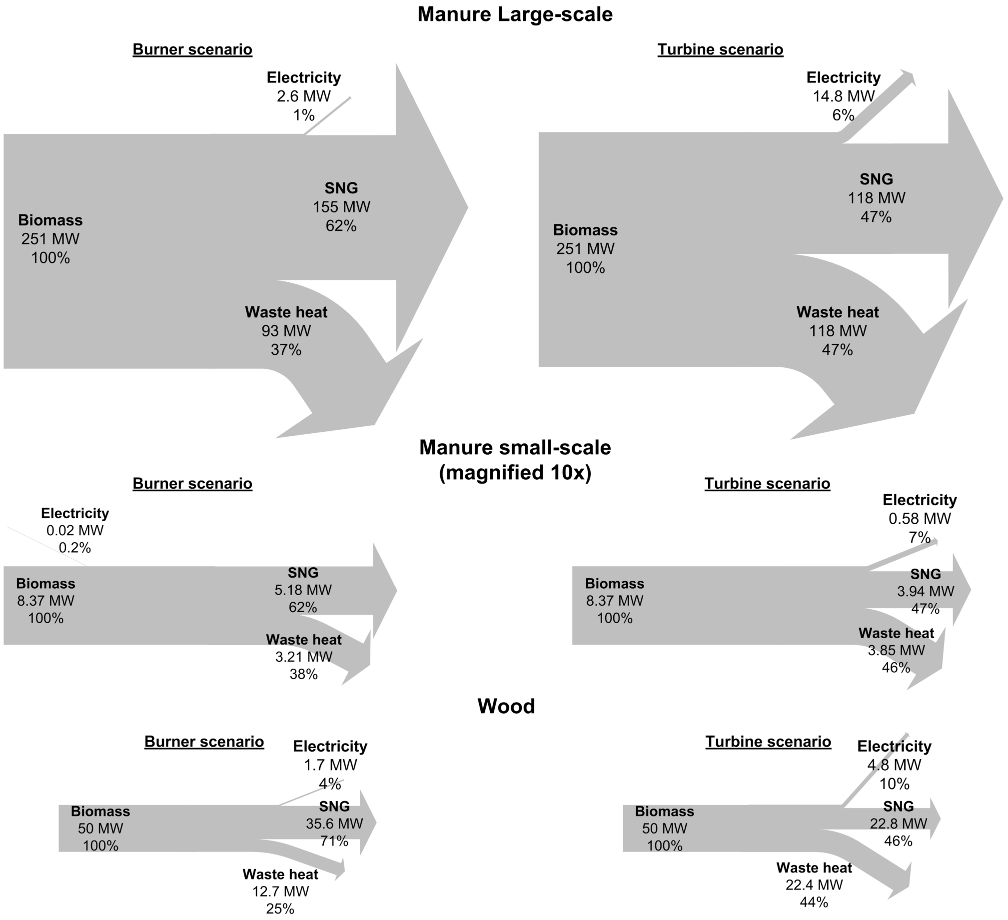


FIGURE 3. Overall energy balance for the different methane production processes. The energy entering and exiting the system is calculated on a Lower Heating Value (LHV) basis for biomass and SNG.

results in a water content close to zero. All separation options yield a methane volume fraction that is slightly too low. Revisions of the regulations are under discussion and, if the Wobbe index and the water content requirements are met, entering the grid with a volume fraction of methane of 94% or above should not be a problem even though current regulations require a methane content over 95% (*19*). Since the three processes are comparable from an economic standpoint, the physical absorption process is assumed to be the most advantageous because it avoids an additional drying process.

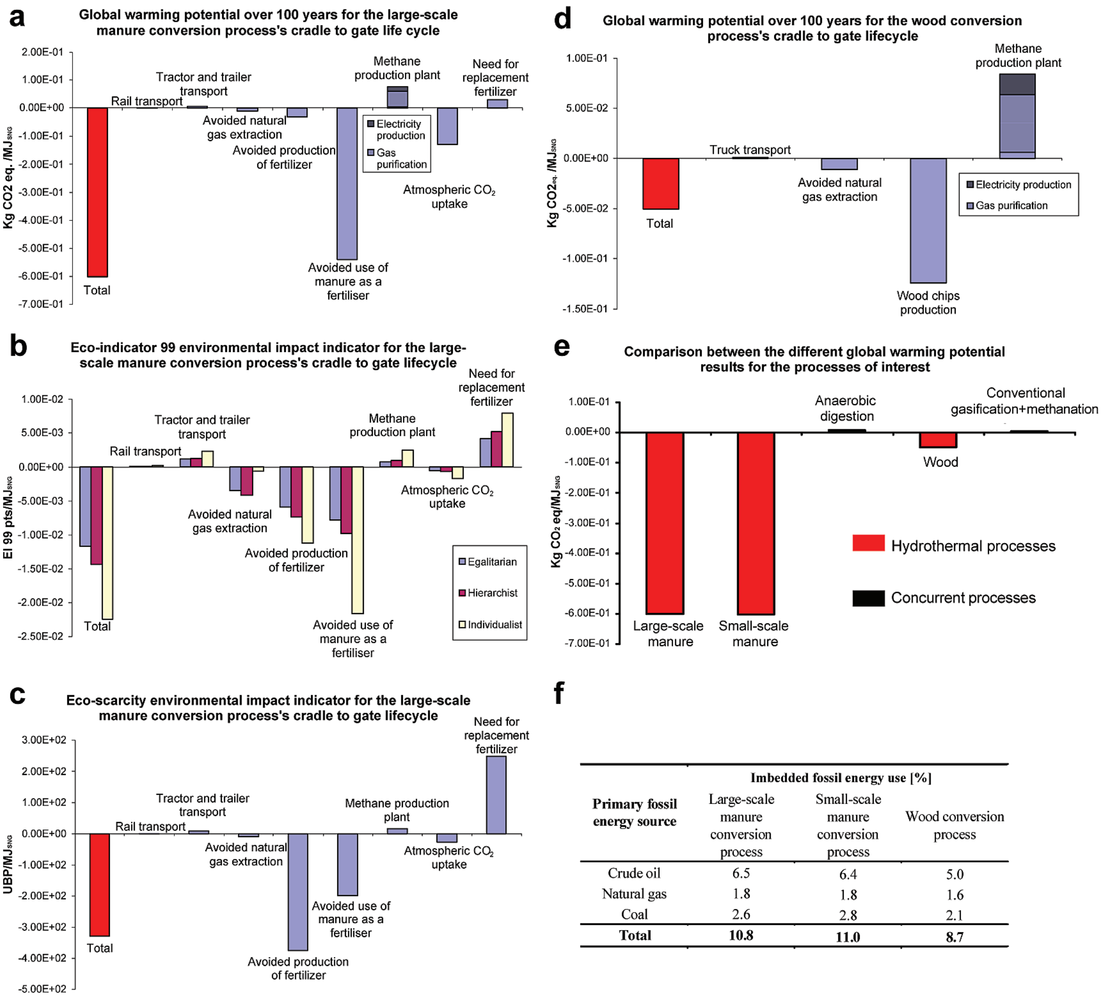
The most advantageous heat generation or heat and electricity cogeneration option is to use a simple burner because prices of methane and electricity (see Supporting Information) favor methane over electricity production in the Swiss context. The conclusion is similar when environmental impact is considered given the low CO_2-emitting electricity production in Switzerland.

In conclusion, the simulation of the catalytic hydrothermal gasification of different biomass feedstocks allowed the design of industrial-scale process configurations. Simulating scenarios with different availability and delivery options in the Swiss context allows the design and comparison of high efficiency integrated process configurations, which can be used for subsequent life cycle assessment.

3.2. Life Cycle Assessment. *Manure Processes.* As shown in Figure 4a, for each MJ of SNG produced, about 0.6 kg of CO_2 emissions are avoided (a 155 MW plant avoids 93 $kg_{eq.}$ of CO_2 emissions per second). Treating manure instead of spreading it avoids N_2O emissions, which account for 97% of the beneficial impact. The bar "atmospheric CO_2 uptake by manure" (Figure 4a) accounts for the renewable nature of organic carbon in manure and thus has a negative global warming potential (GWP). Transport of manure has a fairly small impact on GWP. Transport by rail proves to be much more environmentally benign compared to tractor transport given that the former contributes less to the GWP than transporting a cargo 8 times larger but over a distance 16 times shorter with a tractor. The production of inorganic salts as a byproduct of SNG avoids the production of mineral fertilizer. Given that this study estimates manure to be less efficient as a fertilizer than the salts, more inorganic fertilizer is produced than is needed to replace manure. For this reason, even though the "need for replacement fertilizer" activity contains the emissions of the spread fertilizer to the atmosphere, it contributes inversely to the GWP with respect to the "avoided production of replacement fertilizer" activity. The negative global warming potential of the avoided natural gas extraction is mostly due to avoided CO_2 (70%) and methane emissions (30%) that occur during extraction and delivery of methane.

The GWP impact of the methane production plant itself is small compared to the whole lifecycle, but is still the main emitter of gases contributing to GWP. The gas purification stage shows the largest contribution due to the emissions of carbon dioxide separated from the methane stream. Sequestering the separated carbon dioxide could reduce the GWP of the total system, but has not been investigated in this study.

The majority of the remaining impact of the manure conversion process results from the heat and electricity cogeneration activity, also due mostly to the carbon dioxide emitted during the combustion of methane in the burner. The remaining activities included in the methane production activity (production and delivery of catalyst, solvent, infrastructure, etc.) have a negligible contribution.



Primary fossil energy source	Imbedded fossil energy use [%]		
	Large-scale manure conversion process	Small-scale manure conversion process	Wood conversion process
Crude oil	6.5	6.4	5.0
Natural gas	1.8	1.8	1.6
Coal	2.6	2.8	2.1
Total	**10.8**	**11.0**	**8.7**

FIGURE 4. LCA results. Parts (a), (b), and (c) show the environmental impact of the large-scale manure conversion process as quantified by its global warming potential (a), Eco-indicator environmental impact indicator for 3 different weighting methods (b), and Eco-scarcity environmental impact indicator (c). The global warming potential is given in equivalent kg of CO_2, which translates the amount of emitted greenhouse gases into the equivalent amount of emitted CO_2 in terms of global warming potential. The Eco-indicator and Eco-scarcity methods quantify the environmental impact in the form of points. Part (d) shows the global warming potential for the wood scenario and part (e) compares this potential among all processes and their competitors. Part (f) shows the imbedded fossil energy use (nuclear fuel not included and renewable resources such as hydroelectric power, geothermal heat, and solar energy not included) for the life cycle of the different hydrothermal processes of interest.

Comparison of the three weighting perspectives for the Eco-indicator environmental impact results in Figure 4b shows that the individualist weighting perspective gives a better result for the whole process compared to its two counterparts, a consequence of heavier concern regarding GWP in this perspective. Even with these differences, the qualitative results of the two weighting methods (shown in Figure 4b and c) remain similar to those given by the GWP (given in Figure 4a).

Wood Process. As shown in Figure 4d, the global warming potential of methane production from wood chips is slightly negative. This is mainly due to wood chip production, which has a negative GWP because it includes the uptake of atmospheric carbon dioxide during wood growing. Given the similarity between the two processes, the wood-to-SNG production plant's impact has a magnitude very similar to that of the manure-to-SNG production plant. However, its relative contribution with respect to the other activities is much more important.

Comparison among Different Processes. Figure 4e shows that the two manure conversion scenarios are quasi-identical and therefore, only the results for large-scale processes are discussed. Both manure processes clearly show a greater environmental performance compared to the wood conversion process, even though Figure 3 showed the opposite concerning energy conversion efficiency. This is because manure itself has a direct global warming effect, mainly through N_2O emissions, while wood does not. Thus, the main advantage of the manure process is that, in addition to its gasification efficiency, it treats a form of environmentally problematic biomass. Anaerobic digestion has a larger total GWP than all other processes because manure and the biofertilizer are assumed identical in terms of nutrient-transfer-to-plant efficiency and emissions per unit nutrient. Therefore, there is no benefit from offsetting emissions due to replacing spread manure with the anaerobic sludge as replacement fertilizer. In addition, the anaerobic digestion methane production plant has a larger environmental load compared to the hydrothermal plant. This is due partly to increased process emissions during the fermentation step and partly to the infrastructure. The infrastructure's impact for anaerobic digestion is almost exclusively (over 99.9%) due to concrete use. Concrete has high-imbedded fossil CO_2 emissions due to its production process and transport. The two wood conversion processes are, considering the uncertainty linked to the modeling process, essentially identical. The small difference is due to greater process emissions during conventional wood gasification followed by catalytic methanation compared to the hydrothermal process.

According to Figure 4f, the imbedded fossil energy for the SNG from the investigated biomass conversion processes is between 8.7 and 11%. This is relatively small compared to processes that convert crops to fuels such as ethanol, where some results suggest that these processes consume more energy than they produce (if one includes all electrical input coming from sources like nuclear, hydroelectric, and geothermal in addition to fossil fuels) (*1*). Given the differences in boundary conditions, these numbers are not directly comparable; however, natural gas, alone, often accounts for more than 40% of imbedded fossil energy (*1*). By comparison, Figure 4f clearly shows that, since SNG is produced from waste or residual biomass, it does not require an energy intensive agricultural production process like the corn-grain to ethanol conversion process.

Thus, optimized flowsheet modeling results were successfully implemented as the core of a life cycle inventory model. The outcome demonstrates the value of coupling flowsheet modeling with combined heat and power integration models and life cycle assessment to evaluate and select more sustainable process variants at an early stage. Life cycle assessment demonstrated that for all the scenarios, GHG emissions during gas treatment are a major environmental hot-spot for the methane production plant. End-of-pipe treatment as well as sequestration options should be further investigated to improve the environmental performance of the process.

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Supporting Information Available

Detailed explanation of the manure pick-up scenario modeling as well as the process, economic, and life cycle inventories modeling procedures for all processes. This material is available free of charge via the Internet at http://pubs.acs.org.

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