

METHANOL PRODUCTION VIA BLACK LIQUOR GASIFICATION WITH EXPANDED RAW MATERIAL BASE

Report from a project within the collaborative research program *Renewable transportation fuels and systems*

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PREFACE

This project has been carried out within the collaborative research program *Renewable transportation fuels and systems* (Förnybara drivmedel och system), Project no. 40759-1. The project has been financed by the Swedish Energy Agency and f3 – Swedish Knowledge Centre for Renewable Transportation Fuels.

f3 Swedish Knowledge Centre for Renewable Transportation Fuels is a networking organization which focuses on development of environmentally, economically and socially sustainable renewable fuels, and

- Provides a broad, scientifically based and trustworthy source of knowledge for industry, governments and public authorities
- Carries through system oriented research related to the entire renewable fuels value chain
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SUMMARY

Gasification of black liquor with downstream synthesis to biofuels in Kraft pulp mills shows advantages regarding energy efficiency and economic performance compared to combustion in a recovery boiler. The good performance is partly due to the strong catalytic effect of the black liquor that enhances gasification reactions. Experimental research has shown that the reactivity can be kept even if the black liquor is significantly diluted with a secondary feedstock. Therefore, as a way to increase the operation flexibility as well as the biofuel production capacity of a mill integrated gasifier, a secondary biomass feedstock could be blended into the black liquor and co-gasified.

The main aim of this work was to evaluate the techno-economic and greenhouse gas (GHG) performances of co-gasification of black liquor with various blends of pyrolysis liquid, crude glycerol and fermentation residues for production of biomethanol of two different qualities, grade AA and crude. Material and energy balances to evaluate the systems' techno-economic and greenhouse gas performances were obtained via process modelling, with input data mainly from pilot and lab-scale experiments.

All investigated cases showed good economic performance under the assumed conditions. The cases where crude glycerol was blended with black liquor showed the best economic performances. The crude methanol could currently be competitive with today's prices of fossil based methanol and the grade AA quality methanol to untaxed fossil gasoline. Each case also showed good energy performance with high energy efficiencies. Blending feedstocks with black liquor showed positive influences on the cold gas efficiency and methanol production efficiency due to the catalytic effect of the black liquor. All co-gasification cases also showed higher efficiencies than that of gasification of pure black liquor.

By calculating GHG emissions following the RED method, the methanol produced in all cases resulted in emissions savings of 82-94 % compared to the fossil fuel reference. Following the ISO standards, the results showed lower GHG emissions savings potentials in the range of 62-88 %.

As a consequence of the blending, the methanol production capacities became very large. In one of the glycerol cases (50-50 blend), the capacity exceeded 1000 MW methanol. In this case, the required annual glycerol volume exceeded the current total European glycerol consumption (crude and refined), putting the glycerol availability as a significant bottle-neck.

The general conclusions were that blending a secondary feedstock with black liquor for co-gasification was an energy efficient and cost effective pathway to convert the feedstock to methanol. The produced methanol, in all considered cases, fulfilled the currently required 60 % GHG emissions reduction target as compared to a fossil fuel reference according to RED.

From the results it was further hypothesized that co-gasification could be a pathway to make black liquor gasification with biofuel production economically feasible also when using only a partial stream of the total black liquor flow from a pulp mill, while also keeping the recovery boiler for processing the rest of the black liquor. This would lead to the potential dual benefit of increased pulp production from debottlenecking the recovery boiler, and significantly reduced technical risk compared to total replacement of the recovery boiler with black liquor gasification. While this concept has been outside the scope of this project to evaluate, it is recommended for further studies.

SAMMANFATTNING

Biodrivmedelsproduktion via svartlutsförgasning ger bättre energieffektivitet och ekonomisk prestanda jämfört med konventionell förbränning av svartlut i en sodapanna. Förgasningsprocessen är mycket energieffektiv, främst på grund av den höga reaktiviteten i svartlut, vilken beror av den katalytiska effekten av alkali som finns i höga koncentrationer. Detta kan utnyttjas för att utvidga bioråvarubasen genom att blanda in och samförgasa exempelvis pyrolysvätska, råglycerol och fermenteringsrester, som därigenom kan åka ”snålskjuts” på svartlutens höga reaktivitet. Detta innebär samtidigt en större driftflexibilitet och en potentiell stor ökning av biodrivmedelsproduktionskapaciteten.

Huvudsyftet med projektet var att klimatmässigt och teknoekonomiskt utvärdera samförgasning av svartlut med biprodukter från biokemisk drivmedelsproduktion (råglycerol och fermenteringsrester) samt pyrolysvätska från skogrester för produktion av två olika kvaliteter av biometanol (råmetanol och grade AA). Utvärderingarna baserades på mass- och energibalanser från processmodelleringar där en stor del erforderliga ingångsdata härrörde från experiment i pilot och labbskala.

Samtliga undersökta fall visade god ekonomisk prestanda under antagna förhållanden. De fall där råglycerol blandades med svartlut resulterade i bäst ekonomi. Råmetanol baserad på svartlut och råglycerol låg i nivå med dagens priser på fossilbaserad metanol och säljpriset för grade AA-metanol i nivå med beskattad fossil bensin. Samtliga fall visade också bra energiprestanda med hög energieffektivitet. Inblandning av andra bioråvaror i svartlut hade positiva inverkan på verkningsgraderna, bland annat på grund av svartlutens katalytiska effekt. Samtliga samförgasningsfall visade också högre effektivitet än vid förgasning av enbart svartlut.

Beräkningar av växthusgasprestandan för metanolen enligt RED-metoden resulterade i CO₂-utsläppsbesparingar i spannet 82-94 % jämfört med fossila drivmedel. Enligt ISO-standarderna visade resultaten något lägre besparingspotentialer i intervallet 62-88 %.

Ytterligare en konsekvens av samförgasningskonceptet var att produktionskapaciteterna blev mycket stora. I ett av glycerolfallen (50-50 blandning) översteg produktionskapaciteten 1000 MW metanol. Detta fall innebar dock att den totalt erforderliga årliga volymen av glycerol var större än den nuvarande totala europeiska glycerolförbrukningen (både rå och raffinerad), vilket gör glyceroltillgängligheten till en stor flaskhals.

De generella slutsatserna från projektet var att inblandning av en sekundär bioråvara i svartlut för samgasning var ett energieffektivt och kostnadseffektivt sätt att omvandla råvaran till metanol. Den producerade metanolen uppfyllde också i samtliga fall de aktuella kraven på 60 % reduktion av växthusgasutsläpp jämfört med den fossila referensen enligt nuvarande RED.

En viktig insikt var att samförgasning kan vara ett sätt att göra svartlutsförgasning med biodrivmedelsproduktion genomförbar genom att endast använda en delström av det totala svartlutflödet, samtidigt som sodapannan kan behållas. På detta sätt kan risker relaterade till brukets kemikalie- och energiåtervinning minskas väsentligt, eftersom sodapannan fortfarande är i drift och sköter det mesta av återvinningen. Samtidigt kan utökad massaproduktion möjliggöras genom avlastning av sodapannan, som i många bruk är en kapacitetsbegränsande process. Detta koncept har varit utanför ramen för detta projekt och rekommenderas därför för vidare studier.

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1 INTRODUCTION

This report summaries the most important results and conclusions from the project. Details concerning investigated cases, assessment methodologies, assumptions and more detailed results and discussions are presented in appendices 1-2 and Carvalho et al (2017).

Black liquor is a residue from pulp making and normally combusted in a recovery boiler to produce process steam to the pulp mill. One alternative to combustion of the black liquor is conversion via gasification for production of synthesis gas. The technology is well demonstrated in pilot scale and the next development step is likely large-scale commercialization. Gasifying the black liquor in an entrained flow gasifier results in a high quality synthesis gas well suited for biofuels production. The gasification process becomes very energy efficient, mainly due to the high reactivity in black liquor caused by the catalytic effect of alkali present in high concentrations (Jafri et.al, 2016). Techno-economic analyzes have shown competitive production costs compared to other technology routes (Landälv & Waldheim, 2017).

The black liquor available for conversion is however determined by the pulp production of the mill and therefore limited, consequently it is also limiting for the biofuel production capacity. One way to increase the capacity and at the same time increase the operation flexibility of the biofuel plant is to blend with a secondary feedstock and co-gasify the blend. Previous research has shown that black liquor can be blended with other feedstocks in well-designed mixing processes while still maintaining the high reactivity. For example, the reactivity of a blend of black liquor and pyrolysis liquids is very close to that of pure black liquor (Bach-Oller et.al, 2015; Jafri et.al, 2017).

Techno-economic assessments have shown that co-gasification of black liquor with pyrolysis liquids combined with downstream methanol production can reduce the production costs compared to pure black liquor gasification due to economies of scale and high conversion efficiency. Co-gasification with 20–50 % addition of pyrolysis liquids on mass basis was found to be the most advantageous solution for smaller pulp mills with capacities below 200 kADt/y¹. Pure black liquor gasification was the most advantageous alternative for larger capacity pulp mills. The economic outcome however depends on the future market price of the pyrolysis liquid, which is highly uncertain (Andersson et al., 2015).

It is therefore of great interest also to investigate blending with other types of renewable feedstocks with potentially lower costs, for example residues from biochemical conversion of biomass feedstocks. Ragauskas et al. (2014) emphasize the importance of an efficient utilization of such by-products, which is crucial for the development of biochemical conversion technologies.

By-products of interest are, amongst others, crude glycerol from biodiesel (FAME) production and lignin rich fermentation residues from lignocellulosic ethanol production. Production of FAME results in up to 10 % (w/w) crude glycerol as a by-product. Cellulose-based ethanol production generates a lignin rich fermentation residue (in the range of 15-30 % (w/w)) containing a significant portion of the original energy content of the feedstock. While co-gasification of black liquor with pyrolysis liquids has been successfully accomplished in lab as well as in pilot scale experiments, co-gasification of black liquor with crude glycerol and fermentation residues has so far only been

¹ Air-dried tons per year

done in lab scale (Kirtania et.al, 2017). The results from the experiments showed that char gasification rate was not decreased by glycerol addition, which indicates that the same temperature normally used for black liquor gasification can be used for co-gasification of crude glycerol and BL. However, no techno-economic assessments or evaluation of the greenhouse gas performance have to the authors' knowledge previously been reported.

1.1 AIM AND OBJECTIVES

The main aim of this work was to evaluate the techno-economic and greenhouse gas performances in order to assess the feasibility of producing biomethanol through co-gasification of black liquor with various blends of pyrolysis liquid, crude glycerol and fermentation residues. Additionally, the Swedish methanol production potential via the co-gasification concepts was assessed.

Biomethanol of two different qualities, grade AA and crude, were considered as final products. The former with a 99.8 % purity can be used as a fuel, fuel additive, energy storage or feedstock for production of other chemicals (Olah et.al, 2006), while the latter (approx. 90 % purity) is suitable as a marine diesel fuel (Winnes et.al,2015).

The main objectives were to

- Calculate the required selling prices of biomethanol to reach a certain Internal Rate of Return (IRR) of the two different quality grades, compared to the case of pure black liquor gasification, analyze the competitiveness, and carry out a sensitivity analysis of crucial parameters.
- Assess the energy and greenhouse gas performance from a well-to-tank perspective.
- Estimate the Swedish technical production potential of biomethanol via black liquor co-gasification with an expanded raw material base.

1.2 METHODS

Material and energy balances for the evaluated cases were obtained using different simulation tools (SIMGAS, Aspen Plus). The process-steam demand of the integrated pulp mill was covered by (i) surplus heat from the methanol plant and (ii) high pressure steam from combustion of falling bark and when necessary fuel biomass in the power boiler. The resulting balances were in turn used for the techno-economic analysis applying a differential or marginal basis, i.e. by comparing a stand-alone pulp mill to a mill with integrated black liquor gasification with downstream methanol production. The production capacity of the pulp mill was 700 kADt of kraft softwood pulp per year.

Figure 1 schematically illustrates the main material and energy flows for the mill integrated biofuel plant. The resulting material and energy balances were used to evaluate the systems' energy efficiencies (see Eq 1-5) as well as the economic and greenhouse gas performance. Table 1 shows used prices of feedstocks and commodities. The methods to calculate the required selling prices to reach a certain IRR are described in detail in Carvalho et al (2017).

Table 1. Prices of feedstocks and commodities.

Energy carrier	Unit	Price
Biomass feedstock ¹	[€/MWh]	20
Pyrolysis liquid ¹	[€/MWh]	42
Crude glycerol ²	[€/MWh]	16
Fermentation residues ³	[€/MWh]	20
Electricity ¹	[€/MWh]	57
NaOH ¹	[€/ton]	385

¹Andersson et.al (2016), ²Quispe et.al (2013), ³Assumed to be the same price as for biomass

The cold gas efficiency (CGE) was used to evaluate the performance of the gasifier. It is defined as the ratio between the thermal output of the product gas leaving the gasifier (E_{syngas}) (in MW) and the thermal input to the gasifier ($E_{\text{feedstock}}$), according to Eq. 1.

$$\text{CGE} = E_{\text{syngas}} / E_{\text{feedstock}} \times 100 [\%] \quad (\text{Eq. 1})$$

The methanol conversion efficiency (MCE) is defined as the energy efficiency of converting the feedstock to methanol. It was calculated according to Eq. 2.

$$\text{MCE} = E_{\text{methanol}} / E_{\text{feedstock}} \times 100 [\%] \quad (\text{Eq. 2})$$

where E is the thermal inputs and outputs (in MW) of the methanol production plant. The CGE and MCE were calculated using Sulphur-free higher heating values of the feedstocks and syngas, due to the fact that the Sulphur is either recovered in the smelt or removed in reduced form from the syngas in the acid gas recovery (Öhrman et al., 2012).

To illustrate the influence of adding a blend-in feedstock on the CGE and MCE, marginal efficiencies were calculated according to Eqs. 3 and 4

$$\text{Marginal CGE} = (E_{\text{BL_mix}} - E_{\text{Pure_BL}})_{\text{syngas}} / (E_{\text{BL_mix}} - E_{\text{Pure_BL}})_{\text{feedstocks}} \times 100 [\%] \quad (\text{Eq. 3})$$

$$\text{Marginal MCE} = (E_{\text{BL_mix}} - E_{\text{Pure_BL}})_{\text{methanol}} / (E_{\text{BL_mix}} - E_{\text{Pure_BL}})_{\text{feedstock}} \times 100 [\%] \quad (\text{Eq. 4})$$

As the black liquor supply is equal for all cases, the marginal efficiencies represent the actual conversion efficiencies of the blend-in feedstocks into syngas (Eq. 3) and methanol (Eq. 4).

The marginal overall system efficiency, $\Delta\eta$, is a measure of the potential efficiency improvements a stand-alone pulp mill could achieve by integrating a methanol production plant. It was calculated according to Eq. 5.

$$\Delta\eta = (\Delta E_{\text{pulp}} + \Delta E_{\text{methanol}}) / (\Delta E_{\text{pulp wood}} + \Delta E_{\text{fuel biomass}} + \Delta E_{\text{blend-in feedstock}} + \Delta E_{\text{electricity}}) \quad (\text{Eq. 5})$$

Here, the energy flows crossing the system boundaries of the integrated pulp mill were compared to the corresponding flows of a stand-alone mill. The systems boundaries were represented with dashed lines in Figure 1. The pulp wood and pulp production parameters, respectively $E_{\text{pulp wood}}$ and E_{pulp} , are also equal in both stand-alone and integrated pulp mills, and therefore the terms ΔE_{pulp} and ΔE_{pulp} cancel out. The blend-in feedstocks were assumed to be delivered to the gate. The pa-

parameter $\Delta E_{\text{electricity}}$ takes into account the loss in electricity production due to the integration, corresponding to the sum of the actual electricity requirement of the integrated pulp mill and the excess electricity sold to the grid in the stand-alone pulp mill.

The efficiencies have also been re-calculated to electrical equivalents, by the first law of thermodynamics. All energy carriers (methanol, feedstock, etc) were converted to their electricity equivalents according to the efficiency (η) of the best-available technologies known to the authors (See Table 4, Carvalho et.al 2017). Electricity equivalents are suitable indicators when comparing systems with mixed energy carriers, in order to acknowledge for the differences in energy quality of the carriers.

The greenhouse gas emissions of methanol were assessed using a life cycle assessment (LCA) approach which includes raw material production and processing in a well-to-tank perspective. The well-to-tank greenhouse gas emissions were assessed using two different approaches, in order to highlight the importance of methodological choices to life-cycle assessment results: (1) the life-cycle based calculation approach as required by the EU Renewable Energy Directive (RED) (European Parliament, 2008), and (2) the ISO standards for life cycle assessment, 14040 (ISO 2006a) and 14044 (ISO 2006b).

For each of the considered co-gasification case, comparisons were made to a reference case with alternative uses of blend-in feedstock, see Appendix 1. The used material balances are presented in Appendix 2 and in Carvalho et.al (2017).

One large uncertainty with the co-gasification concept is connected to the potentially available volumes of blend-in components. The future availabilities of crude glycerol and fermentation residues were not explicitly considered in this work and thus the feasibility of providing the required volumes was not assessed. For instance, the availability of crude glycerol for co-gasification will depend on the development of the biodiesel production as well as on potential other alternative uses of the glycerol. The future availability of glycerol and fermentation residues was, as mentioned, not estimated, but the resulting blend-in feedstocks volumes was discussed in the light of the current production and/or consumption of those. Furthermore, the assessment of the domestic production potentials took the ages of the Swedish recovery boilers into concern. It was here assumed that Swedish recovery boilers older than 30 years could potentially be replaced with black liquor co-gasification.

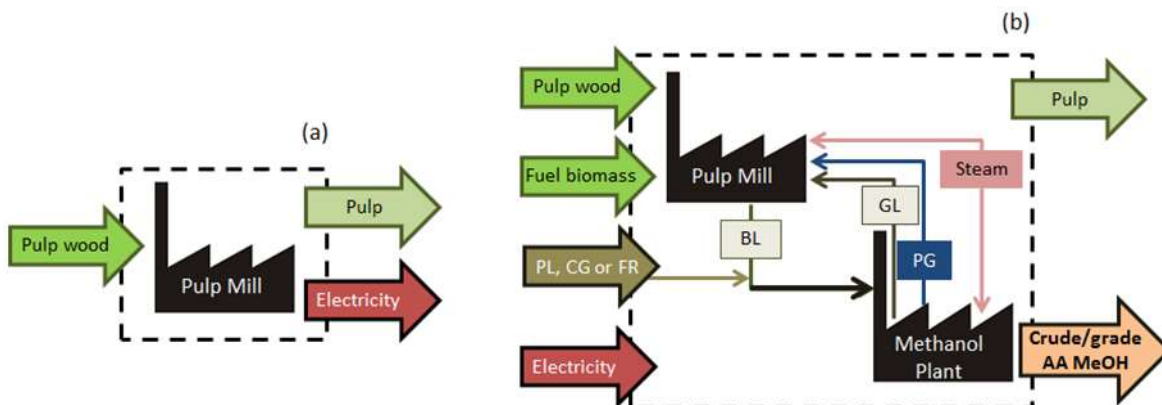


Figure 1. Schematics of a stand-alone (a) and an integrated pulp mill (b). BL: black liquor, GL: green liquor; PG: purge gas; PL: pyrolysis liquid; CG: crude glycerol and FR: fermentation residues.

2 RESULTS AND DISCUSSIONS

2.1 TECHNO-ECONOMIC ASSESSMENT

Figure 2 shows the resulting energy inputs and outputs for all the evaluated cases. By adding feedstock with a higher heating value to the fixed volume of black liquor the total energy input to the methanol plant increases drastically. Blending ratios of 20 % (by mass) added feedstock thus represents an increase in energy terms in the range of 47-53 % compared to pure black liquor. For the 50-50 blends, the energy inputs were up to three times higher than for pure black liquor.

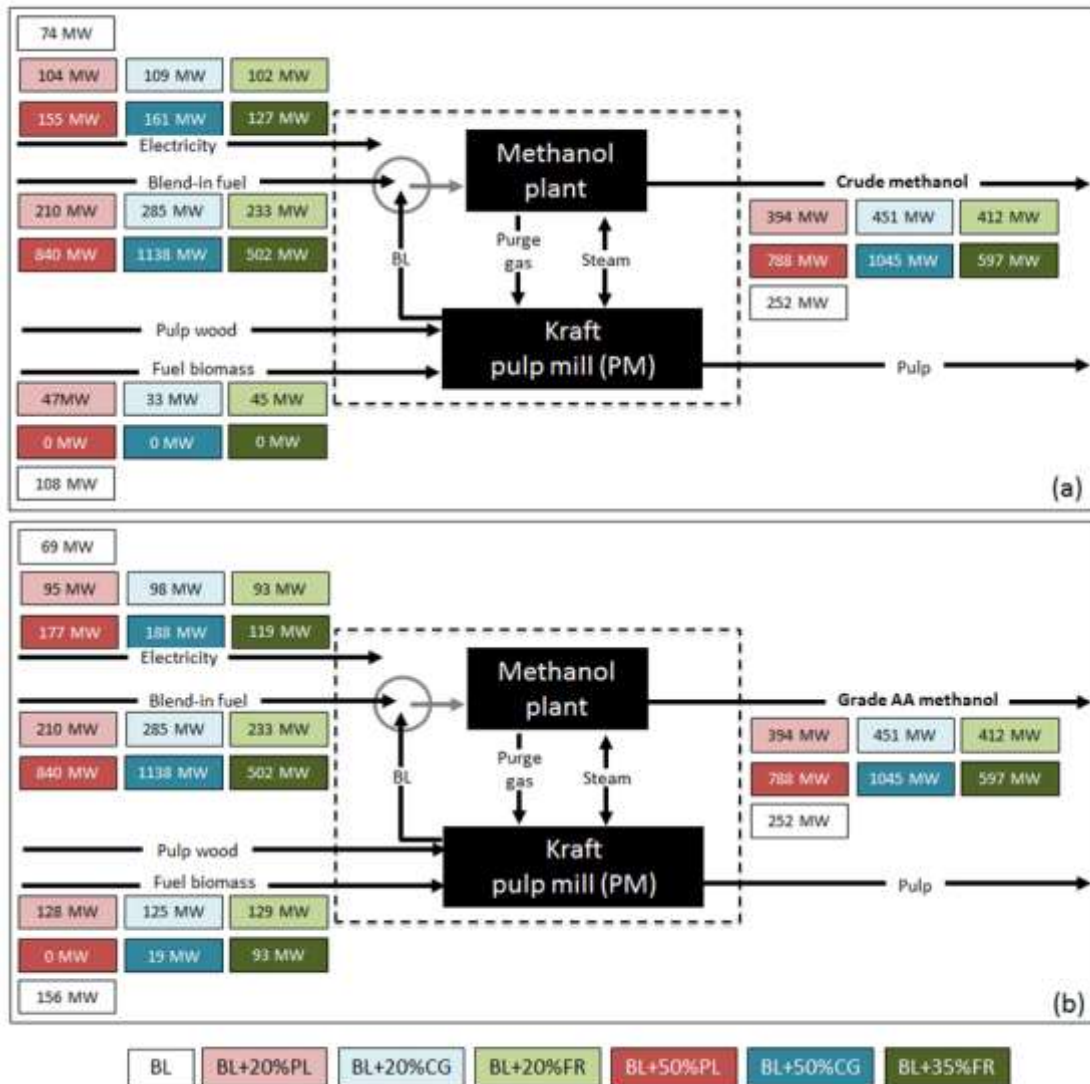


Figure 2. Energy flows for (a) crude methanol and (b) grade AA methanol production. A black liquor supply of 509 MW was used for all the cases. BL: black liquor; PL: pyrolysis liquid, CG: crude glycerol; FR: fermentation residues. Note that blending ratios in the legend (+20 to +50 %) are shown on mass basis.

As shown in the figure, the methanol production capacity when gasifying only the available black liquor of the pulp mill was 252 MW. The capacity increased by up to 80 % for the lower blending cases, and up to 300 % for the higher blending cases. This resulted in that the highest production capacity exceeds 1 GW in the case with addition of 50 % crude glycerol. This result means that by using only a partial stream of the total black liquor flow from a pulp mill, significant economies-of-

scale can still be obtained. This would also allow keeping the recovery boiler for processing the rest of the black liquor. Such a concept would reduce the technical risk compared to total replacement of the recovery boiler with black liquor gasification and lead to the potential dual benefit of increased pulp production from debottlenecking the recovery boiler.

Figure 3 shows the resulting cold gas efficiencies (CGE) and methanol conversion efficiencies (MCE). In addition to this, the figure also shows the corresponding marginal efficiencies, which represents the conversion efficiencies of the blend-in feedstocks into syngas (marginal CGE) and methanol (marginal MCE) using the BL gasification as a baseline (i.e. without black liquor but still subject to the alkali content of the black liquor).

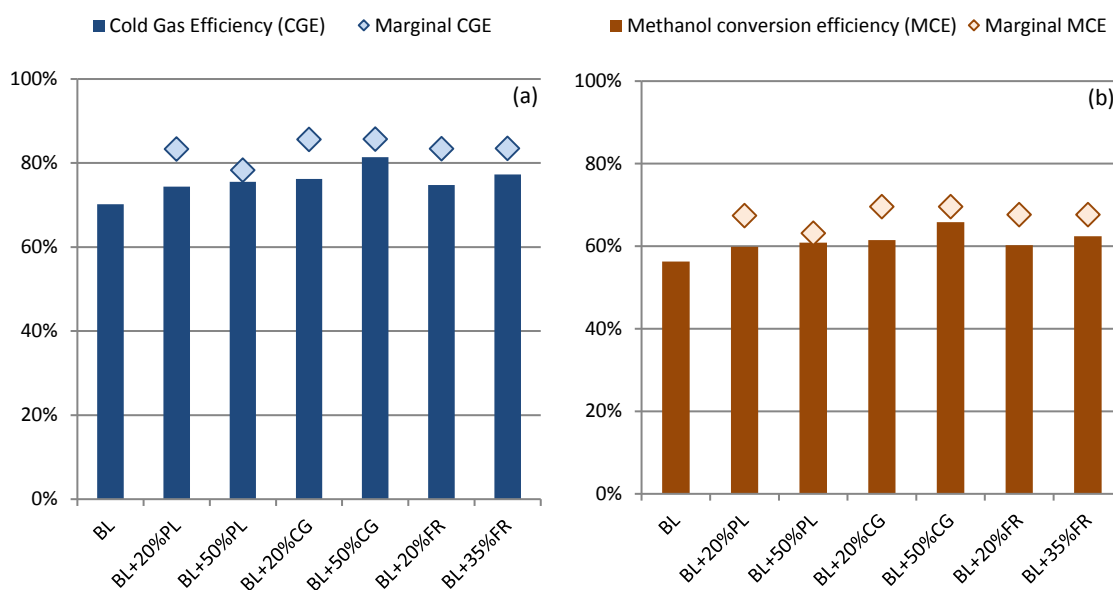


Figure 3. (a) Cold gas efficiency (CGE) for the different cases and marginal CGE for the blend cases; (b) methanol conversion efficiency (MCE) and marginal MCE.

All blend cases showed higher efficiencies than when gasifying pure black liquor. Also the marginal efficiencies illustrate that the actual gasification conversion and methanol conversion efficiencies of the blended secondary feedstocks is aided by the catalytic effect of the alkali in the black liquor. By mixing with black liquor, the marginal efficiencies of the secondary feedstocks are thus higher than the overall CGE and MCE, respectively.

The overall energy efficiencies on marginal basis are presented in Figure 4. for both considered methanol qualities. The results are presented in terms of (i) energy flows in MW and (ii) energy recalculated into electricity equivalents.

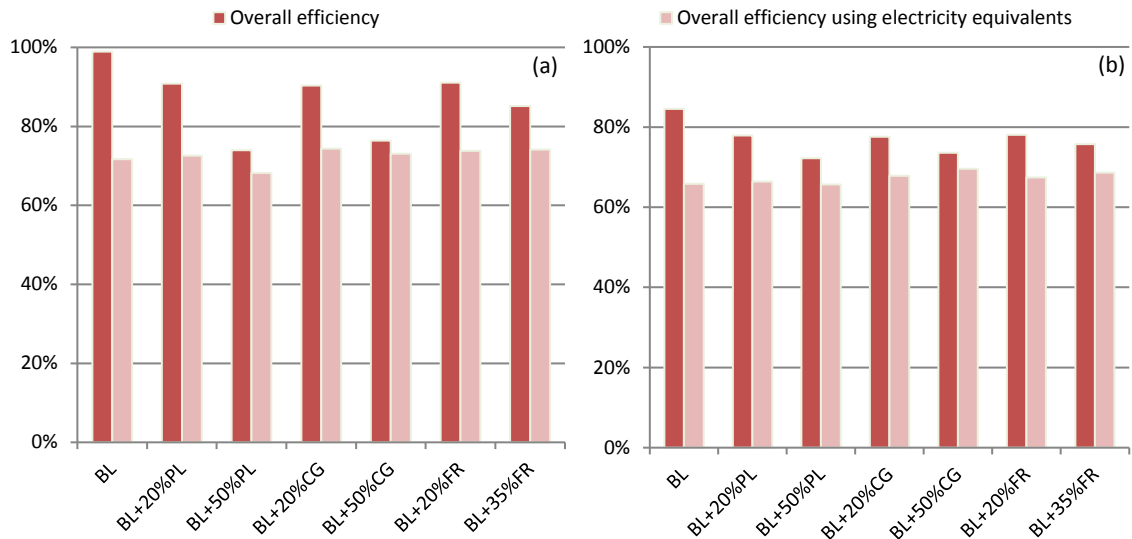


Figure 4. Marginal overall system efficiency using (i) energy flows in MW and (ii) electricity equivalents for (a) crude methanol and (b) grade AA methanol production.

As shown, the two different efficiency measures seem somewhat inconsistent and not really in agreement. They however illustrate different aspects of the efficiency improvements of integrating a methanol plant and of adding blend-in fuels with different blend ratios.

When using the electricity equivalents (lighter bars), the variations can be seen to be relatively small between the different cases, with resulting efficiencies in the range of 68 to 74 %. When instead using energy flows in conventional power units (MW) (darker bars), pure BL gasification resulted in the highest overall efficiency. When crude methanol was produced, the efficiency was as high as 99 % calculated on marginal basis (i.e. compared to the stand-alone pulp mill). The efficiency was reduced with increasing blending ratios, and the lowest overall efficiency was found for the 50-50 blend with pyrolysis liquids (72 %). While this may seem contradictory, it can be explained by the strong efficiency gain caused by the process integration itself. This is further discussed in Carvalho et al. (2017).

In conclusion, all the calculated efficiencies can however be considered as high compared to values for methanol production typically found in the literature (see e.g. Andersson et al 2014, Börjesson et al 2013). To make the different production systems more comparable and also make the primary energy needed for electricity production explicitly, the marginal overall efficiency was also calculated with the electricity converted into biomass i.e. the biomass that would be required to produce the same amount of electricity, assuming a conversion efficiency of 46.2 % (Carvalho et al (2017), Table 4).

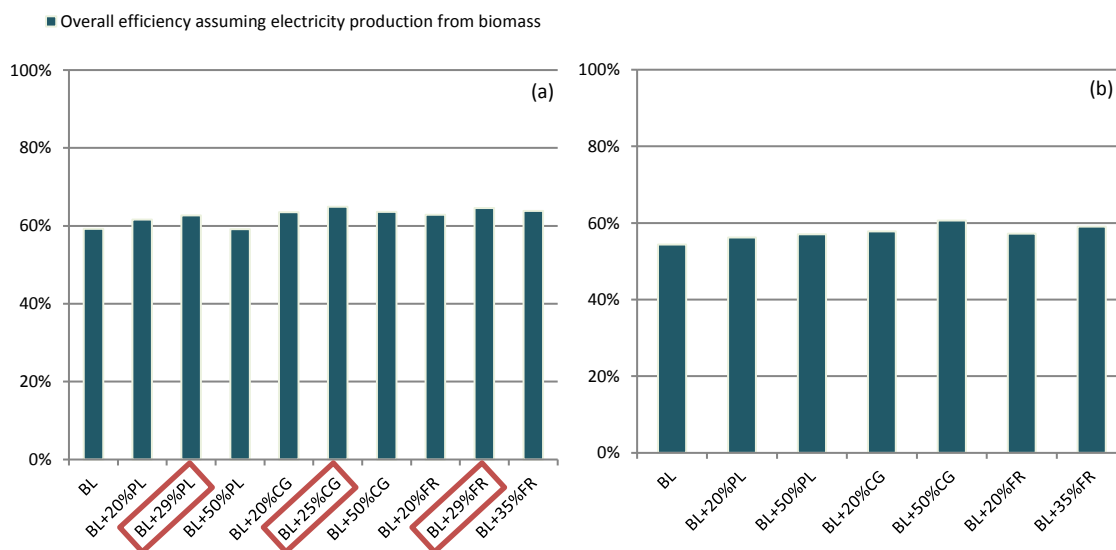


Figure 5. Marginal overall systems efficiency by converting the electricity portions in terms of biomass equivalents for (a) crude methanol and (b) grade AA methanol production. (a) includes the optimum blending ratios, as regards the overall steam balance (see the text for a description).

As illustrated in Figure 5, the marginal overall efficiencies become lower for all cases when compared to the electricity equivalents. This is in particular true for the pure black liquor cases, which now result in lower efficiencies than all of the co-gasification cases. These efficiency values also agree well with the ones calculated using electricity equivalents and increase with increasing blend ratios. Only for the high-blend cases producing crude methanol a different trend can be seen, as the efficiency decreases or remains unchanged, compared to the corresponding low-blend cases. These systems produced a large excess of steam used that was assumed to be used for electricity production in condensing turbines with low efficiency. See also Carvalho et.al (2017) for more detailed explanations.

The results from the process simulations showed a certain over-production of steam, for the crude methanol cases. In order to minimize the steam excess and avoid the use of condensing turbines, the amount of blend-in fuels could thus be reduced. The optimal blending ratios² were calculated to 29 %, 25 % and 29 % for pyrolysis liquids, crude glycerol and fermentation residues respectively, as illustrated in Figure 5.

The required selling prices for the different methanol qualities to reach an Internal Rate of Return (IRR) of 15 % are presented in Figure 6 and Figure 7. The costs shares are divided into: (i) capital (accounting for 15 % IRR), (ii) blend-in fuels including costs for required additives such as NaOH, (iii) fuel biomass, (iv) electricity and (v) operation and maintenance (O&M). No taxes were added to the final methanol price. The details of the economic evaluation are described in Carvalho et al (2017).

² "Optimal" is here defined from a technical point of view, as the blending ratio where the excess steam production is eliminated, without requiring addition of fuel biomass to meet the mill's steam demand.

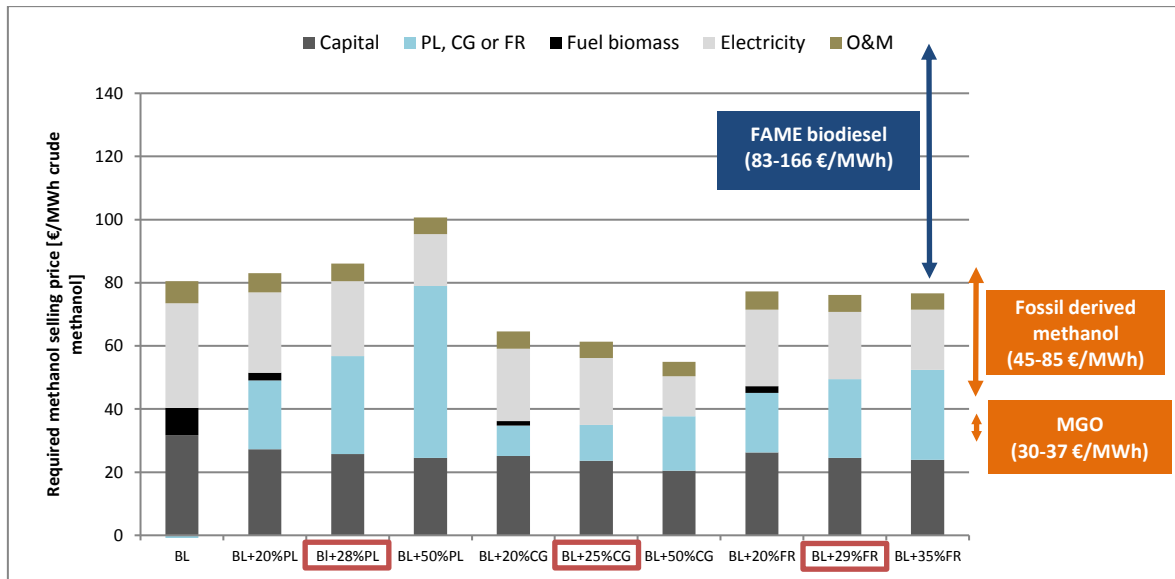


Figure 6. Required crude methanol selling price and comparison with prices of marine gas oil (MGO) (Ship&Bunker, 2016) and fossil derived methanol (Methanex, 2016) as well as production costs of bio-diesel (IRENA, 2016).

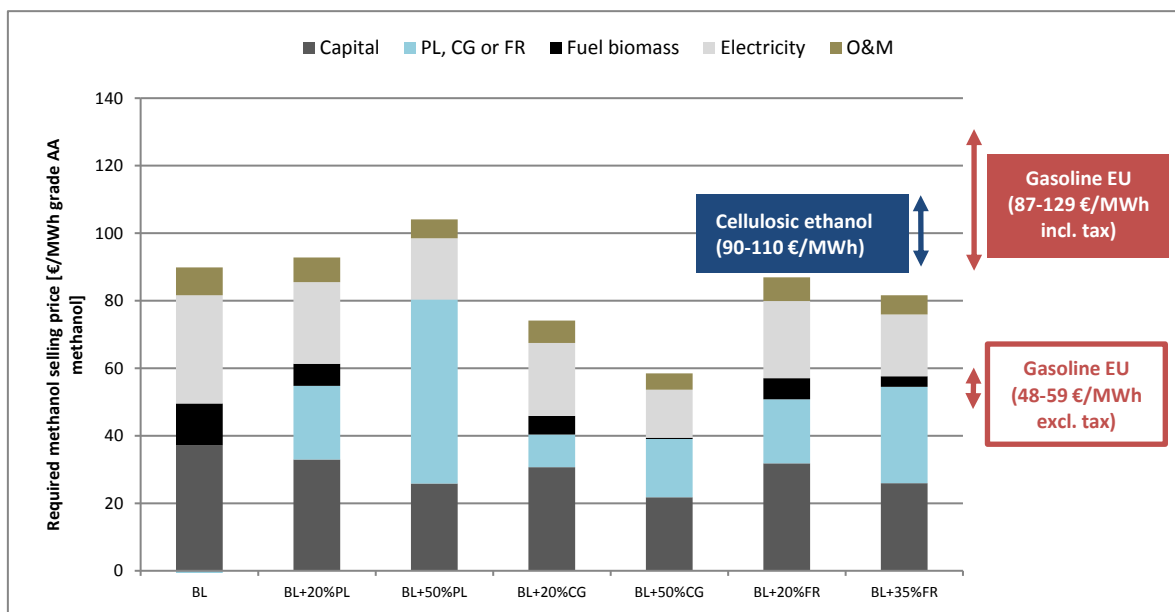


Figure 7. Required grade AA methanol selling price and comparison with production costs of alcohols (Landälv & Waldheim, 2017) as well as gasoline in the EU with and without taxes (European Commission, 2017).

As seen in the figures, the results (values and trends) differ depending on the type of blend-in feedstock. For pyrolysis liquids, the required selling price increases with increasing blending ratios. This is also in agreement with Andersson et al. (2016), who concluded that for large pulp mill capacities, as in the present study, the cost for purchase of the pyrolysis liquids offsets the economy of scale-effects. This is however not the case for the less expensive blend-in feedstocks glycerol and fermentation residues. For these cases, higher blends result in lower required selling prices. The largest decrease in required selling price was observed for the crude glycerol cases, leading to a decrease in the range of 17-34 % compared to the reference case.

An interesting result worth highlighting is that the required selling prices for crude methanol in almost all cases (except the pyrolysis cases) are shown to be competitive against both fossil methanol and other types of biofuels. Marine Gas Oil (MGO) is however available at significantly lower prices (Ship & Bunker, 2016).

The selling price for grade AA methanol was compared to EU gasoline price ranges, with as well as without taxes. As shown in Figure 7, the methanol produced using a high blending share of glycerol can actually even compete with untaxed gasoline under assumed conditions. For all the cases, the required selling prices were lower than, or within the price range of, taxed gasoline.

Further results and discussion as well as a sensitivity analysis are presented in Carvalho et al (2017).

2.1 GREENHOUSE GAS PERFORMANCE

The results of the evaluations of the greenhouse gas performance according to RED and ISO methods of all co-gasification cases and their respective reference case are shown in Figure 8 to Figure 10. For each co-gasification case, a reference case where the black liquor and the blend-in feedstock are converted separately is presented as a comparison. The alternative usage of the blend-in feedstocks were assumed to be direct gasification with subsequent methanol production for logging residues, anaerobic digestion and biogas production for the glycerol, and combustion of the fermentation residues for heat and power production (see Appendix 1). As such, Figures 8 to 10 show the resulting GHG emissions per MWh of fuel or electricity, both for the co-gasification cases and for each reference case ("Ref") assuming alternative conversion routes for the respective resource. The resulting GHG emissions of methanol from co-gasification are compared both to a fossil reference and to the resulting GHG emissions from alternative treatment of BL and blend-in feedstocks.

All cases, including the reference cases, showed greenhouse gas emissions below the RED emission reduction target of 60 % as compared to a fossil fuel reference (i.e. 122 kg CO₂-eq. MWh⁻¹ or 34 g CO₂-eq. MJ⁻¹. (In the updated RED-proposal, the fossil reference is a slightly higher and a 60 % reduction corresponds to 136 kg CO₂-eq. MWh⁻¹ or 38 g CO₂-eq. MJ⁻¹)). Further results and discussions are presented in Appendix 1.

Black liquor blended with pyrolysis liquids: Using the RED method (Figure 8), the specific greenhouse gas emissions were in the range of 36-54 kg CO₂-eq. MWh⁻¹, where the low blends were in the lower region. When applying the ISO method, the emissions more or less doubled to 70-115 kg CO₂-eq. MWh⁻¹. The reference cases turned out to be marginally better than the co-gasification cases due to losses during the production of pyrolysis liquids.

Black liquor blended with crude glycerol: Figure 9 shows that the crude glycerol co-gasification cases resulted in the lowest greenhouse gas emissions, in the range of 19-43 and 34-63 kg CO₂-eq. MWh⁻¹ for RED and ISO calculations, respectively. The higher blend-in shares represent the lower levels. Both the RED and ISO methods resulted in higher emissions for the reference cases, mainly due to leakage of methane from the digestion and biogas upgrading system. Applying the ISO-method, the upstream emissions of glycerol was an important contributing parameter.

Black liquor blended with fermentation residues: The greenhouse gas emissions from the different cases vary depending on the calculation method used, see Figure 10. Using the RED method,

the emissions are approximately 30-40 kg CO₂-eq. MWh⁻¹ for all cases. The reference cases are all marginally lower. The results applying the ISO method shows changes in soil organic carbon (SOC) as another contributing process, resulting in that co-gasification and reference cases become similar in performance (all in the range of 40-50 kg CO₂-eq. MWh⁻¹).

Due to many uncertainties, the results of the GHG calculations should be viewed as first screening results, and therefore be interpreted with caution. Among the main uncertainties are implementations of the studied technologies, availability and assumed alternative use of blend-in feedstocks, and changes in surrounding energy systems (see Appendix 1).

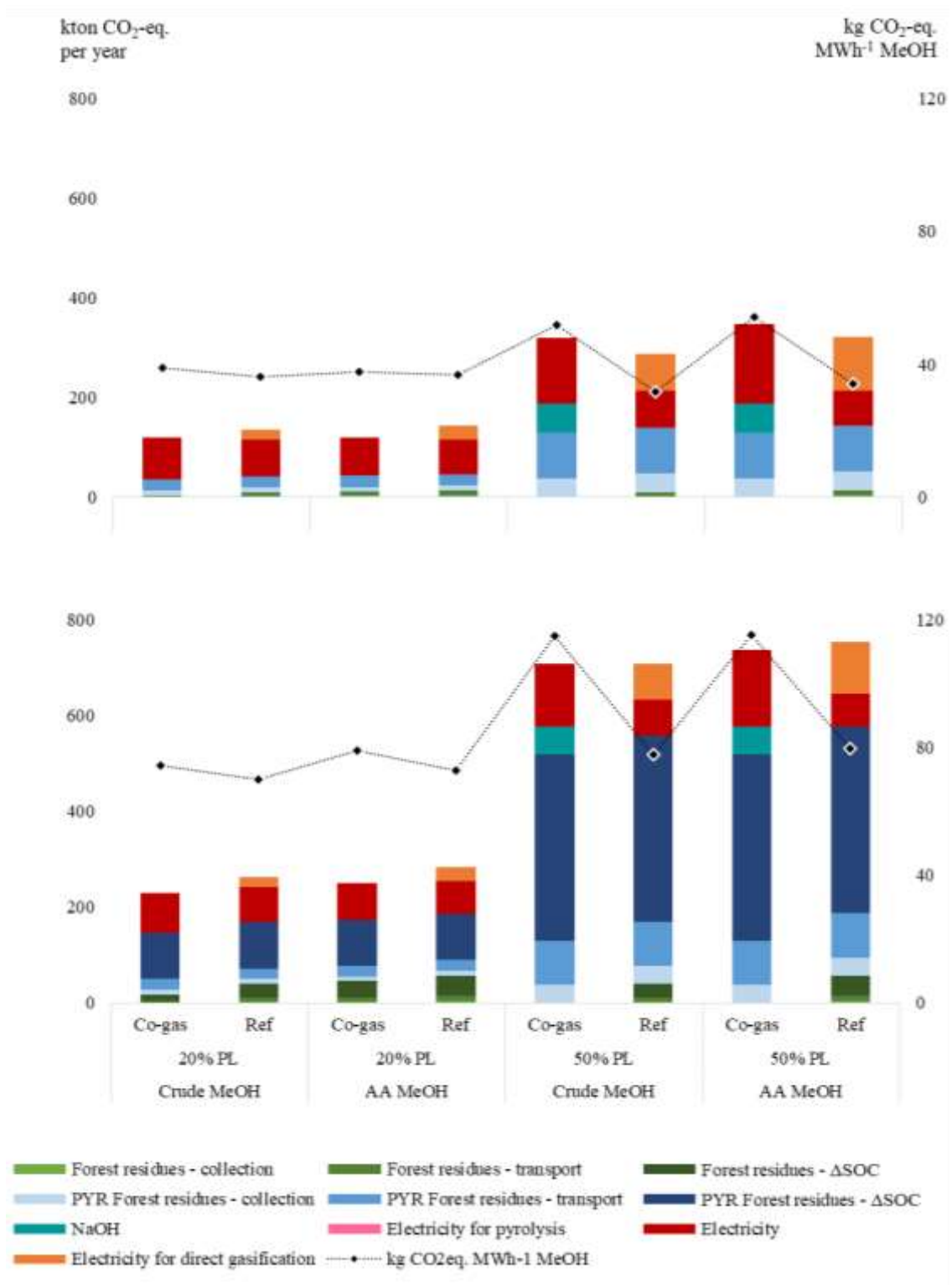


Figure 8. GHG emissions from co-gasification of black liquor and pyrolysis liquid, calculated following the RED method (upper) and the ISO method (lower).

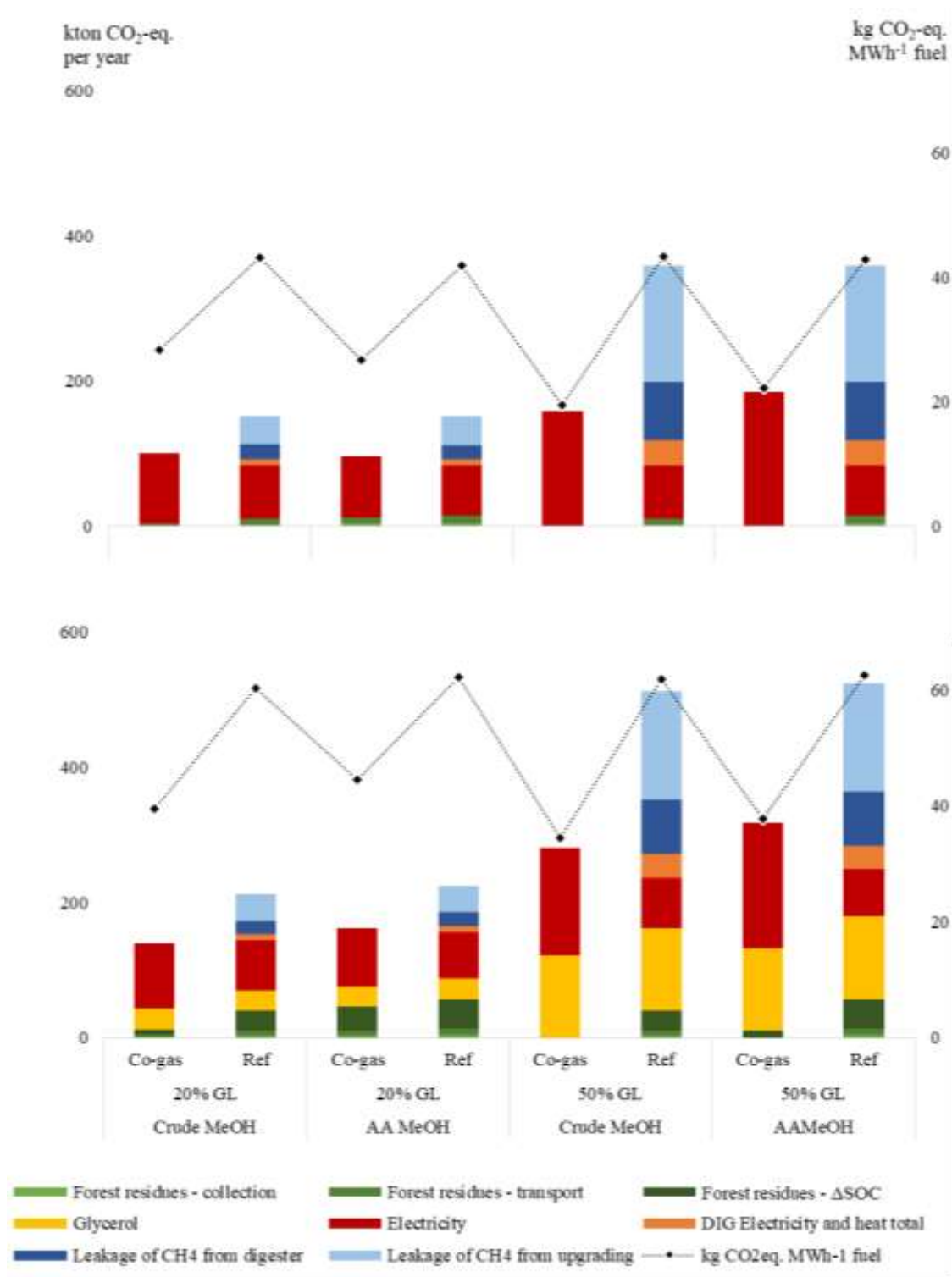


Figure 9. GHG emissions from co-gasification of black liquor and glycerol, calculated following the RED method (upper) and the ISO method (lower).

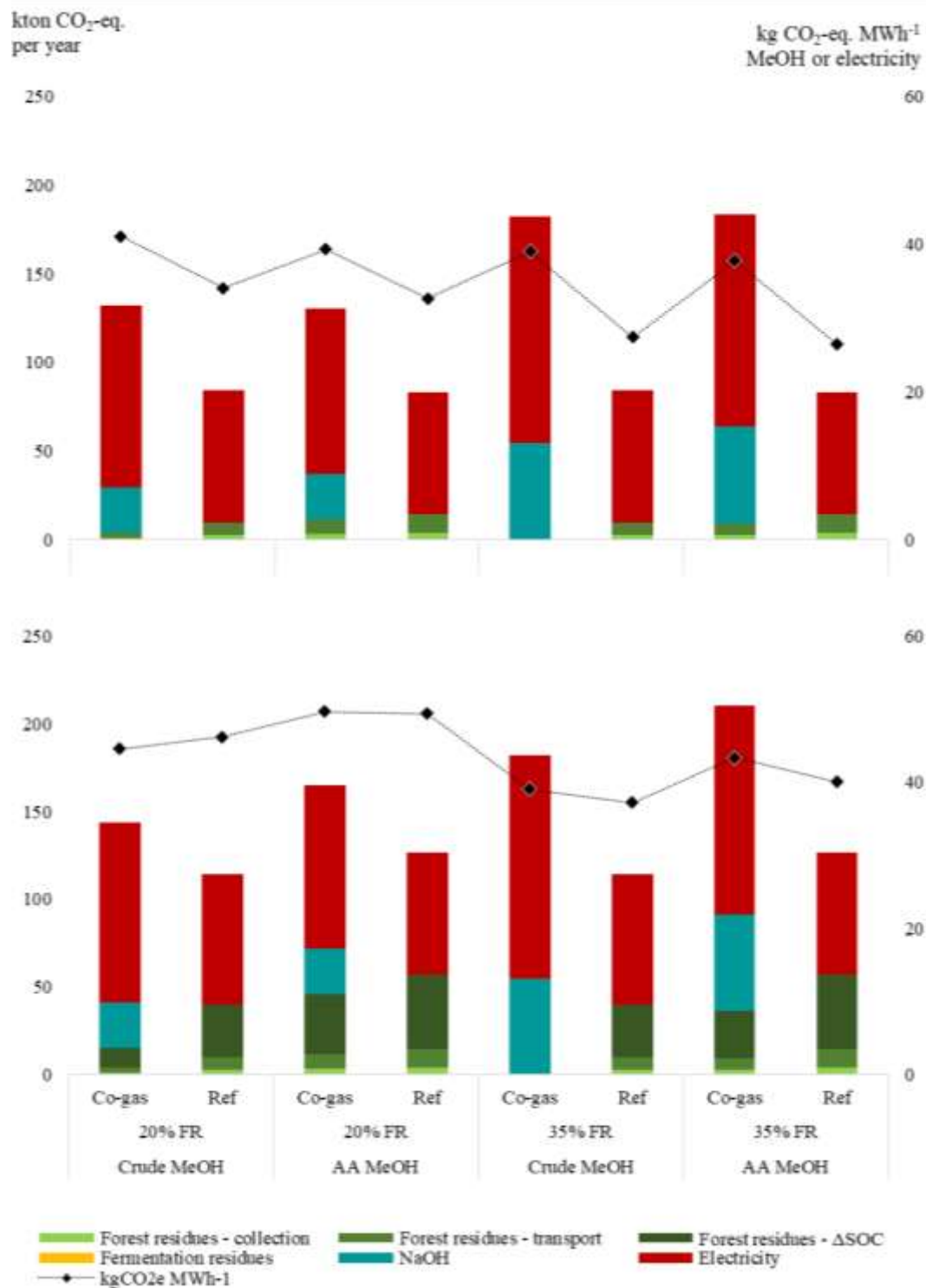


Figure 10. GHG emissions from co-gasification of black liquor and fermentation residues, calculated following the RED method (upper) and the ISO method (lower).

3 ESTIMATION OF PRODUCTION POTENTIAL

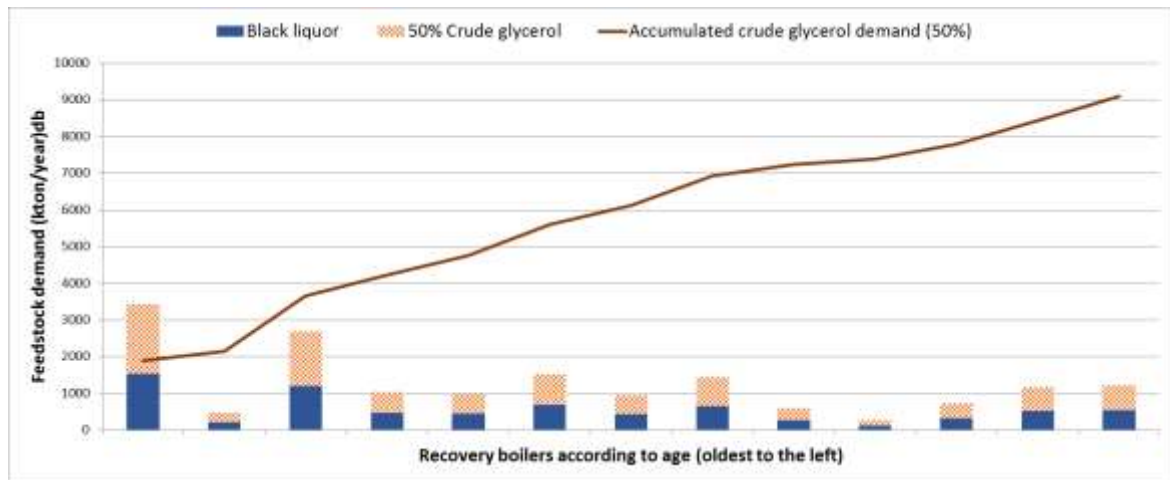


Figure 11 and.

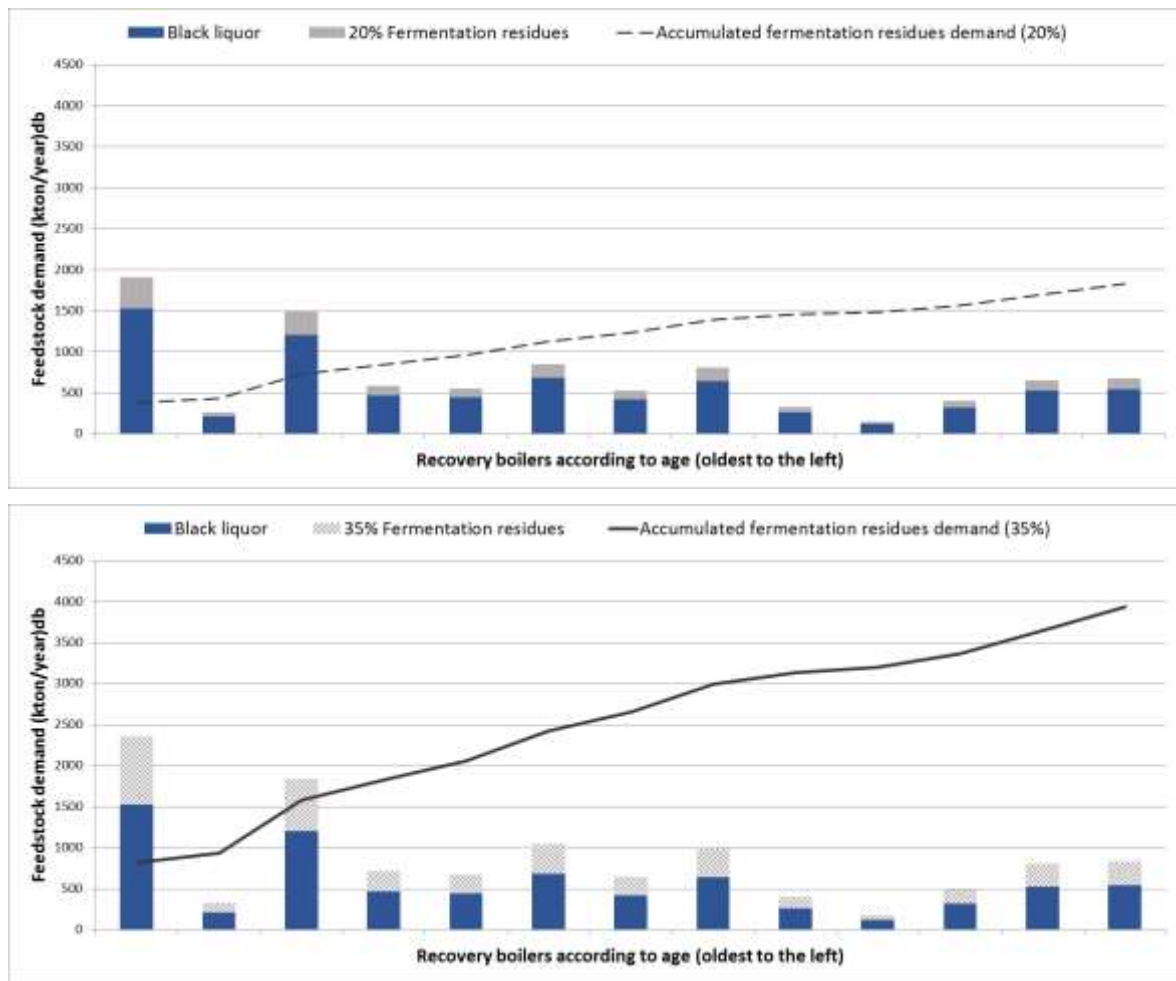


Figure 12 show the accumulated volumes of glycerol and fermentation residues required when replacing the 13 of the currently oldest Swedish recovery boilers (SNRBC & Chemrec, 2015) with the co-gasification concept for each of the two blending ratios.

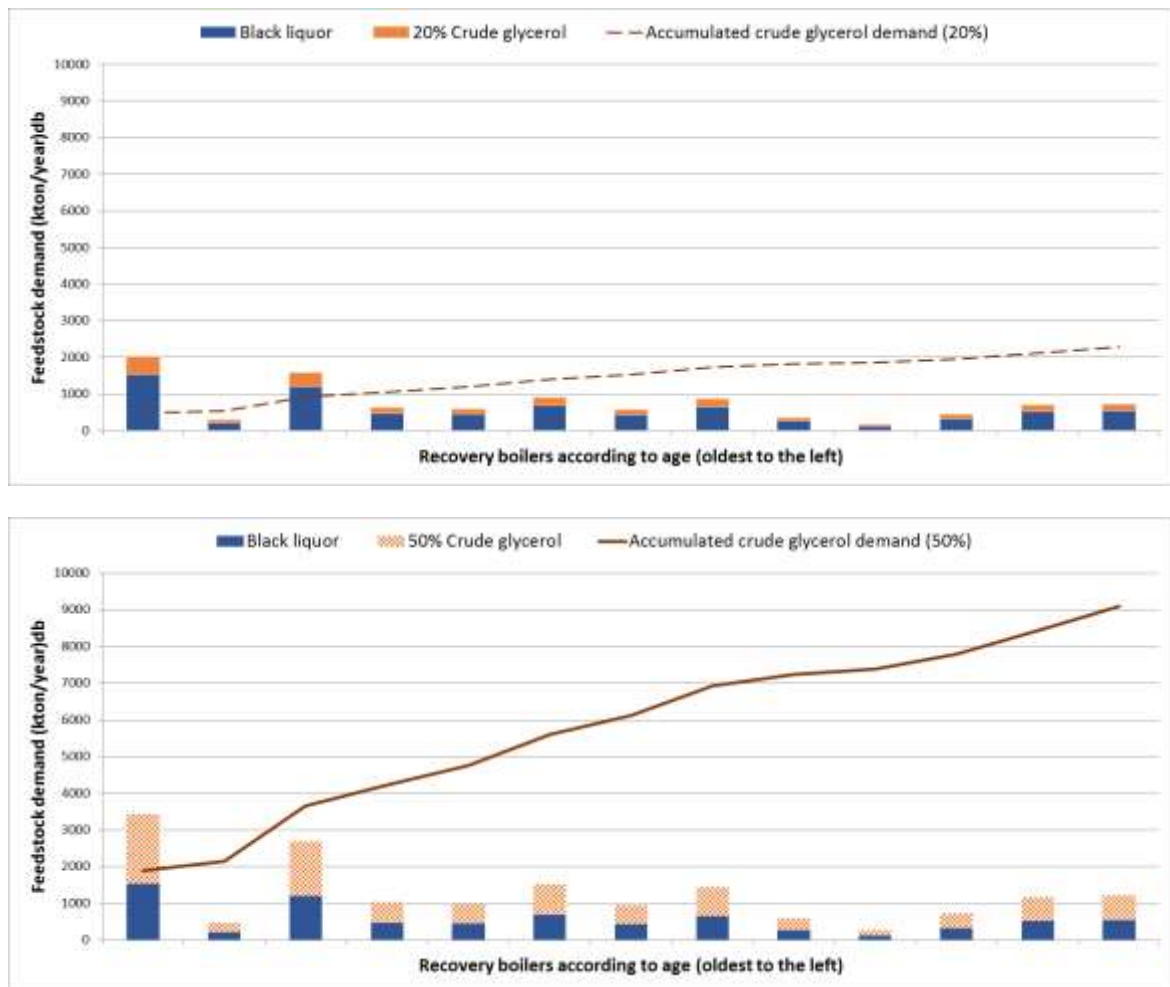


Figure 11. Accumulated glycerol volumes for 20 % and 50 % co-gasification respectively (lines) required to replace aging Swedish recovery boilers with biofuel production based on co-gasification of black liquor and crude glycerol. Bars show feedstock demand for each individual pulp mill that is converted.

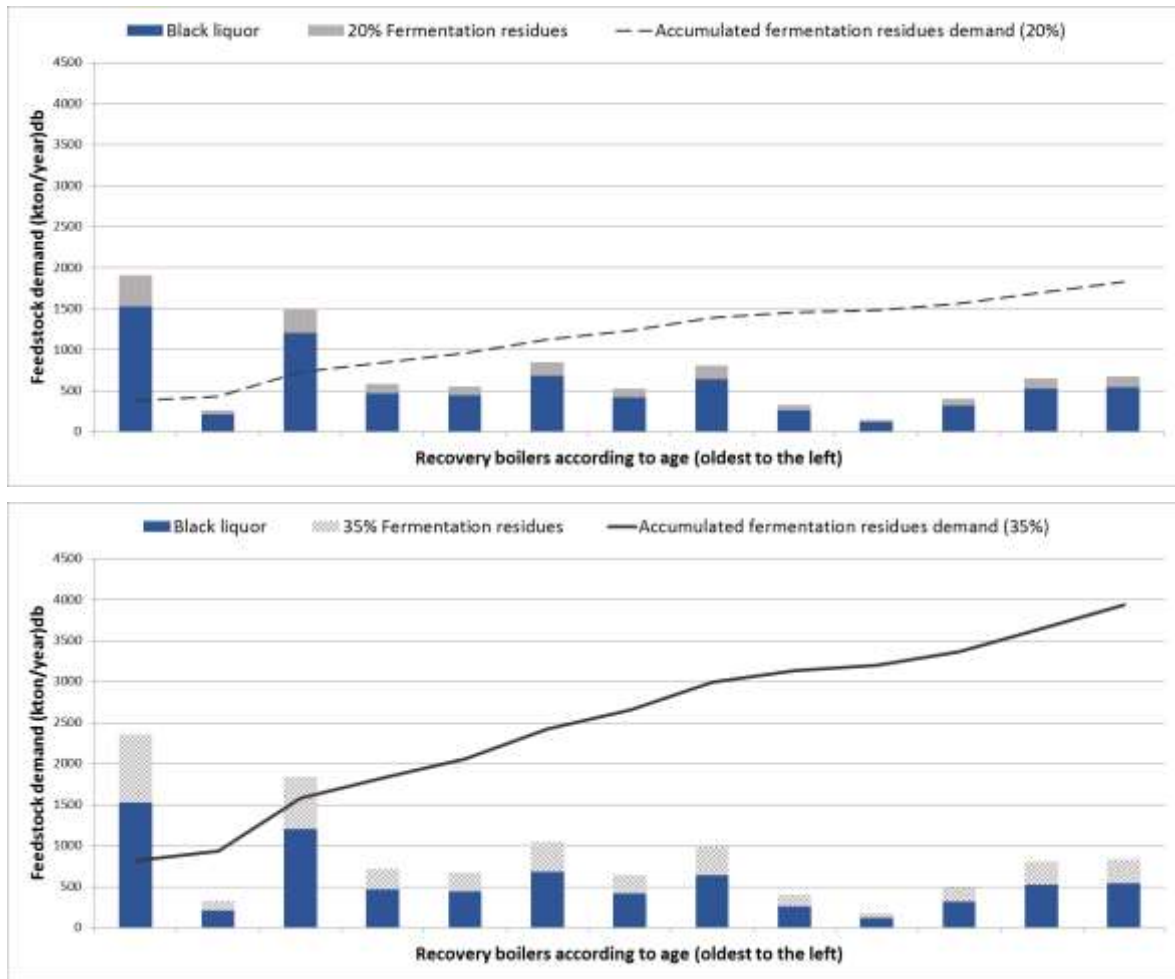


Figure 12. Accumulated volumes of fermentation residues for 20 % and 50 % co-gasification respectively (lines) required to replace aging old Swedish recovery boilers with biofuel production based on co-gasification of black liquor and fermentation residues. Bars show feedstock demand for each individual pulp mill that is converted.

As illustrated in

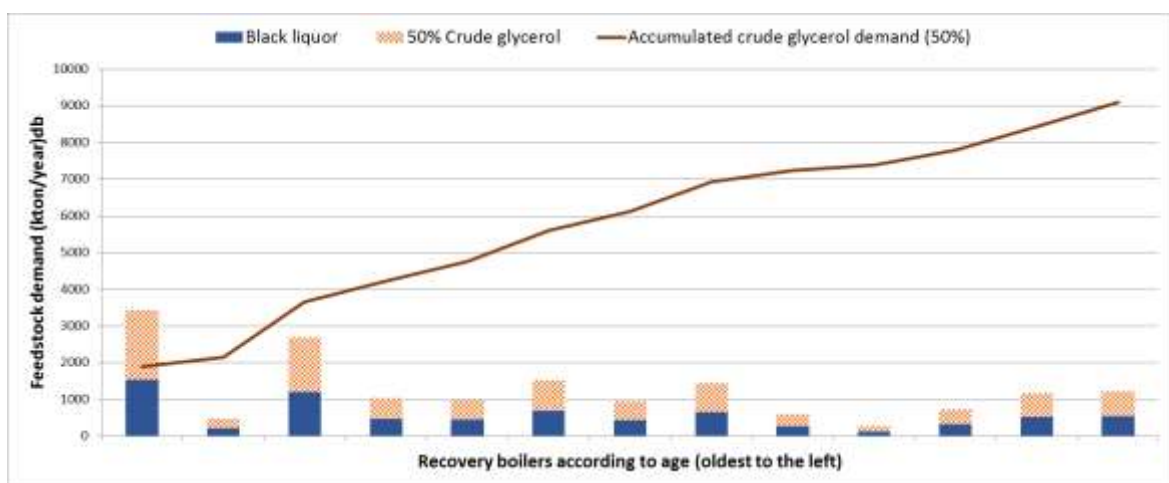


Figure 11, glycerol volumes in the range of 500-1,900 ktons would be required annually if the oldest Swedish recovery boiler was replaced with co-gasification. Converting one of the two smallest pulp mills applying the lower blending ratio would require in the range of 35-60 ktons per year of glycerol.

To put these numbers into a context, the total European glycerol consumption in 2016 (crude and refined) was around 800 kton (OLEOLINE, 2016). This means that the use of glycerol for co-gasification, in particular in large mills and with high blending ratios, would have an exceptionally strong impact on the European glycerol market.

As shown in .

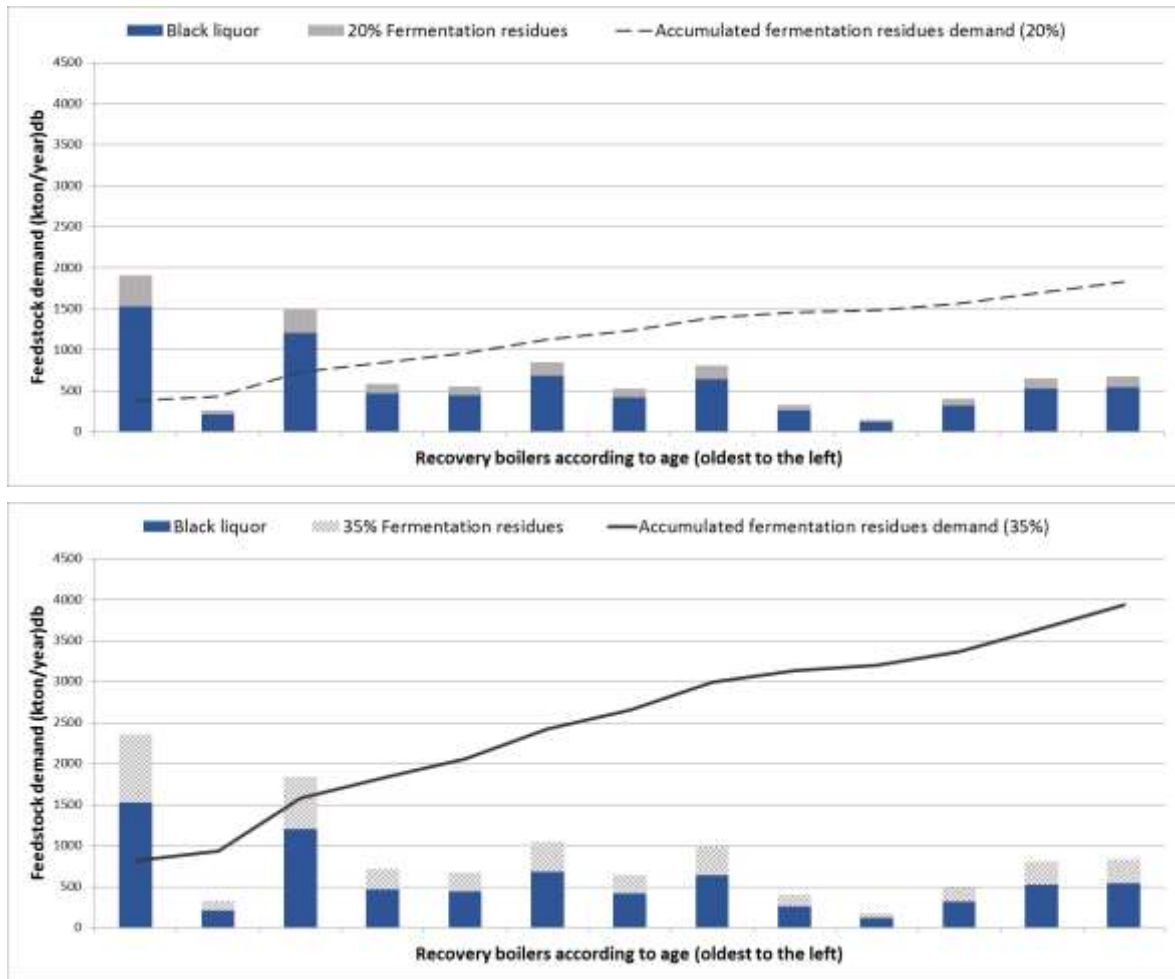


Figure 12, slightly lower volumes of fermentation residues were required compared to the glycerol cases. However, replacing the oldest recovery boiler would require in the range of 380-820 ktons of fermentation residues per year. Assuming a 20 % (w/w) yield of fermentation residues, a lignocellulosic ethanol production in the range of 2,000-4,000 ktons per year would be needed to cover the residue demand. As a comparison, the total global ethanol production (all feedstocks) in 2017 was estimated to reach approximately 92,000 ktons.

4 CONCLUSIONS

This study evaluated the techno-economic and greenhouse gas performances of producing two different methanol qualities in a pulp mill via integrated co-gasification of blends of black liquor and three other biomass feedstocks: pyrolysis liquids, crude glycerol and fermentation residues. The following conclusions were drawn:

- All investigated cases showed good economic performance. The required selling price to reach an IRR of 15% for crude and grade AA methanol produced via pure black liquor gasification, i.e. the reference case, were around 80 € per MWh and 90 € per MWh, respectively.
- Blending the black liquor with pyrolysis liquids showed higher required selling prices than the reference case (82-100 € per MWh for crude and 93-104 € per MWh for grade AA) (with the higher levels for higher blending ratios), while blending with a less expensive feedstock (crude glycerol) the required selling prices became significantly lower, 55-65 € per MWh for crude and 58-74 € per MWh for grade AA methanol (the lower levels with higher blending ratios).
- The crude glycerol co-gasification cases showed such good economic performances that the crude methanol could be competitive to current prices of fossil based methanol and the grade AA methanol to untaxed gasoline.
- All cases showed good energy performance with high energy efficiencies. Blending feedstocks with black liquor showed positive influences on the cold gas efficiency and methanol production efficiency due to the catalytic effect of the black liquor. All co-gasification cases also showed higher efficiencies than pure black liquor gasification.
- Calculation of different types of efficiencies revealed that the largest gain in overall efficiency was caused by the integration itself and not by co-gasification.
- By calculating GHG emissions following the RED method, the methanol produced in all cases result in emissions savings of 82-94 % compared to the fossil fuel reference. Following the ISO standards, the results for the same fuels instead ranged from 62-88 % GHG emissions savings. This difference was due to wider system boundaries in the case of ISO calculations which included effects on soil organic carbon (SOC) from logging residues removal, and a share of the emissions from RME production in the case of glycerol.
- The production capacities became very large, in particular for the 50-50 blends. In the case of a 50-50 glycerol blend, the methanol production capacity exceeded 1000 MW, which would put significant stress on both the capital requirement, and the requirement for blending feedstock. One main limiting factor is therefore the availability of the required volumes of blend-in components.
- An important insight was that co-gasification could be a pathway to make black liquor gasification with biofuel production economically feasible also when using only a partial stream of the total black liquor flow from a pulp mill, while also keeping the recovery boiler for processing the rest of the black liquor. This would lead to the potential dual benefit of increased pulp production from debottlenecking the recovery boiler, and significantly

reduced technical risk compared to total replacement of the recovery boiler with black liquor gasification. While this concept has been outside the scope of this project to evaluate, it is recommended for further studies

- The general conclusions were that blending a secondary feedstock with black liquor for co-gasification was an energy efficient and cost effective pathway to convert the feedstock to methanol. The produced methanol, in all considered cases, fulfilled the currently required 60 % GHG emissions reduction target as compared to a fossil fuel reference according to RED. However, from a GHG-performance perspective, co-gasification was not necessarily found to provide a better alternative than other usage options of the blend-in feedstocks.

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APPENDIX 1. ASSESSMENT OF THE GREENHOUSE PERFORMANCE

METHODOLOGY

The greenhouse gas (GHG) emissions of methanol were assessed using a life cycle perspective and methods which includes raw material production and processing in a so called well-to-tank analysis. The well-to-tank GHG emissions were quantified following the life-cycle based calculation approach as required by the EU Renewable Energy Directive (RED) rules for calculating GHG emissions from liquid biofuels (2009/28/EC Annex V). The resulting life-cycle GHG emissions were combined with global warming factors as defined by the IPCC in order to estimate the impact on climate, and more specifically the global warming potential (GWP). The global warming factors used were 34 g CO₂-eq. per g CH₄ and 298 g CO₂-eq. per g N₂O (Myhre et al. 2013).

In addition to the EU RED method for calculating GHG emissions, parallel assessment is made according to the ISO standards for life cycle assessment, 14040 (ISO 2006a) and 14044 (ISO 2006b). A reason for using a second parallel calculation approach for life-cycle GHG emissions is the importance of method choices to life-cycle assessment results. Where RED calculations could be considered more relevant to current European policy requirements, ISO standards leave more room for interpretation and choice. In this study, the ISO calculations thus represent a type of sensitivity analysis to EU RED results, as they highlight some potential implications of changed calculation rules.

Comparing the two calculation approaches of RED and ISO, one important difference is that the latter includes changes to soil organic carbon stock. According to the RED calculation rules, potential changes in soil organic carbon stock due to removal of logging residues are not included as long as the overall use of the land as forest land does not change. This is because only emissions “*from carbon stock changes caused by land-use change*” are to be included in calculations of GHG emissions from biofuels (RED Annex V), where land use change is defined by the Swedish Hållbarhetsförrordning (SFS 2011:1088, §5) as a shift between two of six land use categories; forest land, grassland, agricultural land, wetland, built-up area and other land. Following ISO standards, however, no such rules are specified. Therefore the RED calculations in this study do not include soil organic carbon changes from removal of logging residues, while the ISO calculations do.

Another potentially important distinction in the RED calculation rules concerns by-products as resources, where the EU RED states that wastes and residues from processing should be considered to have no greenhouse gas emissions prior to their collection. The directive specifically mentions crude glycerol (glycerine) as one such processing residue:

“Wastes, agricultural crop residues, including straw, bagasse, husks, cobs and nut shells, and residues from processing, including crude glycerine (glycerine that is not refined), shall be considered to have zero life-cycle greenhouse gas emissions up to the process of collection of those materials.” (RED Annex V, paragraph 18)

In their guidelines for the EU RED calculation method, the Swedish Energy Agency further mentions black liquor and crude glycerol as by-products to be considered to have zero life-cycle GHG

emissions (Swedish Energy Agency 2012). Also logging residues are stated as zero-emission residues in the guidelines, but with requirements to fulfil the EU RED land criteria, e.g. regarding the type and status of forest land where residues are collected from (RED Article 17).

SYSTEM DESCRIPTION AND LIFE-CYCLE INVENTORY

Each of the co-gasification cases were compared to a reference scenario with alternative handling of the biomass. The underlying modelling results for each co-gasification case are available in Carvalho et al. (2017) and Appendix 2 (Tables C1 to C5).

For calculations of methanol GHG emissions, the pulp produced was not considered in the assessment. Also, the use of pulpwood was entirely allocated to pulp, and thus not included in the calculation of GHG emissions.

In all cases, electricity was considered as Nordic electricity mix with GHG emissions of 125.5 g CO₂-eq. kWh⁻¹, or 34.9 g CO₂-eq. MJ⁻¹ (Martinsson et al. 2012).

In all reference cases, the alternative use of black liquor was direct gasification for methanol production (raw and grade AA), as illustrated in Figure B 1. The simulation results in terms of mass and energy balances for black liquor gasification are available in Appendix 2. (Table C4)

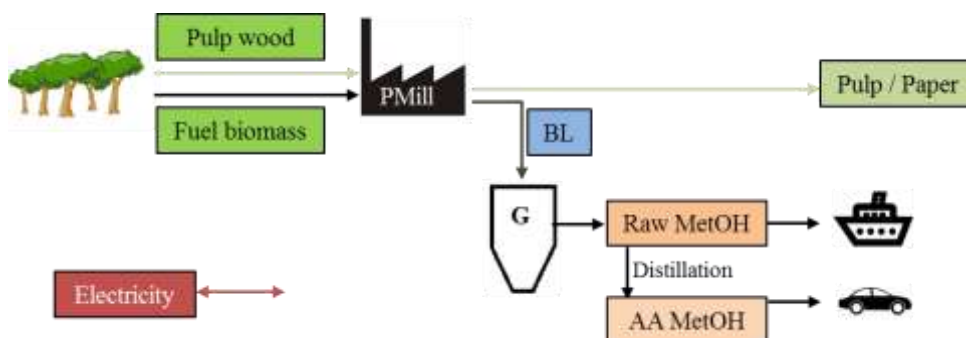


Figure B 1. The considered alternative use of black liquor is direct gasification, which is applied in all studied reference cases.

For logging residues, GHG emissions were considered from collection and transport in both RED and ISO calculations, and from changes in soil organic carbon stock only in ISO calculations. The GHG emissions data used for logging residues are 17.3 g CO₂-eq. kg⁻¹ DM for collection and 42.2 g CO₂-eq. kg⁻¹ DM for transport (Lindholm et al. 2010). Emissions from changes in soil organic carbon were assumed to be 179 g CO₂-eq. kg⁻¹ DM, based on a scenario with two rotations (240 years) in northern Sweden (Lindholm et al. 2011). The same data for logging residues were used also for the fuel biomass input to the integrated pulp mill and biofuel plant, in all studied cases.

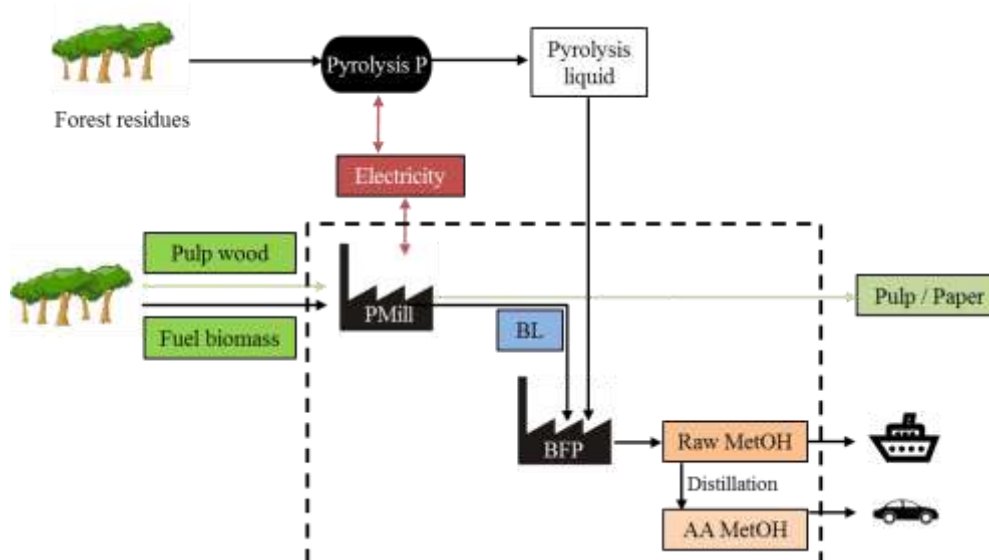
For co-gasification with black liquor, logging residues were converted via pyrolysis to pyrolysis liquid (Figure 3 (a)). The pyrolysis process was assumed to take place in a stand-alone plant where 1 MJ of forest residues is assumed to require input of 32 J of electricity, and yield 0.6 MJ of pyrolysis liquid (at 29 % moisture content), which is based on modelling work by Zetterholm et al. 2017. The assumption of a stand-alone plant was in contrast to an integrated pyrolysis and CHP (combined heat and power) plant, where a higher total efficiency could be achieved (McKeough et al. 2005). Therefore a stand-alone plant could be considered a worst-case scenario for the design of the

pyrolysis system, but here the less complex setup with a stand-alone pyrolysis plant was preferred as a base case.

In the cases of high blend (50 % mixture) of pyrolysis liquid to black liquor, sodium hydroxide (NaOH) was added to the blend with 12 kg NaOH per 100 kg of pyrolysis liquid. As a result, pyrolysis liquid makes up 46 % of the final mix, NaOH makes up 5.4 %, and black liquor 49 % (dry mass basis). For the GHG emissions calculations, 0.48 kg CO₂-eq. kg⁻¹ NaOH was assumed (BioGrace 2012, updated GWP100 for methane).

For the reference cases, the alternative use of logging residues was considered to be direct gasification (Figure B2a). The mass and energy balances were based on process simulations presented in Appendix 1. Important modeling constraints were to produce a sufficient amount of process steam to meet the heat demands of the methanol plant as well as for the pre-treatment of the logging residues. Surplus heat was used to produce electricity via a condensing turbine. For each MJ input of logging residues, the downstream biofuel production via gasification yielded approximately 0.66 MJ of methanol.

a) Co-gasification of BL and pyrolysis liquid from forest residues.



b) Alternative use of forest residues: direct gasification (Gas).

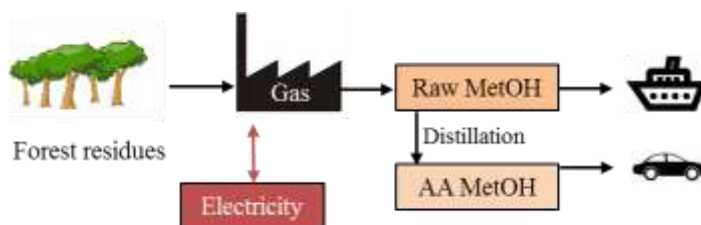


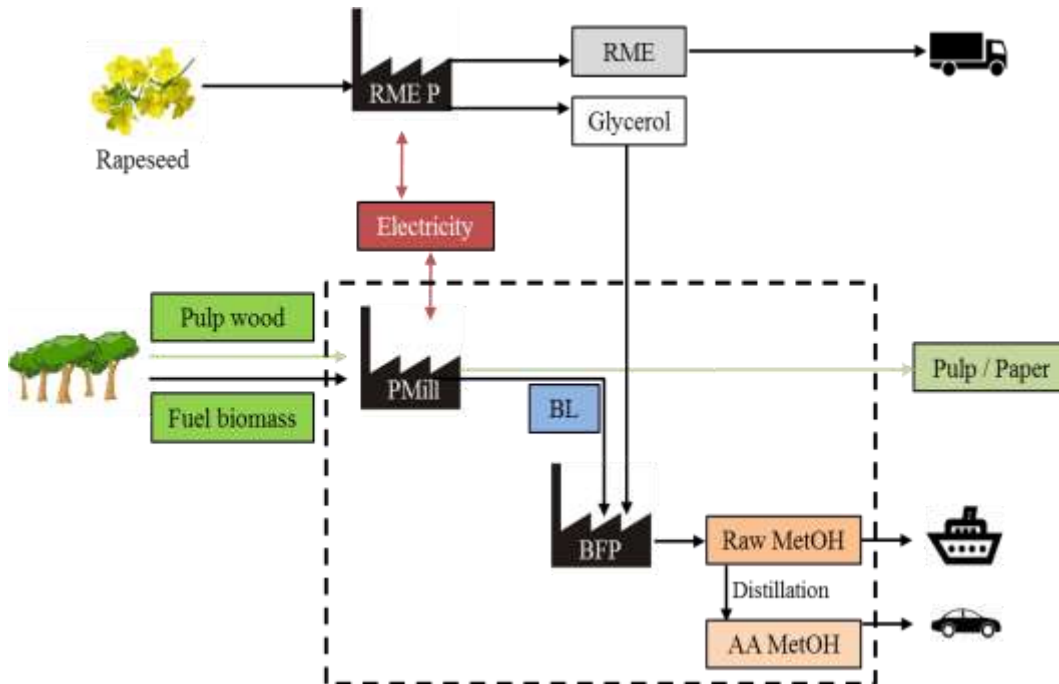
Figure B 2. Schematic overview of the studied system for co-gasification of black liquor (BL) with pyrolysis liquid made from forest residues (a). In the reference case, the alternative use forest residues was direct gasification (b).

The alternative use of glycerol considered was assumed to be anaerobic digestion (Figure B3), and the considered digestion process was based on assumptions of co-digestion with manure, and on general data for anaerobic digestion in a large size plant followed by upgrading of biogas to vehicle

fuel. The assumed digestion of glycerol was thus based on a literature study, where the co-digestion with manure is important in order to assume that all necessary nutrients are available, and in order to avoid potential limitations to glycerol as a sole substrate.

The composition of crude glycerol was assumed to be (on a mass basis) 63 % glycerol, 0.4 % water, 31.6 % MONG (matter organic non glycerol) whereof 25 % FAME, and 5 % ash.

a) Co-gasification of BL and crude glycerol from RME plant (RME P).



b) Alternative use of crude glycerol: anaerobic digestion (AD).

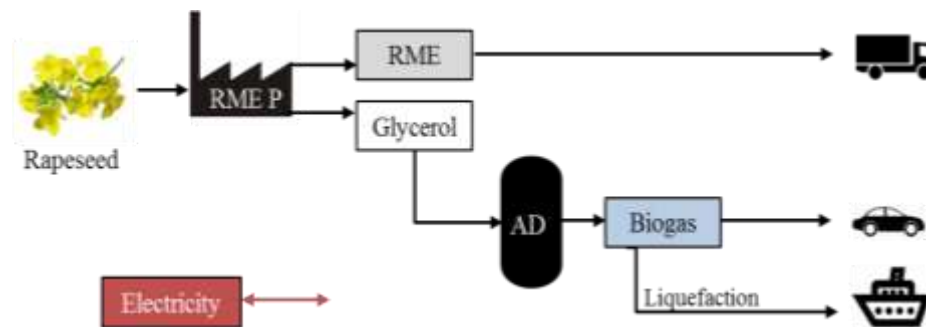


Figure B 3. Schematic overview of the studied system for co-gasification of black liquor (BL) with crude glycerol from biodiesel (RME) production (a). In the reference case, the alternative use of crude glycerol is anaerobic digestion (b).

ANAEROBIC DIGESTION

The chemical maximum methane yield from anaerobic digestion of crude glycerol was calculated theoretically using Buswell's formula, followed by assumptions for calculating a methane yield from the digestion process. For Buswell's formula, we assumed that all fatty acids were oleic acid (18°C), resulting in 0.41 kg CH₄ per 1 kg crude glycerol digested. From the resulting chemical yield, we assumed that $[1/\text{HRT}] \%$, where HRT is the hydraulic retention time, of the substrate is short-circuited in the CSTR and thus not digested. The HRT was assumed to be 25 days (based on

Andriamanohiarisoamanana et al. 2016, Rodríguez-Abalde et al. 2017), meaning $1/\text{HRT}=4\%$ of methane yield lost. Theoretically, 100 % of the remaining yield could be achieved if 100 % of the organic material is digested, but it is not practically achievable. In this study, we assumed that 90 % of the organic material was digested³. Out of this material, another 5 % was assumed to be lost in the form of microorganism biomass, which resulted in a methane yield of 0.34 kg CH₄ per 1 kg crude glycerol. The final production of biogas and methane was estimated to 0.335 kg CH₄ per 1 kg crude glycerol, after subtracting methane slip from the digestion plant (0.5 % of produced methane), and methane slip from upgrading (1 % of methane to be upgraded) (Tufvesson et al. 2013).

The calculation of methane yield from anaerobic digestion of glycerol was based on the assumption that glycerol was co-digested with manure, in order to avoid potential inhibitory effects from the crude glycerol. Concerning crude glycerol as a substrate for anaerobic digestion, the three main potentially inhibiting factors were considered to be i) intermediate products, ii) long fatty acids, and iii) inorganic salts (Viana et al. 2012). For i) the intermediate products, we assumed a low blend and a continuous addition of glycerol, in order to avoid intermediary products accumulating to a critical level. Thus, no inhibitory effect from intermediate products was considered. Regarding ii) long chain fatty acids, these come from the mono-, di- and triglycerides as well as FAME (RME) in the crude glycerol. In the reactor, the fatty acids separate from these molecules and can inhibit digestion. From this point of view, the ~25 % of FAME in the considered crude glycerol was not beneficial, as it makes up most of the long fatty acid content. According to the literature, low concentrations may cause inhibitory effects, and again, a low blend and continuous adding of glycerol was assumed to prevent the concentration from increasing. No inhibitory effect from long chain fatty acids was therefore considered, not ignoring the uncertainties of such an assumption. The third inhibitory factor considered is iii) inorganic salts. The considered crude glycerol appears to have relatively low contents of inorganic salts compared to values reported by literature as problematic (Viana et al. 2012), and also so here a dilution and continuous adding of glycerol was assumed to keep inorganic salts at acceptable levels. Thus, no inhibitory effect from inorganic salts was considered. Conclusively, no inhibitory effects were considered since co-digestion with manure was assumed, but for the calculations of GHG emissions, only the part of digestion representing crude glycerol was included.

³ This assumption is important to decide the final amount of CH₄ fuel produced, but not important to the final GHG emissions from 1 MJ CH₄ fuel produced. This is because the methane slip from the plant and from the upgrading processes is calculated as a fraction of total methane produced, and therefore an increased methane yield also results in increased methane slip. The two factors cancel each other out so that GHG emissions per MJ fuel produced are approximately the same irrespective of the fraction of organic material which is digested.

LCI DATA

The methane slip from the digestion plant and the upgrading process (water scrubber) was included in the calculation of GHG emissions. Furthermore, electricity and heat inputs were included: 7 kWh electricity and 25 kWh heat per ton of substrate (glycerol) and for the digestion plant, and 0.25 kWh electricity per Nm³ biogas for the upgrading process (Tufvesson et al. 2013).

For the sensitivity analysis of including upstream GHG emissions for crude glycerol, corresponding to a share of GHG emissions from RME production, a mean value was derived based on Börjesson and Tufvesson (2011). From the whole biofuel production system producing RME, the percentage of GHG emissions allocated to RME ranged from 61 % (energetic base) to 72 % (economic base), and 3 % was allocated to glycerol (energetic and economic bases). Here an allocation of 65 % to RME was used as an average, with 3 % allocated to glycerol. Assuming total GHG emissions of RME to be 50 g CO₂-eq. MJ⁻¹, which can be considered a high emissions scenario based on unfertilized grassland as reference land use, a 3 % allocation to glycerol corresponds to 88 g CO₂-eq. kg⁻¹ glycerol (using 38 MJ kg⁻¹ for RME). If wheat cultivation were instead to be considered as the alternative land use, GHG emissions from RME would be cut by more than half to approximately 20 g CO₂-eq. MJ⁻¹ RME, and the upstream impacts of glycerol would consequently follow. On the other hand, if the price for glycerol was to change, the allocation to glycerol using an economic basis would also change. For the purpose of this sensitivity analysis, only the value of 88 g CO₂-eq. kg⁻¹ glycerol is used.

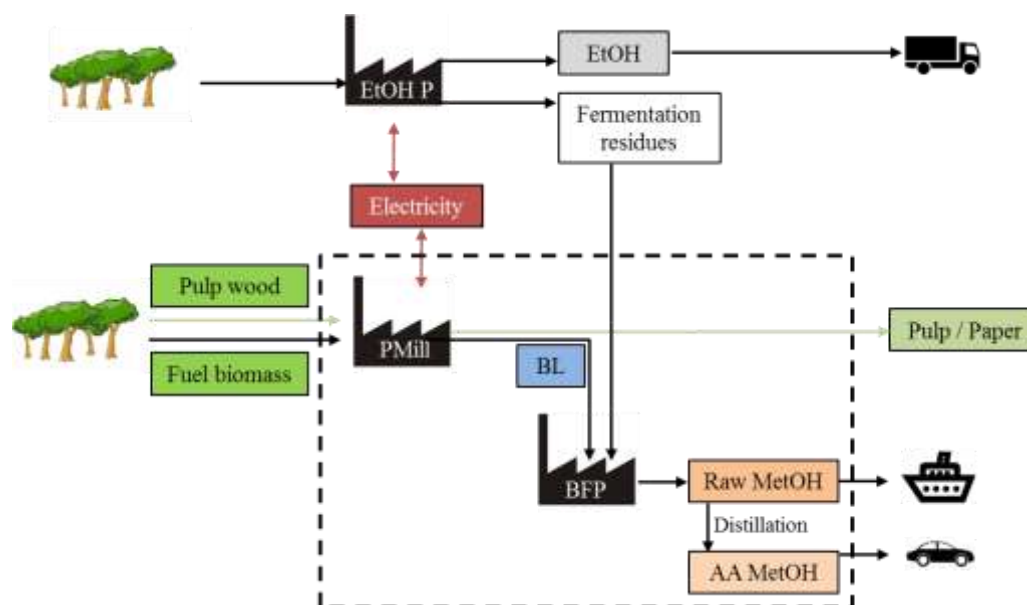
FERMENTATION RESIDUES

In the case of fermentation residues, the alternative use considered was combustion with power generation (Figure B4). Thus the comparison of the final products, methanol in the co-gasification case as compared to methanol and electricity in the reference cases for black liquor and fermentation residues, respectively, does not fully follow the same logic as the other co-gasification cases. Nonetheless, since data for other conversion processes were not available in the current project, the combustion scenario allowed for a comparative reference case for fermentation residues.

For the co-gasification of fermentation residues and black liquor (Figure B4a), sodium hydroxide (NaOH) was added to the blend with 0.2 kg NaOH per kg of fermentation residues. The added salt is recovered together with the pulping chemicals in the BL. As a result of the NaOH addition, the low-blend (20 %) mix finally consisted of 3.8 % fermentation residues, 19 % NaOH, and 77 % black liquor (on dry mass basis), and the high-blend (35%) mix consists of 6.5 % fermentation residues, 33 % NaOH, and 61 % black liquor. For the GHG emissions calculations, 0.48 kg CO₂-eq. kg⁻¹ NaOH was assumed (BioGrace 2012, updated GWP100 for methane).

For the alternative use of fermentation residues for electricity generation, a conversion efficiency of 0.3 (based on Naturvårdsverket 2005) was used to estimate the amount of electricity generated from each energy unit of fermentation residues. The energy content of fermentation residues was 18 MJ kg⁻¹ (80 % DM) and thus 1 kg of fermentation residues at 80 % DM yields 6 MJ of electricity. No additional energy inputs were considered. Notably, this scenario for fermentation residues is distinctly less elaborated as compared to the conversion alternatives for the other by-product feedstocks in this report. This is explained by the lack of suitable data for a biomass-to-biofuel conversion process for fermentation residues within this project.

a) Co-gasification of BL and fermentation residues from ethanol plant (EtOH P).



b) Alternative use of fermentation residues: combustion with power generation.

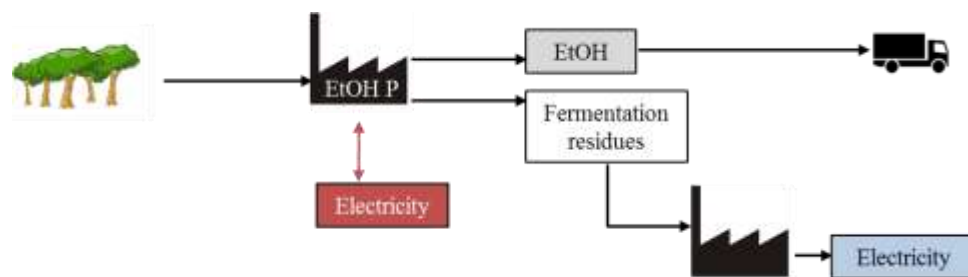


Figure B 4. Schematic overview of the studied system for co-gasification of black liquor (BL) with fermentation residues from lignocellulosic ethanol production (a). In the reference case, the alternative use of fermentation residues is combustion with power generation (b).

RESULTS AND DISCUSSION

Black liquor and pyrolysis liquid

The GHG emissions vary from case to case. For the low-blend cases, the reference case resulted in slightly higher total GHG emissions (see Figure B5), but because the reference cases also produced more methanol, the specific GHG emissions were largely equal between the cases. Using the RED calculation method, GHG emissions were 10-11 g CO₂-eq. MJ⁻¹, and contrastingly, the results from the ISO method were 20-22 g CO₂-eq. MJ⁻¹. For the higher-blends cases, the specific GHG emissions were higher in the co-gasification cases than in the reference cases: 14-15 g CO₂-eq. MJ⁻¹ (as compared to 9-10 g CO₂-eq. MJ⁻¹) using the RED method and approximately 32 g CO₂-eq. MJ⁻¹ (as compared to 22 g CO₂-eq. MJ⁻¹) following the ISO method. Thus, the RED method resulted in GHG emissions below the RED reduction target of 60 % reduction as compared to a fossil fuel reference (38 g CO₂-eq. MJ⁻¹), while the ISO results for the high-blend cases put the emissions at that approximate level. From this comparison, it was obvious that method choices were decisive to the total GHG emissions of the produced methanol, while the compared scenarios, including the comparison of grade AA or crude methanol, yielded somewhat similar results.

Out of the total GHG emissions of the produced methanol, the main contributing processes are electricity for gasification and collection and transportation of forest residues. As an example of the uncertainty connected to the future scenario where the considered pulp mill and biofuel plant may be built, the electricity production in the Nordic countries could change, rendering the data for the Nordic electricity mix used in these calculations inaccurate. The results of the calculations presented here should be viewed as first screening results, and therefore be interpreted with caution.

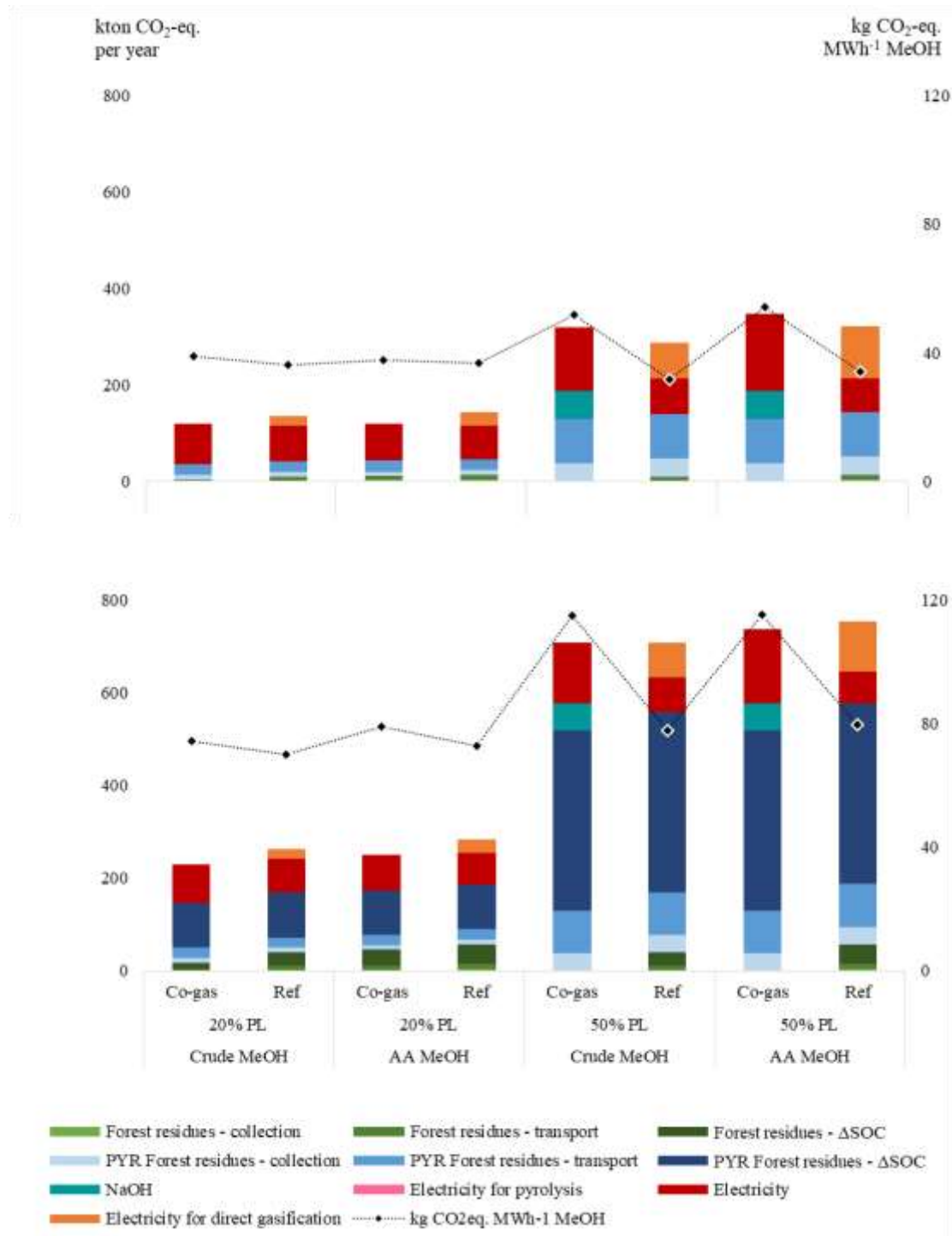


Figure B 5. GHG emissions from co-gasification of black liquor and pyrolysis liquid, calculated following the RED method (upper) and the ISO method (lower).

Black liquor and glycerol

In terms of GHG emissions, both RED and ISO methods resulted in higher total GHG emissions for the reference scenarios (Figure B6). This in turn leads to higher GHG emissions from the fuels (methanol and methane) produced in the reference scenarios: 12 and 17 g CO₂-eq. MJ⁻¹ calculated with RED and ISO methods, respectively, as compared to the methanol produced in the co-gasification scenarios: 5-8 and 10-12 g CO₂-eq. MJ⁻¹ for RED and ISO calculations. Notably, the leakage of methane from the digestion scenario contributed significantly to the total GHG emissions of the reference scenarios, especially for the high-blend (50 % glycerol) cases. The actual methane slip varies between plants and practices, and the leakage from upgrading could potentially be reduced e.g. by oxidation of methane (Tufvesson et al. 2013). It is thus important to note that the resulting GHG emissions from the reference scenario could vary significantly with altered assumptions for methane slip. Still, the results showed emissions below the current RED 60 % reduction target compared to a fossil fuel reference (38 g CO₂-eq. MJ⁻¹). No significant differences could be found in comparing the production of grade AA or crude methanol.

Grid electricity was a main contributor to the total GHG emissions for the gasification cases. Applying the ISO method, upstream emissions of glycerol was also important. Regarding GHG emissions from grid electricity, the same statement can be made for this co-gasification case as for the previous; the unknown future development of the energy system will find current assumptions inaccurate, and therefore, results should be interpreted with caution. Regarding upstream emissions of glycerol, the market situation for crude glycerol could affect what is considered a reasonable allocation to RME and glycerol respectively. Assumptions on rapeseed cultivation also influence the total GHG emissions. Considering that the current assumptions have a significant impact on the ISO results, upstream impacts should be taken into account when considering future uncertain scenarios for black liquor co-gasification with glycerol.

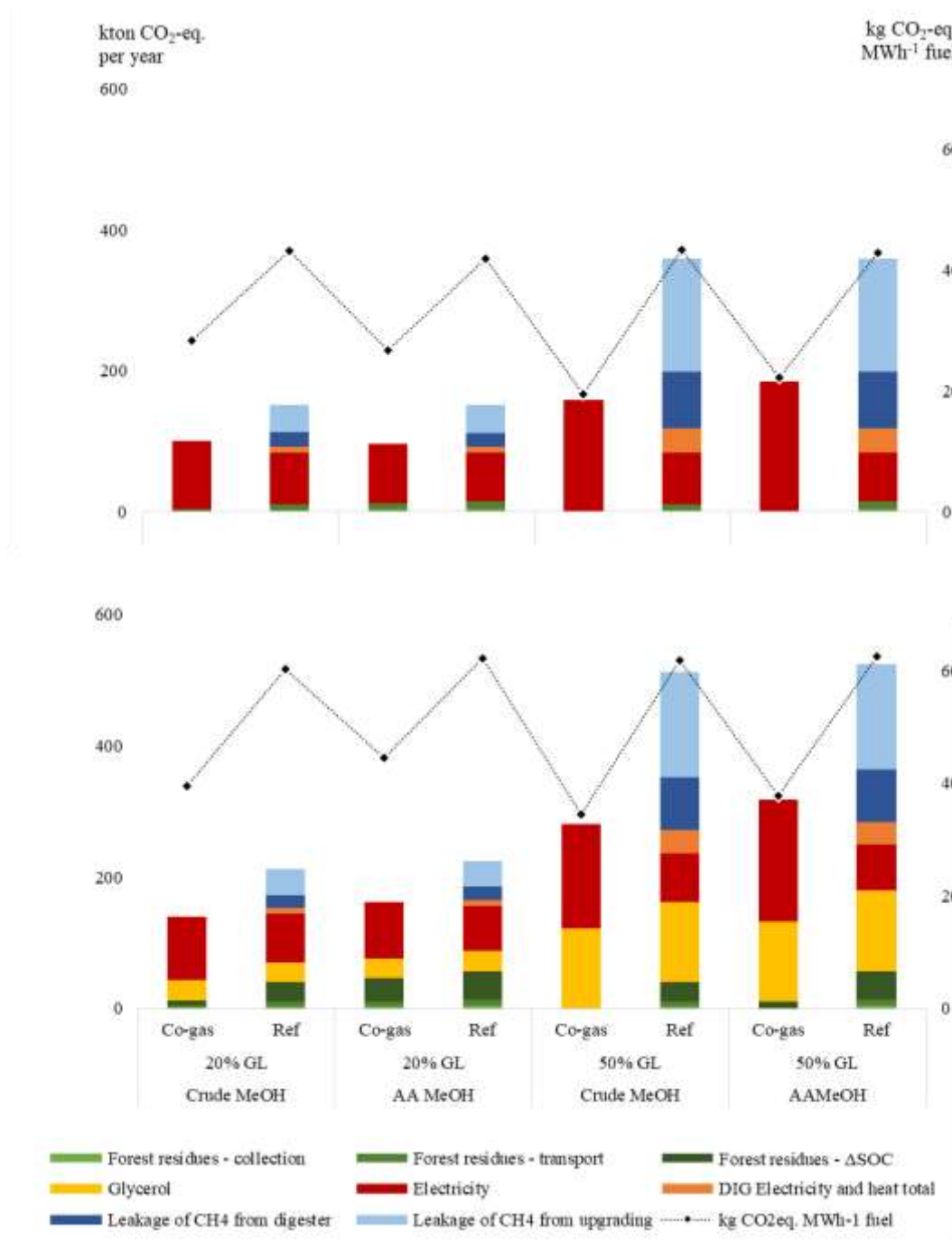


Figure B 6. GHG emissions from co-gasification of black liquor and glycerol, calculated following the RED method (upper) and the ISO method (lower).

Black liquor and fermentation residues

The comparison of GHG emissions from the different cases vary depending on the calculation method used (Figure B7). Using the RED calculation method, net electricity for gasification was the main contributor to the total GHG emissions (roughly 7-11 g CO₂-eq. MJ⁻¹) in all cases. The GHG emissions per MJ methanol or electricity produced were consequently slightly lower in the reference scenarios. On the contrary, the results applying the ISO method showed changes in soil organic carbon as another important contributing process, and render the co-gasification and reference scenarios similar in terms of GHG emissions per MJ of methanol or electricity produced

(roughly 10-14 g CO₂-eq. MJ⁻¹). This aspect also influences the comparison of crude to grade AA methanol, where grade AA methanol resulted in slightly higher GHG emissions. Here, the method for calculation was decisive both to the final results in terms of total GHG emissions, and to the conclusion regarding comparison of co-gasification and reference handling.

With grid electricity as an important source for GHG emissions in all cases and scenarios, again, the future development of the energy system is decisive to the final GHG emissions from the studied systems. What has been said regarding the other studied systems is also valid here; results should be viewed as a first set of screening results, and interpreted with caution. With this in mind, it can be noted that the present results for methanol in the co-gasification scenario showed emissions below the current RED 60 % reduction target compared to a fossil fuel reference (38 g CO₂-eq. MJ⁻¹).

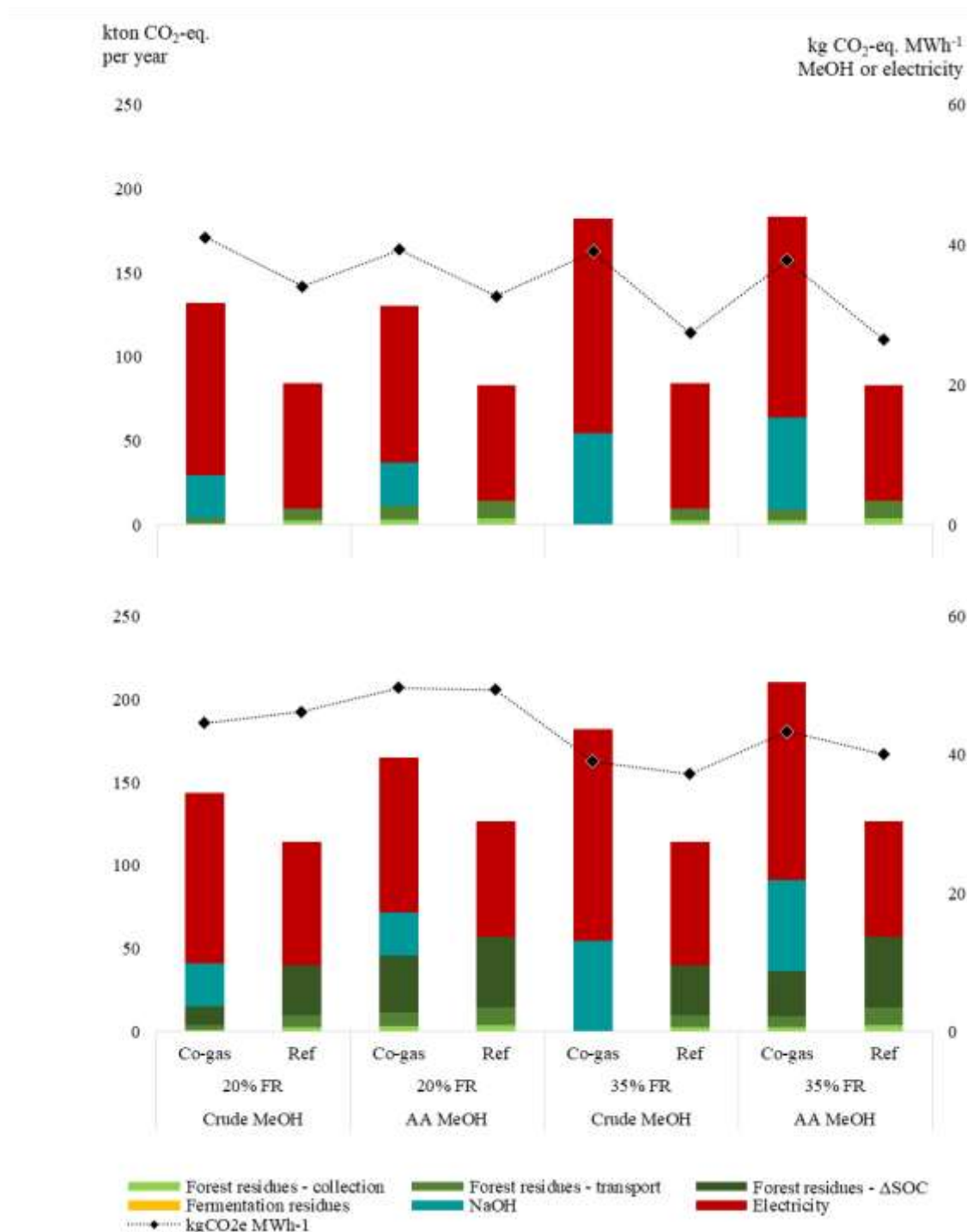


Figure B 7. GHG emissions from co-gasification of black liquor and fermentation residues, calculated following the RED method (upper) and the ISO method (lower).

COMMON OBSERVATIONS AND UNCERTAINTIES

Within each of the studied co-gasification systems, the method choices for calculating life-cycle GHG emissions largely influenced the total GHG emissions of the fuels produced. In the case of co-gasification of black liquor and fermentation residues, also the comparison between co-gasification and the reference scenarios was influenced. Using the RED method, the fuels studied in this

report resulted in emissions savings of 82-94 % compared to the RED fossil fuel reference. By calculating GHG emissions according to the ISO standards, the corresponding GHG emissions savings for the same fuels were in the range of 62-88 %.

Notably, the choice of calculation method influences the resulting GHG emissions significantly. It should also be noted that while the RED method results represent the current policy guidelines, they do not necessarily represent the most accurate GHG emissions of the studied fuel production systems. While the results applying the RED method in this study meet the current policy goal of 60 % reduction of GHG emissions as compared to a fossil reference, this should be re-evaluated with future policy changes.

One such potential changed policy scenario was illustrated by the results following the ISO guidelines, where implications of including changes to soil organic carbon, and upstream GHG emissions of certain by-products, were presented. The assumption regarding upstream emissions of residual biomass could potentially be critical to results for residue-based products (Olofsson, 2017). Since the value of residual biomass is likely to increase in an expanded circular bioeconomy, this could motivate an increased allocation of the environmental impact of the main production systems and also to the residual biomass streams. An example of including upstream GHG emissions was shown in the present work for glycerol, and future assessments should re-evaluate such assumptions.

In the light of uncertainties connected to future energy and resource scenarios, inventory data, and calculation approaches, the results presented here cannot carry decisive conclusions regarding the best use of the studied biomass resources from a GHG emissions or fuel production point of view.

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APPENDIX 2. MASS BALANCES

The following sections, and tables C 1 to C 5, show the mass balances of the studied gasification systems. For each co-gasification case, 173.6 tons of black liquor (wet basis) was input to the bio-fuel plant hourly. The plant was assumed to run 8000 h per year.

CO-GASIFICATION OF BLACK LIQUOR AND PYROLYSIS LIQUID

Table C 1. Mass and energy balance for co-gasification of black liquor and pyrolysis liquid.

Co-gasification: black liquor and pyrolysis liquid						
		[Units]	20% Pyrolysis liquid		50% Pyrolysis liquid	
			Crude MeOH	AA MeOH	Crude MeOH	AA MeOH
Flows going IN	Pulp wood	[t/h]	107	107	107	107
		[MJ/kg]				
		ds [%]				
	Bark	[t/h]db	16	16	16	16
		[MJ/kg]db	19.56	19.56	19.56	19.56
		ds [%]				
	Fuel biomass	[t/h]	9	24	0	0
		[MJ/kg]ds	18.97	18.97	18.97	18.97
		ds [%]				
	Pyrolysis liquid	[t/h]ar	43	43	174	174
		[MJ/kg]	17.41	17.41	17.41	17.41
		ds [%]	75	75	75	75
	NaOH	[t/h]ar	x	x	20	20
		water [%]	x	x	25	25
	Electricity	[MW]el	104	95	131	159
Flows going OUT	Electricity	[MW]el	0	0	0	0
	Pulp	[Adt/h]	82	82	82	82
	Crude MeOH	[t/h]	66	x	132	x
		[MJ/kg]	21	x	21	x
	AA MeOH	[t/h]	x	62	x	125
		[MJ/kg]	x	23	x	23
	Hot water	[t/h]	1323	1323	2596	2596
		[°C]	95	95	95	95

CO-GASIFICATION OF BLACK LIQUOR AND GLYCEROL

Table C 2. Mass and energy balance for co-gasification of black liquor and glycerol.

Co-gasification: black liquor and glycerol						
			20% Glycerol		50% Glycerol	
		[Units]	Crude MeOH	AA MeOH	Crude MeOH	AA MeOH
Flows going IN	Pulp wood	[t/h]	107	107	107	107
		[MJ/kg]				
		ds [%]				
	Bark	[t/h]db	16	16	16	16
		[MJ/kg]db	19.56	19.56	19.56	19.56
		ds [%]				
	Fuel biomass	[t/h]	6	24	0	7
		[MJ/kg]	18.97	18.97	18.97	18.97
		ds [%]				
	Glycerol	[t/h]ar	43	43	174	174
		[MJ/kg]				
		ds [%]	99.6	99.6	99.6	99.6
	Electricity	[MW]	96	85	158	184
Flows going OUT	Electricity	[MW]el	0	0	0	0
	Pulp	[Adt/h]	82	82	82	82
	Crude MeOH	[t/h]	75	x	175	x
		[MJ/kg]	21	x	21	x
	AA MeOH	[t/h]	x	71	x	166
		[MJ/kg]	x	23	x	23
	Hot water	[t/h]	1402	1402	2999	2999
		[°C]	95	95	95	95

CO-GASIFICATION OF BLACK LIQUOR AND FERMENTATION RESIDUES

Table C 3. Mass and energy balance for co-gasification of black liquor and fermentation residues.

Co-gasification: black liquor and fermentation residues						
			20% Fermentation residues		35% Fermentation residues	
		[Units]	Crude MeOH	AA MeOH	Crude MeOH	AA MeOH
Flows going IN	Pulp wood	[t/h]	107	107	107	107
		[MJ/kg]				
		ds [%]				
	Bark	[t/h]db	16	16	16	16
		[MJ/kg]db	19.56	19.56	19.56	19.56
		ds [%]				
	Fuel biomass	[t/h]	8	24	0	19
		[MJ/kg]	18.97	18.97	18.97	18.97
		ds [%]				
	FR	[t/h]ar	43	43	93	93
		[MJ/kg]	19.33	19.33	19.33	19.33
		ds [%]	80	80	80	80
	NaOH	[t/h]ar	9	9	19	19
		Water [%]	20	20	20	20
	Electricity	[MW]	102	93	127	119
Flows going OUT	Electricity	[MW]el	0	0	0	0
	Pulp	[Adt/h]	82	82	82	82
	Crude MeOH	[t/h]	69	x	100	x
		[MJ/kg]	21	x	21	x
	AA MeOH	[t/h]	x	65	x	95
		[MJ/kg]	x	23	x	23
	Hot water	[t/h]	1330	1330	1861	1861
		[°C]	95	95	95	95
	Extra LPS	[t/h]	0	0	16	6

GASIFICATION OF BLACK LIQUOR (REFERENCE CASE)

For each case, 174 tons of black liquor (DM) is input to the system, producing either crude or grade AA methanol.

Table C 4. Mass and energy balance for gasification of black liquor.

Black liquor gasification (reference)				
		[Units]	Crude MeOH	AA MeOH
Flows going IN	Pulp wood	[t/h]	107	107
		[MJ/kg]		
		ds [%]		
	Bark	[t/h]db	16	16
		[MJ/kg]db	19.56	19.56
		ds [%]		
	Fuel biomass	[t/h]	21	30
		[MJ/kg]ds	18.97	18.97
		ds [%]		
	Electricity	[MW]el	74	69
Flows going OUT	Electricity	[MW]el	0	0
	Pulp	[Adt/h]	82	82
	Crude MeOH	[t/h]	42	x
		[MJ/kg]	21	x
	AA MeOH	[t/h]	x	40
		[MJ/kg]	x	23
	Hot water	[t/h]	867	867
		[°C]	95	95

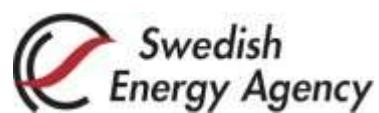
DIRECT GASIFICATION OF LOGGING RESIDUES (REFERENCE CASE)

Table C 5. Mass and energy balance for direct gasification of logging residues.

Gasification of logging residues (reference case)						
			Corresponding to 20 % PL case		Corresponding to 50 % PL case	
		[Units]	Crude MeOH	AA MeOH	Crude MeOH	AA MeOH
Flows going IN	Fuel biomass	[t/h]	64	64	257	257
		[MJ/kg]ds				
		ds [%]	100	100	100	100
	Electricity	[MW]el	19	28	74	109
Flows going OUT	Electricity	[MW]el	0	0	0	0
	Crude MeOH	[t/h]	38	x	153	x
		[MJ/kg]	21	x	21	x
	AA MeOH	[t/h]	x	36	x	145
		[MJ/kg]	x	23	x	23
	Hot water	[t/h]	304	304	1230	1230
		[°C]	95	95	95	95



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