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1	Techno-economic study of NMMO
2	pretreatment and biogas production from
3	forest residues
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20 ABSTRACT

Biogas is nowadays getting more attention as a means for converting wastes and lignocelluloses 21 to green fuels for cars and electricity production. The process of biogas production from N-22 methylmorpholine oxide (NMMO) pretreated forest residues used in a co-digestion process was 23 economically evaluated. The co-digestion occurs together with the organic fraction of 24 municipal solid waste (OFMSW). The process simulated the milling of the lignocelluloses, 25 NMMO pretreatment unit, washing and filtration of the feedstock, followed by an anaerobic 26 27 co-digestion, upgrading of the biogas and de-watering of the digestate. The process also took into consideration the utilization of 100,000 DW (dried weight) tons of forest residues and 28 200,000 DW tons of OFMSW per year. It resulted in an internal rate of return (IRR) of 24.14% 29 prior to taxes, which might be attractive economically. The cost of the chemical NMMO 30 31 treatment was regarded as the most challenging operating cost, followed by the evaporation of the washing water. Sensitivity analysis was performed on different plant size capacities, treating 32 33 and digesting between 25,000 and 400,000 DW tons forest residues per year. It shows that the minimum plant capacity of 50,000 DW tons forest residues per year is financially viable. 34 Moreover, different co-digestion scenarios were evaluated. The co-digestion of forest residues 35 together with sewage sludge instead of OFMSW, and the digestion of forest residues only were 36 shown to be non-feasible solutions with too low IRR. Furthermore, biogas production from 37 forest residues was compared with the energy produced during combustion. 38

39

40 KEYWORDS: anaerobic digestion, NMMO pretreatment, lignocellulose, forest residues,
41 economic analysis

42

43 HIGHLIGHTS

• Biogas from co-digestion with pretreated forest residues was simulated and evaluated

• Plant capacity of > 50,000 DW tons/year forest residues is financially feasible

- The cost of the NMMO was regarded as the largest operating expenditure
- Biogas production was compared with the energy produced during incineration

49 1. INTRODUCTION

The global market and demand for biogas as a vehicle fuel, electricity production, and even as 50 a heating energy source has had a positive trend. The biogas is produced in household digesters 51 to provide cooking or lightening energy to replace kerosene or LPG, while the larger plants 52 burn it in gas engines to produce electricity or upgrade it to almost pure methane to inject in the 53 gas grids or compress it to CBG (compressed biogas) and sell as car fuel. The traditional 54 substrates utilized for biogas production are municipal solid waste, organic wastes from 55 industrial and agricultural activities, as well as high strength wastewater are. However, these 56 sources are limited, and there is a demand for the development of new technologies utilizing 57 other substrates. Lignocellulosic-rich materials have a great potential as an alternative feedstock 58 for anaerobic digestion, since they are found in high abundance globally. 59

The degradation of lignocelluloses into biogas is a complicated process, since lignocelluloses 60 have a recalcitrant structure which is naturally designed to prevent enzymatic degradation. 61 Lignocelluloses are formed in a compact and crystalline structure and often contain a high 62 amount of lignin. In order to permit degradation of these materials in an anaerobic digester, the 63 64 structure has to open up and/or the lignin has to be degraded or removed. This can be performed by using different pretreatment methods [1], such as mechanically, e.g., by milling; physically 65 66 by steam explosion or radiation; chemically by acids, bases or solvents; and biologically by enzymes or fungi [1-3]. 67

Solvent pretreatment on lignocelluloses was shown to be an effective method due to the low 68 degradation of the carbohydrates in the material under the applied, relatively mild conditions. 69 Furthermore, pretreatment with a solvent does not require neutralization, and almost a complete 70 71 recirculation of the treating chemical is possible [4]. The pretreatment using the solvent N-72 methylmorpholine oxide (NMMO) has previously been studied on bagasse [5] and on spruce [6] for ethanol production, and on spruce, rice, and triticale straws [7] as well as pure cellulose 73 [8] for biogas production. NMMO is an organic solvent that interrupts inter- and intra-molecular 74 bonds [9] in the lignocelluloses, making the carbohydrates of the material more accessible and 75 thereby facilitating the enzymatic degradation. NMMO is an environmentally friendly cellulose 76 solvent, and used in industrial scale in the lyocell process [10, 11], where cellulose fibers are 77 treated to produce textile. Since no toxic compounds are produced within the NMMO 78 pretreatment and the recirculation of the solvent is possible [10, 12], this process can be 79 regarded as environmentally friendly. 80

Techno-economic analysis is a useful tool to examine the profitability and performance of a 81 proposed process. Recently, Shafiei et al [6] performed a techno-economical study on 82 bioethanol production from NMMO pretreated wood. They found that the process is feasible 83 when bioethanol production is combined with a subsequent biogas production utilizing the 84 pentoses. Conversion of lignocellulosic pentoses to ethanol is one of the obstacles in the 85 utilization of lignocelluloses to ethanol, since the ordinary industrial yeast species are unable to 86 assimilate pentoses [13]. Furthermore, the production of biogas from lignocelluloses has several 87 advantages compared to bioethanol production, since the overall energy efficiency is much 88 89 higher in biogas production compared to that in ethanol production [14].

The focus of this study was therefore to develop a feasible industrial process for NMMO 90 pretreatment and subsequent utilization of forest residues (branches, tops, barks, and needles) 91 92 in anaerobic digestion. Forest residues were selected because they are the most abundant lignocellulosic waste stream in Sweden, and several other countries. In 2008, 1.6 Mtons total 93 solids (TS) /year of the tree tops and branches were delivered from the forests in Sweden, and 94 this is expected to increase to 3.5 Mtons total solids /year by 2018 [15]. Moreover, the total 95 energy potential of bioenergy production from the forest is calculated as being 49 TWh [15]. 96 An industrial scale process was designed and simulated using SuperPro Designer[®] 8.0 97 simulation software (Intelligen, Inc., NJ, USA) based on unpublished biomethane potential test 98 (BMP) experimental data provided by Kabir et al. [16]. A process including an NMMO 99 pretreatment step with filtration, evaporation and recirculation of NMMO and washing water 100 together with a following co-digestion step was evaluated to determine economic feasibility 101 and profitability, such as capital costs for the total plant, annual operating costs, and unit costs. 102 Finally, sensitivity analyses were performed on different scenarios, where effects of the plant 103 size, different co-digestion set-ups as well as the methane price and the water consumption were 104 evaluated. 105

106 2. PROCESS DEVELOPMENT AND FINANCIAL ANALYSIS

107 2.1 Process description

A novel process of the NMMO pretreatment of forest residues prior to anaerobic digestion was developed. The process includes the feedstock handling, pretreatment by NMMO, anaerobic digestion, and upgrading of the biogas as well as the dewatering of the digestate. It is assumed that the plant is located close to a power plant, so that steam and electrical power are readily available. It is further assumed that the plant is situated in Sweden with a high availability of forest residues. The type of forest residues investigated in this study includes the rejected topsand branches.

The base case is constructed for 100,000 tons DW (dry weight) forest residues per year. However, capacities ranging from 25,000 to 400,000 tons DW forest residues/year were also studied. The plant is in operation for 7,920 h/year, and the construction material was chosen to be stainless steel 304. The cost index was set at 2012.

119

120 2.2 Pretreatment unit

The forest residues arrive at the plant in truck trailers, where the price of the feedstock includes 121 122 the handling all the way to the plant. The feedstock contains 42% carbohydrates, 44% lignin, 75% total solids (TS), and 64% volatile solids (VS) [17]. The forest residues have a C/N ratio 123 124 of 325 [18]. The raw material is then placed into a grinder, which reduces the size of the biomass to 2 mm. After grinding, the biomass is conveyed to the pretreatment unit. The pretreatment is 125 performed using 85% NMMO solution in water for 12 h at 90°C. During the pretreatment, the 126 lignocellulosic structure is opened up, resulting in less intra-molecular linkages and less 127 cellulosic crystallinity [9]. The pretreated biomass is then washed with water and filtered using 128 a rotary vacuum filtration unit (Figures 1 and 2). The NMMO-solution is then evaporated back 129 to 85% for reuse in the pretreatment unit. The recovery in the washing step is expected to be 130 99.5%. The use of the rotary vacuum filtration allows for a minimum usage of water during the 131 washing, in order to save energy in the following evaporation unit. Previous experimental 132 studies were performed with 500 mL washing water for 200 g NMMO/biomass mixture [16], 133 where these conditions were applied in the base case of the simulation study. The evaporation 134 135 unit was designed with a mechanical vapor design (MVR). The MVR design with two effects and two compressors was found to be the most energy efficient and an economically beneficial 136 137 alternative for the evaporation of NMMO water solution in a previous investigation, focusing on NMMO pretreatment of spruce prior to ethanol production [6]. The same design for the 138 139 evaporation step was applied in this study.

140

141 2.3 Biogas and digestate production

The washed and pretreated forest residues are mixed with the organic fraction of municipal 142 waste (OFMSW, Figures 1 and 2), in order to a achieve a C/N ratio of between 20-30 which is 143 regarded as the optimum ratio [19]. Two-thirds of the OFMSW and one-third of the forest 144 residues are used in the base case, which results in a C/N ratio of 30. The OFMSW in the 145 simulation consist of 60% carbohydrates, 17% fats, 8% proteins based on the dried weight, and 146 the water content was estimated to be 67 % water. The cost of OFMSW is set to zero. The 147 methane yield of similar substrate mix was 0.470 Nm³/kg VS [20], which corresponds to a 148 conversion rate of 86.7%. The methane production from forest residues is based on 149 experimental results from lab scale BMP tests showing a yield of 0.137 Nm³ CH₄ per kg total 150 solids of forest residues [16], which corresponds to a conversion rate of 73.4%. The two 151 fractions are together passed through a screw press prior to the anaerobic digester, together with 152 extra water in order to reach a TS of 12% in the incoming stream. The digester runs at 153 154 thermophilic conditions (55°C) with a hydraulic retention time of 20 days. It is a fixed roof storage tank, which allows for mixing, constructed of stainless steel. The gas produced is a 155 156 mixture of the main components methane and carbon dioxide, and trace amounts of some other components, such as hydrogen sulfide, nitrogen, and hydrogen, which are neglected in the 157 158 study. The gas produced in the anaerobic digester is upgraded to 98% methane content, using the water scrubber technique. The water scrubber technique is regarded as a low cost technique 159 [21], and is globally the most widespread upgrading technique [22]. This upgrading step 160 consists of a gas compressor, an absorption tower where the carbon dioxide is absorbed in 161 water, and a degasification tower, where the carbon dioxide and water are separated. The 162 upgraded methane is then injected into the biogas/natural gas grid. The solid residuals 163 remaining from the process, so called digestate residues, are dewatered in a centrifugal 164 separator to 45% TS, together with 10 kg flocculating agent polyacrylamide per ton TS, in order 165 to improve the dewatering process [23]. The solid fraction after the dewatering step is lignin-166 rich, which has a high heating value, and can be used as fuel for combustion in combined heat 167 and power (CHP) plants [24]. In this study, the dewatered digestate is sold to CHP plants. 168 However, due to the high nutrient value, the digestate residue can also be used as a fertilizer in 169 agriculture or on forestland. Consequently, the dewatering process would then be unnecessary. 170

171

172 **2.4 Process simulation and economic calculations**

SuperPro Designer[®] 8.0 (Intelligen, Inc., NJ, USA, licensed to the University of Borås) was 173 used for the simulation of the main steps of the process. The software performs the rigorous 174 material and energy balance calculations. The purchase costs of the equipment were calculated 175 176 with the built-in software calculations, except for the purchase cost of the tanks, which was calculated according to Turton et al [25]. Other than the purchase costs, SuperPro Designer 177 estimates the cost for the installation, the process piping, instrumentation, insulation, electrical 178 utilities, buildings, yard improvements, and auxiliary facilities. The total direct plant cost (DC) 179 is a sum of these costs and was 329% of the equipment purchase cost at base conditions. The 180 total indirect plant costs, such as engineering (25% of DC) and construction fee (35% of DC) 181 was based on the equipment purchase cost, and was obtained by the above- mentioned software. 182 183 The fixed capital investment (FCI) was calculated as a sum of the direct costs, the indirect costs, the contractor's fee, and the contingency. The contractor's fee and contingency were estimated 184 185 to be 5% and 10%, respectively, of the sum of the direct cost and the indirect cost together [26]. The project is regarded as 100% equity financed. The project life is set to 20 years and the 186 187 depreciation period to 10 years. The construction period is set to 30 months and a startup period of 4 months is used. The working capital was assumed to be 5% of the fixed capital investment 188 [27], and the cost index for all calculations was set at 2012. 189

The annual operating cost was calculated as the sum of the expenses for raw materials, utilities, 190 labor, waste management, and facility dependent cost and can together with the product prices 191 be found in Table 1. The maintenance and insurance costs are regarded as facility dependent 192 operating costs, and are together 1 and 2%, respectively, of the total plant capital costs [28-30]. 193 The methane price used in the present study was the price of methane sold in the market in 194 Sweden, minus the cost for the connection and distribution into the gas grid, including 195 compression and cost for tank stations. The methane price used in this study was 1.895 euro/kg 196 [31]. A value of 22% taxation rate is assumed, which is the current corporation tax in Sweden 197 since 2013 [32]. 198

Furthermore, the plant was divided into sections in order to determine the cost distribution forthe different parts of the plant. These calculations were performed using the base case.

201

202 2.5 Sensitivity analysis

Different plant sizes were investigated in a sensitivity analysis in order to study the effect of the capacity on the construction and production costs. Plant sizes with the feed capacity of 25,

50, 100, 200, and 400% of the base case were studied. The cost prediction of the total investment 205 costs, annual operating costs, and production cost per unit methane produced, as a function of 206 the plant capacity was studied and simulated. Cash flow analysis was performed where the net 207 present value (NPV) was set to zero and the process time was equal to 20 years. The internal 208 209 rate of return (IRR) was calculated, and was regarded as being financially feasible at 15% rate of return (IRR) or higher, in order to cover the firms costs of raising funds and making a 210 sufficient profit [33]. The IRR is the discount rate, when the NPV is set to zero and was 211 calculated as [33]: 212

213
$$NPV = \sum_{t=0}^{n} \frac{A_t}{(1+r)^t} = 0$$

215 *NPV, t, n,* A_t and *r* are net present value, project year, total project lifetime, the cash flow in 216 year *t*, and the discount rate, respectively.

217

The cash flow analysis was performed in order to study the effect of the methane price, the water consumption in the washing step following the NMMO pretreatment, and the price of the feedstock on the economic feasibility of the process under different scenarios. A co-digestion study where the forest residues were co-digested with sewage sludge instead of OFMSW was also performed, as well as a scenario where only forest residues were digested.

223

3. RESULTS

225 **3.1 Process development and economic calculations**

The plant was divided into five different sections (1) the NMMO pretreatment, (2) the filtration 226 and evaporation following the NMMO pretreatment, which also includes the recirculation of 227 water and NMMO, (3) the anaerobic digestion of both forest residues and OFMSW, (4) the 228 upgrading of the biogas, and (5) the dewatering of the lignin-rich digestate. The fixed capital 229 investment (FCI) for the different sections can be found in Figure 3. The most capital-intensive 230 sections are the anaerobic digestion, followed by the filtration and evaporation, and the 231 upgrading. Auxiliary capital investments, buildings, and yard improvements are excluded from 232 the calculation. 233

A block flow diagram of the process is presented in Figure 1, which gives an overview of the 234 process. The material composition of the streams in the block flow diagram is presented in 235 Table 2. The developed process flow sheet, showing the equipment used in all processes, is 236 237 presented in Figure 2. All process steps were run continuously, except the NMMO-pretreatment 238 reactor, which was operated in batch mode. For this purpose, four staggered NMMO pretreatment reactors of each 970 m³ were used to perform 1975 batch pretreatments per year. 239 The base case was considered to pretreat and utilize 100,000 DW tons forest residues/year, 240 together with 200,000 DW tons OFMSW/year, and the plant was calculated to produce about 241 975 GWh (98 MNm³) methane per year. The produced amount of dewatered digestate and 242 carbon dioxide are 290 and 133 kt/year, respectively. The consumption of electricity was 48 243 GWh per year, steam 355 GWh, and water 5,043 kt per year. In order to pretreat 100,000 DW 244 tons forest residues per year, six batches of NMMO treatment per 24 h were performed. The 245 246 fixed capital investment (FCI), is a sum of direct fixed capital, working capital, and startup cost, and was calculated for the base case as being 145,053,000 €. The annual operating cost is a sum 247 248 of raw materials, labor costs, energy and power, waste management, as well as facility dependent costs. For the base case, this cost was calculated as being 103,810,000 €/year. The 249 250 total revenue per year is a sum of the revenues of produced methane, carbon dioxide, and the dewatered lignin-rich digestate. The annual revenue for the base case was calculated as 251 136,179,000 €/year. This gives a net profit value (taxes and depreciation are included) of 252 181,333,000 €, at 7.0% interest rate over 20 years project lifetime. A cash flow analysis, with 253 the net profit value set at zero resulted in an internal rate of return of 24.14% prior to taxes, and 254 255 20.39% after taxes, at a process time of 20 years.

The costs for the distribution of the upgraded methane into the distribution gas grid, were 256 calculated according to as described by Benjaminsson and Linné [31]. The authors performed 257 a techno-economic study of 300 GWh biogas plant in Sweden. For this size of plant, the cost 258 of a gas pipeline for 40 km connected to the distribution gas grid was 0.001 €/kWh, the 259 260 distribution cost 0.007 €/kWh, and the compression and tank station cost was 0.012 €/kWh, a total of 0.020 €/kWh. Calculating with an 8% price increase in Sweden between 2007 and 2012 261 262 [34], the price for gas grid distribution, compression, and tank stations are set to 0.285 €/kg 263 methane.

The total annual operating costs divided into different cost items are presented in Figure 4. The costs of the raw materials have the highest share of operating costs, followed by facility dependent costs, which include maintenance, depreciation, insurance, and other factory expenses. The cost for the NMMO corresponds to 80% of the material cost for the base case with 99.5% recirculation, and the cost for the forest residues corresponds to 15%. The annual operating cost divided into the different sections is presented in Figure 5, where the price of the materials is excluded. Filtration and evaporation represent the biggest part of the annual operating costs, followed by the anaerobic digestion, where the costs for materials are excluded.

272

273 **3.2 Sensitivity analysis**

Different plant sizes were investigated in a sensitivity analysis in order to study the effect of 274 the plant capacity on the construction and production costs. Plants treating 25, 50, 100, 200, 275 and 400 thousand DW tons forest residues per year were studied in co-digestion with 50, 100, 276 200, 400, and 800 thousand DW tons OFMSW per year, respectively. All the estimations of 277 total investment costs, annual operating costs and production cost per unit methane produced, 278 as a function of the plant capacity is presented in Figure 6. The revenue per unit was calculated 279 as being 2.12 €/kg produced methane, which is higher than the production cost for all plant 280 sizes. However, a cash flow analysis of the five different plant size scenarios show that only 281 plant capacities of 50,000 tons per year and above are financially viable with an IRR over 15%. 282 This is in contrast to the IRR of the plant size of 25,000 tons per year, which was 5.08% prior 283 284 to taxes.

The economic feasibility of the process was further analyzed through different scenarios. The 285 effect of water consumption in the washing process following the NMMO pretreatment was 286 evaluated with 50% more and 50% less water consumption. The effect of 20% increase and 287 20% decrease on the methane price and the cost of feedstock was also calculated. Cash flow 288 289 analysis was performed and the resulting IRR's were compared with the base case and are presented in Figure 7. The water volume during the washing step following the NMMO 290 291 pretreatment has a large effect on the IRR. The use of more water during the washing step requires a larger and more expensive evaporation unit, which in turn results in a lower IRR. 292 293 Furthermore, the price of the produced methane has a large impact on the IRR, while the cost 294 of forest residues has a minor effect.

295

296 **3.3 Co-digestion scenarios**

In order to achieve a proper C/N ratio, forest residues can be co-digested with other nitrogen 297 rich substrates. Sludge from wastewater treatment (sewage sludge) has been studied as an 298 299 alternative co-digestion source. Due to the high nitrogen content, one part of sewage sludge together with two parts of forest residues result in an optimum C/N ratio of about 20, compared 300 with two parts of OFMSW and one part of forest residues in the base case (Table 3). In Sweden, 301 biogas plants get paid for the digestion of sewage sludge (Table 1), which will increase the unit 302 revenue. However, our calculations showed that the co-digestion with sludge results in a unit 303 production cost of 2.78 €/kg and a unit revenue of 2.75 €/kg. The IRR of the process was 304 calculated as being 3.52% (Table 3), which is lower than the financially feasible limit of 15% 305 and is therefore considered to be a non-feasible solution. 306

The process can be further designed to digest forest residues exclusively, which is not a real scenario, since it is unfavorable to digest forest residues by itself due to the low nitrogen content. However, the simulation of the pretreatment and anaerobic digestion of forest residues only can give us a better insight in the contribution of forest residues in the co-digestion process. With the exclusive digestion of forest residues, the IRR is negative (Table 3). The unit production cost has increased to 9.35 \notin /kg CH₄, while a higher unit revenue comes from the higher fraction of lignin in the digestate residue which was sold to a combustion plant.

314 Moreover, a sensitivity analysis has been performed in order to study the effect of different scenarios when only the forest residues are digested (Table 4). The effect of circulation of 315 316 NMMO was evaluated, as well as the effect of the methane price. An increase in the recirculation of NMMO from 99.5% to 99.99% will decrease the unit production cost by a 317 318 factor of three, while an increase in methane price increases the unit revenue. The unit revenue 319 was the same as the unit production cost, after an increase of the NMMO recirculation to 320 99.99%, together with a methane price increase of 25%. However, none of the present scenarios reached the targeted IRR of 15% (Table 4). 321

322

323 **3.4 Anaerobic digestion versus combustion**

The energy produced from anaerobic digestion of NMMO-pretreated forest residues can be compared with the energy production of the same amount of forest residues when incinerated. Combustion of the feedstock in a combined heat and power plant (CHP) will produce 17 MJ/kg TS, with the assumption of 90% efficiency in the CHP [35]. On the other hand, when biogas produced from the anaerobic digestion of only forest residues is utilized in a CHP with 90% efficiency [36], the energy generated can be calculated as being 12 MJ/kg TS. This is with the assumption that the lignin-rich residue from the anaerobic digestion is combusted separately, and the energy produced by this process is included in the above-mentioned calculation. Both processes are assumed to yield similar fractions of electricity and heat. It can, therefore, be concluded that the combustion of forest residues in CHP will yield about 1.5 times more energy compared with that in the anaerobic digestion.

There are another aspects that should also be considered when comparing anaerobic digestion 335 or combustion of forest residues. Utilization of these materials for vehicle fuel production is 336 only possible if they are converted to biogas. There is a large demand for alternative fuels 337 produced from renewable resources worldwide, since a considerable part of the total 338 339 greenhouse gas emissions originates from the transport sector [37]. Moreover, the organic nutrients cannot be retained and recycled back to soils after combustion, which in turn will 340 341 result in the removal of structural material from the soil. On the other hand, the digested residue left after anaerobic digestion can be utilized as a sustainable fertilizer. Additionally, combustion 342 343 is also connected with other serious problems as well, such as fly ash disposal and super heater 344 corrosion.

345

347 **4. DISCUSSION**

The anaerobic digestion of NMMO pretreated forest residues, co-digested with household 348 organic wastes in the base case is an economically viable process, with an IRR over 15%. The 349 analysis of different sections of the process shows that the price of the raw material, i.e., 350 NMMO, used for the pretreatment has the largest share of the costs. A challenge for the future 351 352 is to increase the recirculation of the NMMO, in order to limit the consumption of the raw material, and thereby the costs. Furthermore, evaporation of the washing water is a costly 353 process, and solving the technical challenge of using less washing water should further improve 354 the economy of the process. 355

In order to reach a financially viable process for the digestion of pretreated forest residues, the methane price needs to be increased substantially. This could perhaps partly be reached by incentives in order to increase the fraction of renewable vehicle fuels production, together with increasing oil price. The European Commission has set the goal that by 2020, 20% of the energy consumed and 10% of the vehicle fuels should be renewable [38]. Furthermore, the cost of gas injection into the gas grid and the cost of the tank stations are probably reduced with larger plant sizes as is the case in the present study.

The use of the biogas produced from the anaerobic digestion of the NMMO pretreated forest 363 residues in a CHP plant was shown to be a less attractive alternative compared with the 364 combustion of the same amount of forest residues. These two processes, however, produce 365 electricity and heat, while the anaerobic digestion process produces high-valued vehicle fuel. 366 Another advantage of producing biogas from the forest residues, compared with combustion, is 367 that the digestion of the feedstock results in a rich solid residue. In this study, this residue is 368 calculated as being sold to combustion plants. As an alternative, it could also be used as a 369 370 nutrient rich fertilizer. The use of the solid residue as a fertilizer is a sustainable way of recycling the nutrients back into the soil, and also structural material being placed back into the 371 372 soil.

Compared with co-digestion of forest residues with OFMSW, the digestion of only pretreated forest residues has a negative IRR. The scenario of digesting only forest residues however, is a fictive scenario, since an optimal C/N ratio of 20-30 should be reached for a sufficient nutritional balance in the digester. Therefore, a co-digestion of nitrogen-rich substrates together with forest residues is required. Many digesters with e.g. sewage sludge or protein-rich substrates have problems with a too low C/N ratio, which means a lack of carbohydrate-rich

substrates. Addition of carbon-rich materials, such as lignocelluloses, was previously shown to 379 both stabilize sensitive processes as well as result in good synergetic effects [39]. These 380 synergetic effects have implied higher methane yields when a lignocellulosic-rich material (i.e., 381 paper tube residuals) has been digested with nitrogen-rich substrate mixture compared to the 382 expected methane production calculated from the methane potentials of the single substrate 383 streams alone. The co-digestion of NMMO-pretreated forest residues with OFMSW has not yet 384 been experimentally studied, but similar synergetic effects can be assumed, which can lead to 385 higher methane yields and a more economically feasible process. The anaerobic co-digestion 386 387 of pretreated lignocelluloses has not yet implemented commercially, but could emerge in the 388 future.

389

390 5. CONCLUSIONS

The possible co-digestion of NMMO pretreated forest residues together with the organic 391 fraction of municipal solid waste is an economically feasible process with an IRR over 15%. In 392 order to avoid nitrogen deficiency, one-third of forest residues were co-digested with two- thirds 393 of OFMSW. Technical improvements such as increased recycling rate of the NMMO solvent, 394 as well as decreased water consumption in the washing step can further increase the economic 395 viability of the process. The co-digestion with sewage sludge instead of OFMSW resulted in 396 lower methane yields, which had a negative effect on the process economy. In general, the co-397 digestion circumstances, such as the type of feedstock used in the co-digestion and the 398 relationships between the different feedstocks have large consequences on the methane yields 399 and thereby the process economy. 400

401

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404

405 **7. REFERENCES**

406 [1] Johnson DK, Elander RT. Pretreatments for enhanced digestability of feedstocks. In: Himmel ME,
 407 editor. Biomass Recalcitrance: Blackwell Publishing; 2008. p. 436-53.

- 408 [2] Taherzadeh MJ, Karimi K. Pretreatment of lignocellulosic wastes to improve ethanol and biogas
 409 production: a review. International Journal of Molecular Sciences. 2008;9:1621-51.
- 410 [3] Hendriks ATWM, Zeeman G. Pretreatments to enhance the digestibility of lignocellulosic biomass.
- 411 Bioresource Technology. 2009;100:10-8.
- 412 [4] Shafiei M, Karimi K, Taherzadeh MJ. Pretreatment of spruce and oak by N-methylmorpholine-N-
- 413 oxide (NMMO) for efficient conversion of their cellulose to ethanol. Bioresource Technology.
- 414 2010;101:4914-8.
- 415 [5] Kuo C-H, Lee C-K. Enhanced enzymatic hydrolysis of sugarcane bagasse by N-methylmorpholine-
- 416 N-oxide pretreatment. Bioresource Technology. 2009;100:866-71.
- 417 [6] Shafiei M, Karimi K, Taherzadeh MJ. Techno-economical study of ethanol and biogas from spruce
- 418 wood by NMMO-pretreatment and rapid fermentation and digestion. Bioresource Technology.
- 419 2011;102:7879-86.
- 420 [7] Teghammar A, Karimi K, Sárvári Horváth I, Taherzadeh MJ. Enhanced biogas production from rice
- 421 straw, triticale straw and softwood spruce by NMMO pretreatment. Biomass and Bioenergy.
 422 2012;36:116-20.
- 423 [8] Jeihanipour A, Karimi K, Taherzadeh MJ. Enhancement of ethanol and biogas production from
- 424 high-crystalline cellulose by different modes of NMO pretreatment. Biotechnology and
- 425 Bioengineering. 2009;105:469-76.
- 426 [9] Cuissinat C, Navard P. Swelling and dissolution of cellulose part 1: free floating cotton and wood
- 427 fibers in N-methylmorpholine-N-oxide-water mixtures. Macromolecular Symposia2006. p. 1-18.
- 428 [10] Adorjan I, Sjöberg J, Rosenau T, Hofinger A, Kosma P. Kinetic and chemical studies on the
- 429 isomerization of monosaccharides in N-methylmorpholine-N-oxide (NMMO) under Lyocell
- 430 conditions. Carbohydrate Research. 2004;339:1899-906.
- [11] Fink HP, Weigel P, Purz HJ, Ganster J. Structure formation of regenerated cellulose materials
 from NMMO-solutions. Progress in Polymer Science. 2001;26:1473-524.
- 433 [12] Hall ME, Horrocks AR, Seddon H. The flammability of Lyocell. Polymer Degradation and Stability.
 434 1999;64:505-10.
- 435 [13] Karimi K, Zamani A. *Mucor indicus*: Biology and industrial applications perspectives: a review.
- 436 Biotechnology Advances. 2013;31:466-481.
- 437 [14] Murphy J, Power N. Technical and economic analysis of biogas production in Ireland utilising
- 438 three different crop rotations. Applied Energy. 2009;86:25-36.
- 439 [15] Thuresson T. Bioenergi från skog uppdaterad bedömning av potentialer och förutsättningar för
 440 svenskt skogsbruk att producera främst primära skogsbränslen. Skogsindustrierna; 2010.
- 441 [16] Kabir M, Sárvári Horváth I. NMMO pretreatment of straw and forest residues for enhanced
- 442 methane production. Manuscript.
- 443 [17] Kabir MM, del Pilar Castillo M, Taherzadeh MJ, Horváth IS. Effect of the N-Methylmorpholine-N-
- 444 Oxide (NMMO) Pretreatment on Anaerobic Digestion of Forest Residues. BioResources. 2013;8:5409-445 23.
- 446 [18] Båth B, Winter C. Bibliografiska uppgifter för växtnarinsstyrning i ekologisk odling i växthus.
 447 VäxtEko, Jordbruksverket; 2008.
- 448 [19] Yadvika, Santosh, Sreekrishnan TR, Kohli S, Rana V. Enhancement of biogas production from
- solid substrates using different techniques—a review. Bioresource Technology. 2004;95:1-10.
- 450 [20] Carlsson M, Uldal M. Substrathandbok för biogasproduktion. SGC, Swedish Gas Technology451 Centre; 2009.
- 452 [21] de Hullu J, Maassen J, van Meel P, Shazad S, Vaessen J, Bini L, et al. Comparing different biogas
- upgrading techniques. Eindhoven, the Netherlands: Dirkse Milieutechniek, Eindhoven University of
 Technology; 2008.
- 455 [22] Bauer F, Hulteberg C, Persson T, Tamm D. Biogas upgrading Review of commercial
- 456 technologies. SGC, Swedish Gas Technology Center; 2013.
- 457 [23] Henriksson G, Del Pilar Castillo M, Jakubowicz I, Enocksson H, Ascue J, Lundgren P, et al.
- 458 Environmental effects of the use of polymers in the biogas industry Pre study. Wasterefinery; 2010.

- 459 [24] Larsen J, Ostergaard Petersen M, Thirup L, Wen Li H, Krogh Iversen F. The IBUS process -
- 460 lignocellulosic bioethanol close to a commercial reality. Chemical Engineering & Technology. 461 2008;31:765-72.
- 462 [25] Turton R, Bailie RC, Whiting WB, Shaeiwitz JA. Analysis, synthesis and design of chemical processes. Upper Saddle River, New Jersey: Pearson Education; 2009. 463
- 464 [26] Kumar D, Murthy GS. Impact of pretreatment and downstream processing technologies on
- 465 economics and energy in cellulosic ethanol production. Biotechnology for Biofuels. 2011;4:27.
- [27] Aden A, Ruth M, Ibsen K, Jechura J, Neeves K, Sheehan J, et al. Lignocellulosic biomass to ethanol 466
- 467 process design and economics utilizing co-current dilute acid prehydrolysis and enzymatic hydrolysis 468 for corn stover. NREL National Renewable Energy Laboratory; 2002.
- 469 [28] Sassner P, Galbe M, Zacchi G. Techno-economic evaluation of bioethanol production from three 470 different lignocellulosic materials. Biomass and Bioenergy. 2008;32:422-30.
- 471 [29] Forgács G, Niklasson C, Sárvári Horváth I, Taherzadeh MJ. Methane production from feather
- 472 waste pretreated with Ca(OH)₂: process devolopment and economical analysis. Waste and Biomass 473 Valorization. March 2013.
- 474 [30] Forgács G, Pourbafrani M, Niklasson C, Taherzadeh MJ, Hováth IS. Methane production from
- 475 citrus wastes: process development and cost estimation. Journal of Chemical Technology &
- 476 Biotechnology. 2012;87:250-5.
- 477 [31] Benjaminsson J, Linné M. Biogasanläggningar med 300 GWh årsproduktion - system, teknik och 478 ekonomi. SGC, Swedish Gas Center; 2007.
- 479 [32] Gauthier Reberg K. Klart med sänkt bolagsskatt. Riksdag & Departement; 2012.
- 480 [33] Dolan T, Cook MB, Angus AJ. Financial appraisal of wet mesophilic AD technology as a renewable
- 481 energy and waste management technology. Science of the Total Environment. 2011;409:2460-6.
- 482 [34] Inflation i Sverige 1831-2012, konsumentprisindex.
- 483 http://www.scb.se/Pages/TableAndChart33831.aspx Statistiska centralbyrån; 2013.
- 484 [35] Naturvårdsverket. Förbränningsanläggningar för energiproduktion inklusive rökgaskondensering. 485 Naturvårdsverket; 2005.
- 486 [36] Hagen M, Polman E, Jensen JK, Myken A, Jönsson O, Dahl A. Adding gas from biomass into the 487 gas grid. GASTECH NV, Danish Gas Technology Center, Swedish Gas Center; 2001.
- [37] Skinner I, Essen Hv, Smokers R, Hill N. EU Transport GHG: Routes to 2050? Towards the 488
- 489 decarbonisation of the EU's 2009.
- 490 [38] Renewable Energy - Targets by 2020, European Commission.
- 491 http://ec.europa.eu/energy/renewables/targets_en.htm 2011.
- 492 [39] Teghammar A, Del Pilar Castillo M, Ascue J, Niklasson C, Sárvári Horváth I. Improved anaerobic
- 493 digestion by the addition of paper tube residuals: pretreatment, stabilizing and synergetic effects.
- 494 Energy and Fuels. 2013;27:277-84.
- 495 [40] Wiberg S. Vatten- och avloppstaxa. Tibro kommun; 2012.
- 496 [41] Dahlberg A, Ekander I. Wood fuel and peat prices No 4 2012. Swedish Energy Agency and SCB,
- 497 Energy and transport statistics 2012.
- 498 [42] Kastberg S. Fuel data. Swedish University of Agricultural Sciences, Unit of Biomass Technology 499 and Chemistry: Länsstyrelsen i Västerbottens län; 1997.
- 500 [43] Schnürer A, Jarvis Å. Mikrobiologisk handbok för biogasanläggninar. Rapport U2009:03. Avfall
- 501 Sverige, Svenskt Gastekniskt Center; 2009.
- 502 [44] Lindén B. Samkompostering av rötslam, halm och djupströgödsel för framställning av ett
- 503 lätthanterligt gödsel- och jordförbättringsmedel. SLU, Swedish University of Agricultural Sciences; 2000.
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508 TABLE LEGENDS

- 509 Table 1. Prices for raw materials, products, and utilities.
- Table 2. Stream components based on data obtained by batch NMMO pretreatment
- 511 experiments and expressed as ton/batch.
- Table 3. Co-digestion scenarios with forest residues, OFMSW, and sewage sludge.
- Table 4. Sensitivity analysis for the digestion of forest residues only.

515 FIGURE LEGENDS

- Figure 1. Block flow diagram of the NMMO pretreatment and biogas production from forestresidues within a co-digestion with OFMSW.
- 518 Figure 2. Process flow diagram of the entire process.
- 519 Figure 3. FCI, Fixed capital investment per section, including equipment prices, installation,
- 520 instrumentation, electricity, piping, insulation, engineering and construction, contractor's fee,
- and contingency. Auxiliary facilities, yard improvements, and buildings are excluded.
- 522 Figure 4. Annual operating costs for the base case divided into cost items.
- 523 Figure 5. Annual operating cost per section. Cost of materials is excluded.
- 524 Figure 6. Sensitivity analysis of total investment and annual operating costs, as well as
- 525 methane production costs, as a function of plant capacity of digested forest residues per year.
- 526 Figure 7. Result of cash flow analysis. Internal rate of return before taxes (IRR) of 50%
- 527 increased or decreased water consumption during washing, and of 20% increased or decreased
- 528 price of methane and forest residues, compared to base case, after taxes.
- 529

Raw materials	€/kg	Reference		
Forest residues	0.057	Market price ¹		
NMMO	4.0	[6]		
OFMSW	-	-		
Fresh water	6.7*10 ⁻⁵	[6]		
Polyacrylamide	2,171	Market price ²		
Sewage sludge	-3.28	3		
Products				
Methane	1.895	Market price ⁴		
Carbon dioxide	0.003	[6]		
Lignin rich digestate	0.030	Market price ⁵		
Utilities				
Electricity	0.0346 €/kWh	[28]		
Steam	0.0084	SuperPro Designer®		
Chilled water	2.28*10 ⁻⁴	SuperPro Designer [®]		
Others				
Waste water treatment	9.79*10 ⁻⁴	[40]		
Labor wage	70.000 €/employee/year	[28]		

¹Based on prices from the fourth quarter of 2011 [41], and the energy content of 1 kg

533 branches and tops [42], ²www.alibaba.com, ³personal communication with Moshe Habagil,

534 *VIVAB, Vatten och miljö i Väst, 2013,* ⁴*methane price sold on the market (www.fordonsgas.se)*

535 *minus the cost for injection and distribution into the gas grid, together with the cost for tank*

536 stations [31] and ⁵www.bioenergiportalen.se [41].

538 Table 2.

Stream	1	2	3	4	5	6	7	8	9	10	11
component											
Cellulose	14.3			14.3				4.1		4.1	
Hemicellulose	7.6			7.6				2.1		2.1	
Lignin	21.0			21.0				21.0		21.0	
Ash	7.6		15.2	7.6				22.7		22.7	
Water	16.8	3.3	205.1	133.8	2166.4	1815.1	117.0	1046.9	820.8	81.1	
NMMO		3.3		666.0	662.7		666.0	3.3		3.3	
Carbohydrates			60.7					8.1		8.1	
Proteins			8.0					1.1		1.1	
Fats			17.2					2.3		2.3	
Polyacrylamide										0.9	
Methane											32.6
Carbon dioxide											67.6
Total	67.3	6.6	306.1	850.4	2829.1	1815.1	783.0	1111.6	820.8	146.8	100.2
(ton/batch)											

541 Table 3.

Co-digestion	C/N	Forest	Unit prod.	Unit	Total raw	IRR % ⁴
substrates	ratio	residues	cost (€/kg	revenue	material (tons	
			CH ₄)	(€/kg CH ₄)	DW /year)	
Forest residues +	29.5 ^{1,2}	33%	1.58	2.12	300,000	20.70
OFMSW						
Forest residues +	$20.5^{1,3}$	67%	2.78	2.75	300,000	3.52
Sewage sludge						
Forest residues	325 ¹	100%	9.35	3.12	300,000	-100

542 ${}^{1}C/N$ ratio for forest residues is set as the middle value of a range between 150-500 according

to[18], ²C/N ratio for OFMSW is set as the middle value of a range between 15-32 according

to [43] and ³ C/N ratio for sewage sludge is set as 5.98 according to [44], ⁴IRR is the internal

545 *rate of return*.

547 Table 4.

Methane price	NMMO	Unit production	Unit revenue	$IRR^1 \%$
	recirculation	cost (€/kg CH₄)	(€/kg CH ₄)	
+0%	99.5%	9.35	3.12	-100
+0%	99.99%	3.21	3.12	-100
+25%	99.99%	3.21	3.21	4.30
+50%	99.99%	3.21	3.69	11.0

548 ^{*I}IRR is the internal rate of return.*</sup>

549



552 Figure 1.



Figure 2.



Figure 3.



Figure 4.



Figure 5.







Figure 7.