1	Prospective life cycle assessment of large-scale
2	biochar production and use for negative emissions
3	in Stockholm
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12 ABSTRACT – Several cities in Sweden are aiming for climate neutrality within a few decades 13 and for negative emissions thereafter. Combined biochar, heat and power production is an 14 option to achieve carbon sequestration for cities relying on biomass-fuelled district heating, 15 while biochar use could mitigate environmental pollution and greenhouse gas emissions from 16 the agricultural sector. Using prospective life cycle assessment, the climate impact of the 17 pyrolysis of woodchips in Stockholm is compared with two reference scenarios based on 18 woodchip combustion. The pyrolysis of woodchips produces heat and power for the city of 19 Stockholm, and biochar whose potential use as a feed and manure additive on Swedish dairy 20 farms is explored. The climate change mitigation trade-off between bioenergy production and 21 biochar carbon sequestration in Stockholm's context is dominated by the fate of marginal 22 power. If decarbonisation of power is achieved, building a new pyrolysis plant becomes a better 23 climate option than conventional combustion. Effects of cascading biochar use in animal 24 husbandry are uncertain, but could provide 10-20% more mitigation than direct biochar soil 25 incorporation. These results help designing regional biochar systems that combine negative 26 carbon dioxide emissions with increased methane and nitrous oxide mitigation efforts, and can 27 also guide the development of minimum performance criteria for biochar products.

KEYWORDS – Biochar, life cycle assessment, climate impact, negative emission technology,
 energy system, district heating, agriculture, dairy farming.

30 ABSTRACT ART



32 Introduction

33 Meeting the climate targets set by the Paris Agreement requires deep, immediate cuts in 34 greenhouse gas (GHG) emissions and changes in resource management. Pathways that could meet these targets consider varying types and degrees of technological development and 35 consumption change^{1,2}. Among the technology-based solutions, one set of scenarios considers 36 the large-scale deployment of negative emissions technologies (NETs)^{3,4}. The necessity, 37 feasibility and share of NETs in the future technological mix are still being discussed, but recent 38 reviews highlight a lack of bottom-up⁵ and upscaling studies^{6,7}, which are needed for actual 39 40 development of the sector.

41 Biochar systems have been presented as one of the most readily available NETs, bringing 42 desirable co-benefits such as improved soil structure, nutrient management and water holding capacity, and reduced soil nitrous oxide emissions $^{8-12}$. Some have also praised biochar systems 43 for being overall more compatible with the planetary boundaries than other bio-based NETs¹³. 44 45 However, several effects of biochar remain uncertain and practical ways of large-scale 46 production and incorporation of biochar to soils have yet to be designed and assessed regionally. 47 In Sweden, woodchips are commonly used in combined heat and power (CHP) plants to 48 produce district heating and power. In efforts to further reduce their carbon footprint and 49 achieve negative emissions, cities and energy utilities are considering the pyrolysis of 50 woodchips for biochar, heat and power production, instead of burning woodchips in 51 conventional CHP plants, for their future installations. The biochar produced in the city could 52 then be used, in cascades, in the agricultural sector as an animal feed additive, manure 53 management additive and soil improver¹⁴. Biochar mixed with manure is also thought to be a more practical way to return carbon to soil than direct biochar application^{14,15}. Thereby, biochar 54 55 systems have the potential to reduce the GHG emissions from the agricultural sector, which are often considered partly inevitable due to the continuous need for human food¹⁶. 56

57 A key feature of biochar systems is their lower level of energy production compared with conventional bioenergy systems¹⁷. To estimate the potential climate effects of biochar in 58 59 relation to its production chain and in comparison with alternative technologies, life cycle 60 assessment (LCA) is an appropriate tool. Previous LCA studies of biochar systems have found 61 sequestration of carbon and fossil fuel substitution effects to be the main contributors to the climate mitigation potential^{18–21}. Few studies were set in an energy context where heat is the 62 main product, included the combustion of biomass as a reference²² or investigated specific 63 industrial applications²³. However, to our knowledge, no previous study has dealt with the 64 65 cascading effects of biochar in animal husbandry before land application with manure, or has analysed the effects of biochar production on a city's district heating network. 66

The aim of this study was thus to analyse the potential climate benefits of large-scale biochar production, connected to Stockholm's district heating system, and biochar use in dairy farming, an illustration of where biochar could serve as an animal feed additive, manure additive and soil improver. The objectives were (i) to identify the requirements for a new biochar system to outperform, from a climate perspective, alternative energy uses of biomass, and (ii) to explore the range of potential agricultural GHG effects when biochar is used in cascades in dairy farming.

74 Methods

75 Scope definition

Functional unit. An input-related functional unit²⁴ was defined as the use of 1 tonne (dry weight) of woodchips acquired on the global market. This choice is motivated by the multiplicity of outputs of biochar systems and enables comparison with previous studies^{18–20,22}. This functional unit places the emphasis on using biomass resources efficiently from a climate change mitigation perspective. *Scenario description.* The LCA was conducted at system level, which took into account the effects of biochar production on the city's energy system and of biochar use in the agricultural sector. The assessment was comparative and prospective²⁵: four large-scale pyrolysis plant configurations (1a-d), producing different amounts of heat, power and biochar from woodchips, a technology under development, were compared with combustion of woodchips for heat and power production in Stockholm (2) or combustion of woodchips in a conventional power plant in Sweden (3) (Figure 1).

The production of heat, power and biochar was coupled to Stockholm's district heating network and the power grid. It included the supply of biomass, its conversion to biochar and use of the co-products for district heating and electricity generation. Pyrolysis oil was assumed to be directly combusted with the pyrolytic gases. The combustion scenarios (2) and (3) included the same supply chain of biomass and its use in modern plants. Apart from transport of bottom ash in these reference scenarios (2 and 3), the management of residues and capital equipment were excluded from the system boundary.



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96 Figure 1: Flow chart showing the three scenarios analysed: (1) Pyrolysis for combined heat and power (CHP)

97 and biochar in Stockholm. (2) Combustion for CHP in Stockholm (reference). (3) Combustion for power

98 production in Sweden (reference). Transportation steps are not shown, but were included in the assessment. 99 Biochar use ratios between feed and manure are given as percentage of dry mass. θ = thermal efficiency, η =

100 power efficiency, and β = biochar yield (% total dry mass). Energy conversion efficiency are given with respect to

101 the lower heating value (LHV) of the woodchips.

103 agricultural benefits through biochar use as animal feed, mixing with manure and land

¹⁰² In scenarios 1a-d, biochar use included transport to an intensive dairy farm, handling on-site,

application, avoided burdens in terms of mineral fertiliser and liming agent production, soil
organic carbon (SOC) stock changes and biochar decay. No increase in crop yield was assumed,
in accordance with Jeffery *et al.*²⁶. Likewise, increase in animal productivity was disregarded
as no data is yet available.

108 The foreground system included the biomass conversion step, energy generation and 109 agricultural effects. The background system included production and supply of biomass and all 110 transport steps. This distinction between background and foreground is particularly important 111 in prospective LCA, because background technologies available in the future, when the 112 technology under study will be deployed, are likely to differ from those available at present²⁵. 113 Geographical boundaries. The case is located in the region of Stockholm, Sweden. 114 Production of biochar through slow pyrolysis was assumed to take place in Lövsta, Stockholm. 115 This choice was motivated by availability of land for such a plant, feasibility of connection to 116 district heating and expected growth in demand for heat in the area. The biochar was assumed 117 to be used on dairy farms located within 300 km north, west and south of Stockholm. The supply 118 of woodchips was based on the existing supply chains used by the main energy utilities and was 119 assumed to comprise a mix of forest residues produced nationally and imported from the global 120 market, mostly from the Baltic region.

121 *Time boundaries.* A large-scale pyrolysis facility operating for at least 25 years and then 122 further retrofitted for continued operations was assumed. The time horizon selected for the 123 impact assessment was 100 years.

Impact categories. The issue primarily assessed was climate change impact, which was evaluated using the static characterisation factor global warming potential with a time horizon of 100 years (GWP₁₀₀). The emissions modelled were of the three main GHGs relevant in energy and agriculture systems: carbon dioxide (CO₂), nitrous oxide (N₂O; GWP₁₀₀ = 265) and methane (CH₄; GWP₁₀₀ = 30.5)²⁷. *Uncertainty*. In prospective LCA, uncertainties tend to be large²⁵ and difficult to estimate. Here, uncertainty regarding the large-scale pyrolysis plant was tackled with four configurations (1a-d) each modelling a different combination of power efficiency and biochar yield. For energy substitutions, two methods were compared. The uncertainty regarding the effects of biochar in the agricultural sector was investigated through three cases (worst, average and best cases) and via a Monte Carlo simulation. Moreover, some possible future developments of the background system were discussed qualitatively.

Intended audience. The intended audience includes researchers and stakeholders with an interest in future biochar production and use. The latter include Swedish municipalities, energy companies and farmer representatives.

139 Biomass production and supply

The woodchips were assumed to be produced from tops and branches harvested at final felling of conventional forestry activities in spruce and pine stands²⁸. These forest residues are a by-product of the timber industry: if not harvested for energy purposes, the forest residues would be left to decompose in the forest and would contribute to forest carbon stocks.

Extracting these forest residues, in Sweden, was recently estimated to have a climate impact of 48.6 kg carbon dioxide equivalents (CO₂-eq) per tonne of dry woodchips (harvesting, forwarding, and chipping) and to induce a loss in forest carbon of 89.1 kg CO₂-eq tonne⁻¹ of woodchips in comparison with no harvesting in a long term perspective²⁸.

To model transport emissions, the mix of woodchips was assumed to comprise three fractions representative of the Stockholm's current consumption, where around 15% of the total is acquired in a radius of 100 km from Stockholm, 50% is transported from more distant Swedish forest by rail and the remaining 35% is transported from the Baltic states by ship²⁹.

Emissions data for transport processes were taken from the Ecoinvent database³⁰ and are presented in Supporting Information (SI). The Swedish emission data were extended to the 154 fraction of imported biomass from the Baltic despite differences in climate, weather and 155 management conditions. This was justified by the fact that Stockholm aims to use only Forest 156 Stewardship Council (FSC)-certified woodchips³¹ and that generic emission data available in 157 the Ecoinvent database for woodchips were of the same order of magnitude.

158 **Biomass conversion**

159 Slow pyrolysis and combustion model. A mass and energy balance of the slow pyrolysis 160 process was constructed. The main output of the model was an estimate of the amount of district 161 heat that an optimised slow pyrolysis plant could generate in the context of Stockholm. The input data included a characterisation of the woodchips²⁹ and biochar properties meeting most 162 criteria of the voluntary European Biochar Certificate (EBC)^{32,33}. The pyrolysis temperature 163 164 was set to 700°C, as currently used in a pilot plant operated in Stockholm, ensuring biochar stability^{12,34,35} and agronomic properties. The yield of biochar was either 21% or 36%³⁶ (% total 165 166 dry mass) and the power to heat ratio was either 0 (no power) or 0.37. The enthalpy of pyrolysis 167 was assumed to be around 8% of the fuel's higher heating value, the mean value of the range reported in previous research³⁷. The woodchips were assumed to have an initial moisture 168 169 content of 50% and to be dried to 10% moisture content, using residual heat from the process, 170 before entering the pyrolysis reactor.

For combustion of biomass, data were taken from the latest CHP plant built in Stockholm^{29,38}
and from the IEA³⁹. The calculated and assumed efficiencies are compiled in SI.

Emissions from biomass oxidation. Pyrolysis and combustion of biomass are accompanied by emissions of air pollutants, some of which are climate forcers, resulting from incomplete combustion of the gases. Emissions of methane and nitrous oxide were assumed to be 0.011 and 0.006 g MJ⁻¹ woodchips, respectively⁴⁰. The same values were used for both pyrolysis and combustion, as nearly all the nitrogen contained in biomass is lost to the flue gas during pyrolysis and as both plants were assumed to be equipped with the same flue-gas cleaningtechnology.

Climate impact of energy substitution. One way of comparing scenarios with different outputs is the avoided burden approach or substitution approach^{18–20,22,24}. Selecting the appropriate substituted product is a key part of LCA studies⁴¹. Here, EF values for different fuels were taken from Swedish-specific data^{42,43}, while fuel displacements were identified with two approaches, one generic and one Stockholm-specific.

In the generic approach, natural gas was selected for substitution, as it is a fossil fuel likely to remain in Europe's energy system for many years. It was assumed that heat and power from the evaluated systems substitute use of natural gas for heat (90% LHV efficiency) and power production (53% LHV efficiency), with EFs of 77.0 and 131 kg CO₂-eq GJ⁻¹ respectively⁴².

189 In the Stockholm-specific approach, fuel displacements were calculated using a model 190 developed by Stockholm Exergi for long-term planning in the city's district heating network. 191 As described in SI, the model calculates the change in consumption of a dozen fuels and net 192 electricity production at city scale, following addition of a new plant in the system. For each 193 scenario, the output of the model was used to calculate the climate impact of energy 194 substitution. The EF for biomass fuel was set equal to that of the biomass used as functional unit, while for other fuels values were taken from⁴². The Stockholm-specific approach included 195 196 three time horizons (2020, 2030 and 2040) with respective EFs of 1000, 550 and 200 g CO₂-eq 197 kWh⁻¹ of electricity⁴³. These time horizons describes current projections for marginal electricity 198 used in Sweden, with old coal plants to be phased out in the coming decade and replaced by 199 efficient natural gas plants⁴³.

200 Biochar effects in dairy farms

201 The agricultural use of biochar was designed to be practical for farmers, to tackle various 202 sources of methane and nitrous oxide emissions in the agricultural sector and to be implementable at large scale in Sweden. Practicality was ensured by integrating biochar use with the current management of manure. In particular, no extra field operations were then needed to apply the biochar to soils as biochar is mixed with manure. The emissions affected included enteric fermentation, manure management and soil emissions. Annually, the pyrolysis plant was assumed to produce 52 500 to 90 000 dry tonnes of biochar, for use in Stockholm and neighbouring regions. These regions have an available area of about 900 000 ha of agricultural land.

210 Reference farm. A dairy farm of 300 cows was modelled, based on characteristics of Swedish dairy herd structures obtained from the Swedish Farmer's Association (LRF)⁴⁴ and the national 211 reporting to the UNFCCC⁴⁵. The dominant manure management system in Sweden, storage of 212 213 slurry under roof and annual application to soil^{46,47}, was modelled. Manure was assumed to be applied to land at a rate of 43 tonne ha⁻¹ of slurry (i.e. 25 tonnes ha⁻¹ of fresh manure), before 214 215 cultivation of high-yielding grass ley requiring fertilisation. The fertiliser value of manure 216 applied to land was calculated as in Hanserud et al.48. Additional mineral fertilisers were assumed to be ammonium nitrate, triple superphosphate and potassium chloride⁴⁹. It was also 217 218 assumed that the farm had access to enough land to accommodate the manure generated and 219 that slurry application rates complied with any local restrictions on phosphorus addition.

The annual emissions of GHG taken into account for one cow, the management of its manure and inputs for growing grass on manure-fertilised land were calculated following the guidelines from IPCC⁵⁰ and were equal to 8.1 tonnes CO₂-eq per year, excluding SOC changes. This amount was potentially affected by biochar.

Explorative modelling of biochar effects. Biochar was assumed to be delivered to dairy farms for addition to feed^{14,33,51}, at an average rate of 0.12 kg dry biochar cow⁻¹ day⁻¹ (about 1% of daily feed intake), and for mixing with manure, at an average rate of 30 kg dry biochar tonne⁻¹ of freshly excreted manure (3% mixing rate). Annual consumption of biochar for one cow and its manure was then 696 dry kg (6% as feed, 94% as manure additive), and for a 300-head farm it was 209 dry tonnes. The resulting application rate of biochar to land (0.80 tonne ha⁻¹ year⁻¹), was significantly lower than in most former studies, but application was assumed to take place every year. With these biochar use rates, about 75 000 dairy cows (22% of all Swedish dairy cows) and 66 000 ha of land (7.3% of available agricultural land in the region) would be needed to consume an annual biochar production of 52 500 tonnes (at 21% biochar yield).

234 The effects of biochar were represented by 11 emissions reduction factors, for four 235 compounds (methane, nitrous oxide, ammonia and nitrate) and six steps (enteric fermentation, 236 slurry storage, slurry application, fertiliser application, lime application, and land 237 methanotrophism). To explore the possible range of effects, the emission reduction factors were 238 given three set of values: worst, average and best case (Table 1). The worst case modelled 239 negative or null effects and a low biochar stability. The best case modelled optimised biochar 240 effects and high stability. The emission reduction factors were selected based on meta-analyses 241 when available, new experimental studies or expected effects otherwise. The agricultural effect 242 of biochar was calculated by multiplying the emissions reduction factors to the emissions from 243 the reference farm, and the total scaled by the biochar yield was taken as an avoided burden for 244 the biochar scenarios (1a-d).

245Table 1. Emission reduction factors (% of emission in reference farm, for each step and gas, available in SI)246and parameters used to explore the potential biochar soil effects and biochar stability.

Step	Gas	Worst, Average, Best	Source		
Enteric fermentation	CH ₄	0%, 2.5%, 5.0%	In-vitro experiments highlighted no increase in emissions, and probably a small decrease could be expected ⁵² .		
	CH ₄ -C	0%, 12.5% , 25%			
Indoor storage	NH ₃ -N	0%, 12.5% , 25%	Assumed effects on manure storage. No negative effects		
	d-N ₂ O-N	0%, 12.5% , 25%			
	NH ₃ -N	0%, 20% , 40%	Assumed reduced ammonia loss by biochar adsorption.		
Slurry application	NO ₃ -N	5.7%, 26% , 41%	Meta-analysis ⁵⁴ . Value for longer studies, with widest 95% confidence interval.		
	d-N ₂ O-N	-10%, 16% , 42%	Meta-analysis9. Value for small application rates.		
Mineral	NH ₃ -N	0%, 20% , 40%	Assumed reduced ammonia loss by biochar adsorption.		
fertiliser	NO ₃ -N	5.7%, 26% , 41%	Meta-analysis ⁵⁴ . Value for longer studies, with widest 95% confidence interval.		
application —	d-N ₂ O-N	-10%, 16% , 42%	Meta-analysis9. Value for small application rates.		

Soil methane sink CH		0%, -25% , -50%	In upland soils, the strength of the methane sink may slightly be reduced ^{55–57} . Increased emissions.		
Parameter	Unit	Worst, Average, Best	Source		
Biochar liming effect	% CaCO ₃	1.0%, 10% , 20%	Assumed liming effect based on liming classes from ⁵⁸ .		
SOC decay rate change	-	-0.80%, 3.8% , 8.1%	Meta-analysis ¹² . Grand mean and 95% confidence interval.		
Biochar carbon recalcitrance	%C	70%, 80% , 90%	Higher value supported by meta-analysis, for woody biomass ¹² . Lower value suggested as in ¹⁴ .		

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248 Agricultural effects of biochar not taken into account were: animal health improvement,

249 entailing lower mortality and medical inputs; and increased biomass yields⁵⁹.

250 Biochar long-term soil effects

251 The carbon sequestration effect of biochar derives from its resistance to biological, chemical 252 and physical degradation in soils. When using GWP₁₀₀ as the climate metric, the figure usually 253 reported is the amount of carbon remaining in the soil after 100 years. What happens after the 254 first 100 years was not considered here. The carbon stability of biochar was set to $80\% (\pm 10\%)$, in accordance with a pyrolysis temperature above 500°C and a molar $H: C_{org}$ ratio below 255 $0.40^{12,14,35}$. Assuming a biochar carbon content of 80%, the mean amount of carbon remaining 256 257 100 years after production was thus 64% of the dry amount of biochar initially applied to soil. 258 Application of biochar to soil is expected to affect SOC stocks. A decrease in the SOC decay 259 rate was modelled as a consequence of annual applications of biochar. The initial SOC level was assumed to be 85 tonne ha⁻¹ of land²². The new steady-state was assumed to be reached 260 261 during the timeframe of the study. In the average case, the change of decay rate (3.8%, metaanalysis¹²) led to an increase in SOC levels, reaching 88.4 tonne ha⁻¹. This SOC change was 262 263 scaled to the functional unit by dividing it by the total amount of biochar applied to the soil over 264 the timeframe of the study (100 years) and multiplying by the biochar yield.

The liming effect of biochar was modelled using the classes defined in the IBI standard⁵⁸.
The EF for substitution of conventional liming agent was taken from Ecoinvent³⁰. Biochar's

liming effect was benchmarked against Swedish agricultural recommendations that vary for
 different soil types⁶⁰ and assumed a reference application of liming agent every 10 years.

269 **Results and discussion**

The study compared the life cycle climate impact of three biomass options for expanding Stockholm's energy system. The ranking of these options varied mainly with assumptions on the background energy system and the magnitude of agricultural effects (Figure 2).



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Figure 2: Climate impact (kg CO₂-eq tonne⁻¹ dry woodchips) for the six systems studied under different substitution
methods (generic natural gas assumption and Stockholm-specific 2020, 2030 and 2040). The high and low bars
indicate the possible variation in biochar agricultural effects and carbon sequestration (worst and best case). In
the Stockholm-specific approach, the scenario 3 PP is modelled as exogenous, i.e. due to the size of the Nordic
electricity market, we assumed that this new plant would not affect the district heating network.

279 α = power to heat ratio, β = biochar yield (% total dry mass).

¹In the Stockholm-specific approach, electricity substitution is split between electricity used in heat pumps and net electricity produced in CHP.

²In the Stockholm-specific approach, heat substitution denotes the change in (non-electricity) fuel consumption
 for the CHP plants and the heat-only boilers.

284

285 Generic energy substitution

286 With the generic approach, where natural gas is the alternative fuel for both heat and power 287 production, a CHP plant outperforms a power-only plant, as expected, as do all biochar 288 scenarios (Figure 2). For the biochar scenarios, distinctions must be made between the levels 289 of agricultural effects achieved. In the worst case, where only biochar C sequestration is 290 achieved with negative agricultural effects, no pyrolysis scenario (1a-d) outperforms the CHP 291 reference (2). With natural gas as the alternative fuel and biochar C sequestration only, the 292 trade-off between bioenergy and biochar production is in favour for bioenergy: increasing the 293 biochar yield (1a to 1b, 1c to 1d) reduces the climate performance (Figure 2, generic). It is only 294 if the full range of agricultural effects is achieved that biochar scenarios can outperform the 295 CHP reference and that the trade-off is in favour of biochar production (Figure 2, generic).

These generic considerations are relevant for understanding the trade-offs of biochar systems^{17,61}. However, they do not reflect the current and future states of Stockholm's energy system where, for instance, heat production is mostly fossil-free, large heat pumps are in operation, and marginal power-consumption may still include some coal power plants.

300

Stockholm-specific substitution

301 In the long run (Figure 2, 2040), with a decarbonised marginal electricity (at 200 g CO₂-eq kWh⁻¹ electricity), all biochar configurations except 1c outperformed the reference plants even 302 with low biochar agricultural effects (worst case). In 2030 (at 550 g CO₂-eq kWh⁻¹ electricity), 303 304 only the plants with high electricity or biochar outputs (1a-b and 1d) ranked better than the 305 references under the condition that the high biochar agricultural effects were achieved (Figure 306 2, 2030). In today's context, with marginal electricity at 1000 g CO₂-eq kWh⁻¹ electricity, no 307 biochar configuration could outperform the reference scenarios except in one case (1b), when 308 high power and biochar yields with high agricultural effects are achieved (Figure 2, 2020).

The direction of the bioenergy-biochar trade-off changed with the time horizon and the magnitude of agricultural effects (Figure 2). The new base-load heat production capacity 311 provided by all scenarios did not contribute to any further substitution of fossil fuel for heat 312 production. Therefore, the performance of biochar systems in Stockholm is bound to the fate of 313 marginal power consumption, which is largely dependent on imports from the Nordic and 314 European Union grids. In the short term, pyrolysis plants with power production (1a, 1b) 315 performed better than without (1c, 1d) (Figure 2, 2020). However, this will no longer be the 316 case if low-carbon electricity production is achieved by other means in the future (Figure 2, 317 2040). Alternatively, biochar-based solutions would benefit from future scenarios where the 318 energy and electricity demands decrease. Indeed, low energy consumption reduces the pressure on biomass resources¹ and thus enables its use for other high-exergy applications, such as 319 320 biochar production.

In the next sections, we shed light on specific parts of the life cycle: production and transport,
system effects at the city level, and biochar use phase.

323 **Production and transport**

At present times, forest and plant technical operations, transportation steps and forest carbon losses accounted for 242 kg CO_2 -eq tonne⁻¹ in the combustion scenarios and 253-262 kg CO_2 eq tonne⁻¹ in the pyrolysis scenarios (Figure 2). A third of these emissions derived from losses in forest carbon stocks. The additional transport and handling of biochar only added 12-20 kg CO_2 -eq tonne⁻¹ woodchips. From a climate perspective, the production burden of the pyrolysis and combustion scenarios was rather similar.

In an optimistic prospective scenario, where efforts are made in all sectors of society to reach climate targets, the production burden would decrease. Other sources of woody biomass, such as willow grown on agricultural land, could even lead to increase in SOC stocks²². Besides, in the average and best cases, SOC increases following biochar application compensated for about 36% to 139% of the forest carbon loss arising from residue harvesting. The consequences of transferring carbon from forest to field for the long-term productivity of each land use were not assessed, but it is believed that higher SOC levels are an indicator of soil fertility with complex

337 feedback mechanisms⁶².

338 Fuel consumption changes at the city level

339 In terms of operation of Stockholm's district heating network, the addition of a new biomass-340 fuelled plant led mainly to lower consumption of biomass in older boilers and to lower 341 consumption of electricity for running heat pumps (HPs) (Table 2). An increase in electricity 342 production from CHP turbines was observed in scenarios 1a-b. To a lower extent, other fuels 343 (multi fatty acids, EO5 oil, biodiesel and tall oil) were also displaced but with a negligible effect 344 on the climate score of the different scenarios. Scenario 1b, due to its high power and biochar 345 production, had a higher merit order than one facility using imported municipal solid waste 346 (Table 2). This singularity did not affect significantly the climate score, and we therefore did 347 not model indirect effects related to alternative waste management abroad. Further analysis of 348 these city-level changes is provided in SI.

349 Table 2: Changes in fuel inputs and product outputs at the city level when introducing a new plant in the district
350 heating network. CHP = combined heat and power; HP = heat pumps.

_	Scenarios	1 a	1b	1c	1d	2 CHP
	Output change	tonne ⁻¹ dry woodchips ^a				
	Net electricity produced in CHPs (MWh)	0.59	0.40	-0.12	-0.29	1.6
	Biochar output from pyrolysis unit (kg)	210	360	210	360	0
	Input change	tonne ⁻¹ dry woodchips ^a				
	Woodchip-like biomass (MWh)	-1.7	-0.96	-1.6	-1.5	-2.0
	Net electricity consumed in HPs (MWh)	-0.27	-0.16	-0.55	-0.25	-0.57
	Imported municipal solid waste (MWh)	0	-0.21	0	0	0
	Other fuels ^b (MWh)	-0.17	-0.09	-0.22	-0.14	-0.37
	Net electricity produced in CHPs (MWh) Biochar output from pyrolysis unit (kg) <i>Input change</i> Woodchip-like biomass (MWh) Net electricity consumed in HPs (MWh) Imported municipal solid waste (MWh) Other fuels ^b (MWh)	0.59 210 -1.7 -0.27 0 -0.17	0.40 360 <i>tonne</i> -0.96 -0.16 -0.21 -0.09	-0.12 210 -1 dry wood -1.6 -0.55 0 -0.22	-0.29 360 <i>cchips^a</i> -1.5 -0.25 0 -0.14	1.6 0 -2.0 -0.57 0 -0.3

^aLHV of woodchips is 18.9 GJ tonne⁻¹ or 5.26 MWh tonne⁻¹. ^bMinor fuel changes for multi fatty acids, EO5 oil, biodiesel and tall oil.

The net biomass consumption change was +0.62 tonne per tonne of woodchips used in the new CHP plant (2), while it varied between +0.68 and +0.82 tonne per tonne of woodchips used in the pyrolysis plants (1a-d). A net change lower than 1 indicates that biomass use in newer, more efficient plants replaces biomass use in older plants. The fact that all values are strictly

³⁵¹

positive is linked to larger electricity or biochar outputs of the scenarios. At the city scale, building a large-scale pyrolysis plant (250 000 dry tonnes of woodchips per year) instead of a conventional CHP plant represented a net increase of 17 to 50 thousand tonnes of woodchips per year, while delivering the same heating service but different amounts of electricity and biochar. Apart from the direct climate impact, the consequences of this additional demand for biomass in Stockholm were not assessed here⁶³⁻⁶⁵.

362 **Biochar effects and variability**

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363 The average biochar use phase, including transport to and on the farm, yielded a climate impact of -3315 kg CO₂-eq tonne⁻¹ of biochar (i.e. -696 and -1193 kg CO₂-eq tonne⁻¹ of 364 365 woodchips at biochar yields of 21% (1a, 1c) and 36% (1b, 1d) respectively) (Figure 3a). More 366 than two thirds derived from sequestration of carbon in the biochar, while the remaining third 367 derived from agricultural emissions reductions and field SOC increase. The average agricultural 368 effect represented on average a 7.9% reduction in emissions from the dairy farm considered 369 and was evenly distributed between methane (11%) and nitrous oxide (11% direct, 2.9% 370 indirect) emissions reductions (Figure 3b). These non-CO₂ gases accounted for a non-negligible 371 25% of the total biochar effect (Figure 3b).



Figure 3: Per process (a) and per elementary flow (b) contribution to the climate impact of biochar agricultural effects and carbon sequestration (kg CO_2 -eq tonne⁻¹ dry biochar) for the three cases modelled (worst, average, best).

376 In the worst case modelled, the biochar effects were 48.4% lower than in the average case 377 and were dominated by biochar carbon sequestration and some increased emissions (loss of 378 SOC and increase in nitrous oxide emissions). In the best case modelled, the biochar effects 379 were 47.7% higher than average and biochar carbon sequestration contributed to only 54% of 380 the total effect. This modelling shows the importance of studying cascading biochar effects as 381 they could, if identified and optimised, double the benefits provided by biochar, thereby 382 mitigating up to 17% amount of non-CO₂ emissions from dairy systems; but also, if not 383 optimised, lead to an estimated increase of emissions of 2.2%.

To compare these results with previous biochar LCAs, one can express the ratio between other mitigation effects and carbon sequestration. For the effects studied here (Figure 3), the ratios were -0.14, 0.44 and 0.88 (worst, average and best cases). Hammond et al.¹⁸ had ratios of about 0.45, which was mostly due to assumed SOC increase. Other studies had ratios ranging from $0.071^{19,23}$ to 0.17^{19} depending on feedstock and effects modelled.

389 Energy system

Substitution approaches. In LCA, substitutions are a source of variability between studies because of choices made by modellers^{41,48,66}. Here, the Stockholm-specific approach showed how energy system modelling could help identify fuel displacements that are more representative of the actual changes in operation of the energy system than a generic approach. This model however introduces more parameter uncertainty that was not evaluated here.

With the generic approach, the energy substitution impact is simply a function of the energy efficiencies and the EF of the selected supply chain for the energy product considered. For the Stockholm-specific approach, the energy substitution is a function of the change in energy supply to the entire city and the EF for each energy source. This means that the Stockholmspecific approach does not only assess the climate impact of a biochar plant but the contribution of this new plant to the existing energy system. 401 Besides, the different time-horizons selected for marginal power consumption in Sweden 402 showed how the climate impact of different scenarios could vary significantly with changes in 403 assumptions regarding the background system⁶⁷.

404 System boundaries. Using different reference scenarios, substitutions methods and 405 timeframes also provides additional information for stakeholders. With the narrowest 406 boundaries, only the performance of the plant can be assessed, which is probably of interest to 407 plant operators for optimisation models or accounting (Figure 2, without energy substitution). 408 At the city scale, more information on fuel markets is obtained, which is of interest for 409 anticipating demand changes and securing supply (Figure 2, with specific energy substitution 410 and Table 2). These demand changes are also associated with indirect effects on land use, whose 411 impacts are difficult to assess and which should be avoided^{63,65,68}.

412 With wider boundaries, more alternative uses of biomass could have been considered. The 413 woodchips used in Stockholm are potentially available on the global market. Therefore, the 414 expansion of Stockholm's district heating network is in competition with other energy systems 415 that could theoretically acquire the same biomass for production of transportation fuel or to 416 replace coal-fuelled plants. In this second case, the additional benefits of a new biomass-fuelled 417 plant in Sweden, regardless of combustion or pyrolysis, would be lower than the displacement 418 of the most carbon-intensive energy sources. This suggests that the climate benefits provided 419 by biochar systems in Sweden are relevant globally only if the material, social and financial 420 means to phase out carbon-intensive energy sources are available to all countries. However, the reality of today's biomass⁶⁹ and carbon⁷⁰ markets may not incentivise such theoretical biomass 421 422 transfers. Besides, the introduction of new biochar technologies remains relevant for continued 423 improvements and competitiveness in Sweden, but also for the development of NETs, which is of international relevance in the longer term^{4,7}. 424

Biochar energy penalty. Compared with biomass combustion, biochar systems produce less heat and power because part of the energy in biomass remains in biochar¹⁷. The biochar energy penalty, defined as the difference in energy substitution between a biochar scenario (1a-d) and a reference scenario (2, 3), was dominated by the fate of power production.

The size of the biochar energy penalty is a critical indicator, as it sets a baseline for the performance of biochar during the use phase. It allows one of the key trade-offs between bioenergy and carbon sequestration⁶¹ to be quantified: unless the use phase effects are greater than the energy penalty, biochar solutions cannot outperform the established technologies from a climate perspective.

434 Agricultural system

435 Knowledge gap in biochar effects. While using biochar for feed and manure management is a convenient way of returning carbon to soil¹⁴, there is a knowledge gap regarding the effect of 436 biochar use in dairy farming systems⁷¹. The modelling of the agricultural effects of biochar 437 438 performed in this study was exploratory rather than predictive, as most values produced are 439 very uncertain and not backed up by local field observations or detailed mechanistic 440 understanding. Among the 11 reduction factors used in the model, 5 were documented with meta-analyses^{9,12,57,72}. The other parameters were selected based on new experimental results⁵² 441 or expectations for engineered biochar^{73,74} regarding leaching and volatilisation of nutrients and 442 effects on storage of manure^{53,75}. Likewise, the effects on manure management^{53,75}, but also 443 composting⁷⁶ and anaerobic digestion⁷⁷, are still not sufficiently well understood to be used for 444 445 anything other than prospective LCA studies. As commercialisation of biochar products for feed and manure management has already started in Europe⁷⁸, the need for rigorous academic 446 447 documentation of these effects is needed, including potential animal health and productivity 448 effects.

Nevertheless, the insights gained through prospective modelling are valuable for developers of biochar products^{73,74}, as it allows assessment of the relative sizes of expected effects. The potentially substantial reduction in agricultural emissions (10% of dairy farm emissions) is to be sought and optimised for different uses and environments. Here, only cascading use of biochar in animal husbandry was considered, but other uses in agriculture, construction or landscaping are possible⁷⁹.

Soil pH restrictions. The contribution of avoided production of liming agent to the climate impact was small, regardless of the biochar's liming class (Figure 3). However, the repeated application of biochar may lead to long-term changes in soil pH. Based on the Swedish liming recommendations for different soil types⁶⁰, restrictions on biochar application rates may be needed on soils that usually require little liming. That is the case of sandy and low-SOC soils⁶⁰ (SI). Restrictions on biochar application rates, whether for alkalinity or other factors, must be considered in future policy support and recommendations to farmers.

462 Policy support for environmental effects. Earlier research recommended to not establish 463 biochar solutions in regions where energy production is carbon intensive and where agricultural vields are already high⁸⁰. However, it was shown here that increase in agricultural yield is not 464 465 a requirement for climate-efficient biochar systems, if other emissions are reduced. 466 Nevertheless, agricultural yield increases are one of the main factors in acceptance of a new 467 practice by farmers, as it contributes directly to better economic performance^{81,82}. Therefore, if 468 environmental benefits are obtained without agricultural productivity increase, policy measures 469 providing payment for climate mitigation are needed to promote implementation.

470 Environmental assessment

Non-assessed climate factors. The present assessment was limited to the three main GHGs.
Effects of biochar application on albedo and heat flux were not included, as such an assessment
would require more site-specific data and as the effects would vary with soil management

474 practices. Anyhow, biochar on the soil surface has a warming effect, due to its dark colour. 475 Meyer and colleagues⁸³ estimated that the albedo contribution could reduce the mitigation 476 potential by up to 15%. Albedo changes can be reduced by permanent soil cover, which is 477 common on dairy farms due to grass cultivation, and incorporation of biochar into the soil. 478 However, soil disturbance may have other consequences for SOC levels. Thus, the albedo 479 penalty reinforces the need for biochar systems to perform significantly better than current 480 technologies.

481 Under the assumptions made on biochar stability in this study, about 123 kg CO₂ tonne⁻¹ of 482 woodchips would be released from biochar decomposition during the first 100 years. The 483 climate metric GWP₁₀₀ does not take into account the effects on climate of progressively 484 releasing the carbon dioxide contained in the biochar. Instead, what is implicitly assumed with 485 this metric is that the progressively decayed carbon over 100 years is released at the production time. Time-dependent metrics⁸⁴ could account for the consequences of slowly released carbon. 486 487 Other environmental impacts. Particle, dust and soot emissions due to biochar mishandling contribute to reducing the climate benefits of biochar-based systems⁸⁵. Their contribution to the 488 489 climate impact of this study may be marginal when clean production principles are applied, but particles also have serious effects in other environmental impact categories⁸⁶ (air pollution, 490 491 human health, biodiversity) that were not assessed here. Likewise, some co-benefits of biochar 492 were not quantified. For example, the reductions in ammonia and nitrate losses were modelled 493 because they lead to avoided indirect nitrous oxide emissions (Figure 3b)⁵⁰, but they also 494 mitigate water-related impacts such as eutrophication^{72,87}.

Final recommendation. At the large-scale envisioned in this study, where woodchips are sourced on the global market, the suitability of biochar systems in Stockholm is subject to the decarbonisation of the electricity market and other carbon-intensive sectors. If this decarbonisation is achieved by 2040, biochar solutions would represent a suitable expansion for the district heating network, thereby providing a sound option for carbon dioxide removal. If agricultural effects of biochar are optimised, through cascading use in animal husbandry, manure management and fertiliser management, the climate benefits of biochar could at best be doubled. Such a prospective development requires research efforts, in both upscaling of pyrolysis technologies and mechanistic understanding of biochar agricultural effects. When developing new biochar products, the life cycle perspective is useful to assess trade-offs and the relative importance of various potential effects.

506

507 ASSOCIATED CONTENT

508 Supporting Information. The Supporting Information (SI) is available free of charge on the 509 ACS Publications website at http://pubs.acs.org. It includes: activity and emissions data for 510 biomass production and supply; slow-pyrolysis model key parameters; description of energy 511 products in Stockholm's district heating network; description of city-scale district heating 512 network model; list of fuel emission factors; description of reference dairy farm model; activity 513 and emissions data for the reference dairy farm; description and diagram of biochar effect 514 modelling; table of emissions reduction factors, application rates and soil effects; description 515 of long-term effects of biochar on soil; detailed results on fuel displacement at city level; table 516 of results; histogram of biochar use phase effects; compiled list of parameters; references.

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520 Author Contributions

521 Elias S. Azzi defined the scope of the study, performed the modelling and wrote the 522 manuscript. Erik Karltun contributed to defining the scope of the study, model development

- 523 and manuscript revision. Cecilia Sundberg initiated the project, contributed to defining the
- 524 scope of the study, revised the model and revised and edited the manuscript.

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