Life-Cycle Assessment of Renewable Fuel Production via Hydrothermal Liquefaction of Manure in Germany

Supporting Information

Choice of manure samples

Manure presents an interesting but also diverse and complex feedstock for HTL. There is quite a number of different types of manure, each having individual characteristics. For our analysis, two characteristics are of special interest - the ash content and the biochemical composition. The former is important due to its influence on the biocrude and upgraded biocrude yield as well as the nutrient recovery step incorporated into the HTL process chain, while the latter strongly influences the composition of biocrude and upgraded biocrude. Table S1 lists various manures, shedding light on their differences in biochemical composition as well as ash content. Two manure compositions were chosen for the feedstock modelling in this work and appear highlighted in Table S1.

Table S1: List of proximate analyses of different types of manures, showing their diversity in biochemical composition, especially considering the amount of ash. The two manure samples used as orientation for the Aspen Plus® model are highlighted in bold face. All values are given in wt%.

Reference	biomass	Fats (F)	Proteins (P)	Carbos (C)	Lignin (L)	Ash (A)	Sum
[1]	beef manure	4.5	18.7	66.1	10.8	43.0	143.0
[1]	broiler manure	6.8	25.3	65.9	2.1	34.7	134.7
[2]	cow dung	11.6	18.9	24.2	14.6	5.8	75.1
[2]	cow dung	13.9	19.3	27.0	14.6	6.4	81.2
[3]	cattle manure	4.3	28.3	-	-	17.5	50.1
[3]	cattle manure	4.8	28.4	-	-	19.9	53.1
[3]	cattle manure	5.3	28.7	-	-	20.2	54.2
[3]	cattle manure	5.1	28.5	-	-	20.3	53.9
[3]	cattle manure	5.1	27.1	-	-	21.1	53.3
[4]	cattle dung	4.3	29.7	42.3	-	23.7	100.0
[5]	cow dung	-	16.9	-	-	24.6	41.5
[6]	cattle manure	-	-	-	-	25.3	25.3
[5]	cow dung	-	13.1	-	-	29.5	42.6
[5]	cow dung	-	8.9	-	-	30.1	39.0
[4]	cattle dung	2.0	24.5	41.3	-	32.2	100.0
[1]	**dairy cow manure**	**5.7**	**14.3**	**74.8**	**5.2**	**38.5**	**138.4**
[2]	poultry manure	9.6	20.9	28.0	6.1	6.6	71.2
[2]	poultry manure	10.4	23.5	32.0	6.1	7.1	79.1
[4]	poultry manure	5.3	32.4	37.8	-	24.5	100.0
[4]	poultry manure	2.4	27.0	37.7	-	32.9	100.0
[1]	laying hen manure	6.1	23.5	68.0	2.4	39.0	139.0
[1]	sheep manure	3.8	21.5	59.0	15.7	28.9	128.8
[2]	swine manure	9.2	18.5	26.2	7.9	5.9	67.7
[2]	swine manure	10.2	19.6	29.0	7.9	6.1	72.8
[7]	swine manure	18.8	26.9	37.6	5.3	11.4	100.0
[8]	swine manure	3.4	16.1	80.0	0.0	11.5	110.9
[1]	**swine manure**	**10.6**	**26.4**	**57.6**	**5.4**	**17.1**	**117.1**
[9]	swine manure	9.4	23.4	40.9	4.8	21.6	100.1

Aspen Plus® model

The Aspen Plus® model described herein is used as basis for the subsequent system analysis in terms of a life cycle assessment (LCA) and has been published before.[10]

Inputs

Table S2 lists all components used to represent the feedstock in the Aspen Plus® models. Distinct differences can be observed for the overall ash content as well as the amount of lipids and proteins. The amounts of biochemical species are taken from Ref. [1], whereby manure 1 is modelled based on cattle (dairy cow) manure and manure 2 is modelled based on swine manure.

Table S2: Mass of all model components [g] representing the feedstock in the Aspen Plus®models for manure 1 and manure 2, based on an input of 1000 g.

	manure 1 [g]	manure 2 [g]		manure 1 [g]	manure 2 [g]
Ash	**385.5**	**170.2**	**Carbohydrates**	**505.9**	**524.6**
Calciumoxide	90.6	40.0	Glucose	227.0	147.7
Potassiumoxide	145.6	64.3	Xylose	232.7	328.8
Magnesiumhydroxid	24.9	11.0	Acetic Acid	46.2	48.0
Sodiumoxide	25.2	11.1	**Proteins**	**90.6**	**231.7**
Ammoniumdihydrogen phosphate	87.0	38.4	3-Mercaptopropionic acid	13.9	13.9
Iron oxide	12.2	5.4	Glycine	4.1	16.7
Lipids	**30.2**	**90.0**	Glutamine	5.1	16.2
Myristic acid	2.3	4.6	Tryphthophan	7.1	22.7
Pentadecyclic acid	2.4	4.8	Phenylalanin	9.0	22.0
Palmitic acid	4.5	17.3	Lysine	8.0	32.5
Oleic acid	9.9	38.1	2-Ethylimidazol	5.3	21.3
Arachidic acid	4.7	9.4	Pyrrolidine	3.9	11.8
Behenic acid	3.4	6.8	Valeric acid	11.1	17.0
Glycerol	3.0	9.0	Tyrosin	9.9	30.1
Lignin	**38.3**	**49.4**	Formic acid	2.5	10.2
4-allyl-2-methoxyphenol	3.2	4.1	Ammonia	10.6	17.4
4-hydroxybenzaldehyde	3.2	4.1	**Extractives**	**8.0**	**8.0**
1,2-benzenediol	0.5	0.5	Phytan	1.0	1.0
Guaiacol	3.2	4.1	Farnesene	1.0	1.0
Phenol	3.2	4.1	Docosane	1.0	1.0
Orcin	3.2	4.1	Pentacosane	1.0	1.0
Benzoic acid	3.2	4.1	Nonacosane	1.0	1.0
1,2-diphenylethanol	3.2	4.1	A-pinene	0.5	0.5
4,4-Biphenol	3.1	4.1	B-pinene	0.5	0.5
Bisphenol A	3.2	4.1	A-terpinene	0.5	0.5
2,3-dihydrobenzofuran	3.2	4.1	Limonene	0.5	0.5
1,4-diphenylbutan	3.2	4.1	Stigmasterol	1.0	1.0
Diphenylether	3.1	4.1	**Estimated water added through hydrolysis**	**58.5**	**73.9**
			Sum	**1000.0**	**1000.0**

Table S3 compares the ultimate and proximate analysis by [1] with the respective Aspen Plus®model results. Relative deviations for ultimate and proximate analysis are in a range from 0.3 to 18.4% and from 0.3 to 20.4%, respectively. In case of the ultimate analysis, nitrogen as the smallest absolute measured quantity shows the highest relative deviation. The same trend can be observed for the proximate analysis with lignin as the smallest quantity.

Table S3: Comparison of ultimate and proximate analysis of manure feedstock with the chosen literature example, including relative deviations. All values are given in wt%, except for deviations, which are given in %.

Manure	C	H	N	O
1	49.3	6.9	2.9	40.2
2	53.1	7.3	5.3	33.7
cattle	50.6	6.7	2.5	40.3
swine	49.7	6.8	4.7	38.8
Deviation	2.5	3.6	18.4	0.3
Deviation	6.9	6.7	13.7	13.2

Manure	Lip	Car	Pro	Lig	Ash
1	3.8	50.6	9.1	3.8	38.6
2	9.8	52.5	23.2	4.9	17.0
cattle	3.5	46.0	8.8	3.2	38.5
swine	8.8	47.8	21.9	4.5	17.1
Deviation	8.7	9.9	2.9	20.4	0.3
Deviation	11.5	9.8	5.9	10.7	0.4

Results

Table S4 lists all important mass in- and outputs considered in the LCA model. Significant differences between the two scenarios can be observed for the biocrude yield and the solids mass. Consequently, also the upgraded biocrude yields differ significantly. Due to the lower amount of ash and therefore higher amount of organics in manure 2, the (upgraded) biocrude yields as well as the cHTG biogas yield is increased. The amount of used hydrogen per kilogram manure in the upgrading step is higher for manure 2, however, the normalized value per kilogram of upgraded biocrude is lower for manure 2 (0.058 kg H_2/kg_{fuel}) compared to manure 1 (0.064 kg H_2/kg_{fuel}).

Table S4: Mass balances for HTL fuel production with both manure samples. Note that both manure inputs are normalized to 1.000 kg of dry matter (DM), including the assumption of additional water being present due to hydrolysis already having taken place. DM content of wet manure is assumed to be 10wt%. DM content of manure after pretreatment is assumed to be ~20wt%.

Pretreatment	**manure 1 [kg]**	**manure 2 [kg]**	**characteristic**
Manure (DM with hydrolysis water)	1.059	1.074	input
HTL			
Slurry	5.059	5.074	input
Biocrude	0.211	0.313	output
HTL gas phase	0.147	0.180	output
HTL AP	4.569	4.523	output
HTL solids	0.131	0.058	output
HT			
Hydrogen	0.136	0.136	input
Biocrude	0.211	0.313	input
Upgraded biocrude	0.170	0.259	output
HT WW	0.021	0.022	output
HT offgas	0.155	0.167	output
cHTG			
Retentate	1.848	1.775	input
Permeate (ww)	2.721	2.748	output

cHTG product gas	0.147	0.190	output
Methane	0.046	0.060	info
Brine	0.538	0.409	output
cHTG AP	1.163	1.176	output
HT Recycle			
HT off-gas	0.155	0.167	input
H_2 in off-gas	0.125	0.121	output
Biogas in HT off-gas	0.030	0.046	output
H_2 used	0.011	0.015	info
H_2 used/kg ubc	0.063	0.056	info
PSA I H_2 recycle	0.113	0.109	intermediate
PSA I H_2 recycle	0.011	0.011	output
H_2 in off-gas	0.001	0.001	info
nutrient recovery			
Brine	0.538	0.409	input
Solids	0.131	0.058	input
Water solids	1.183	0.525	input
$MgCl_2$	0.054	0.023	input
H_2SO_4	0.134	0.060	input
NaOH	0.060	0.027	input
Waste solids	0.092	0.041	output
Struvite	0.125	0.055	output
Wastewater	1.883	1.005	output

Table S5 lists the elemental analysis of the modelled biocrude and upgraded biocrude streams. All values are in a typical range of literature values that have been obtained for other feedstock. [11–13]

Table S5: Elemental analysis of modelled biocrudes and upgraded biocrudes. All values are given in wt%.

Modelled stream	C	H	N	O
Biocrude 1	77.4	9.2	2.1	11.3
Biocrude 2	78.8	9.9	3.2	8.1
Upgraded biocrude 1	86.3	13.3	0.4	0.1
Upgraded biocrude 2	85.9	13.6	0.4	0.1

Figure S1 shows the distillation curves of the modelled biocrudes and upgraded biocrudes based on manure 1 and manure 2 as feedstock. Distillation curves of biocrudes reveal significant differences, which can be explained by the different biochemical compositions of the different manures. The most prominent difference can be observed in the range between 300 °C and 450 °C, where the majority of relevant higher boiling (decarboxylated) fatty acids (C14-C22) and derivatives can be found. Since modifications to these molecules during HTL still keep the boiling points in the same range and the majority of the molecules almost exclusively end up in the biocrude, the effect of different amounts of lipids in the feedstock is very pronounced.[11–13] Distillation curves of upgraded biocrudes are more similar, especially in the ranges up to 150 °C and between 325°C and 600 °C. Upgraded biocrude from manure 2 shows a steeper rise between 300 °C and 325 °C, due to the higher amount of n-alkanes originating from lipids in the feedstock. In contrast, the fraction between 150 °C and 175 °C shows a steeper rise for the upgraded biocrude originating from manure 1. The origin of molecules in this region is generally more complicated to explain, due to the complexity of the HTL reaction network and the interaction between various components originating mostly from proteins and carbohydrates. These reactions are summarized as Maillard-reactions.[14–17]

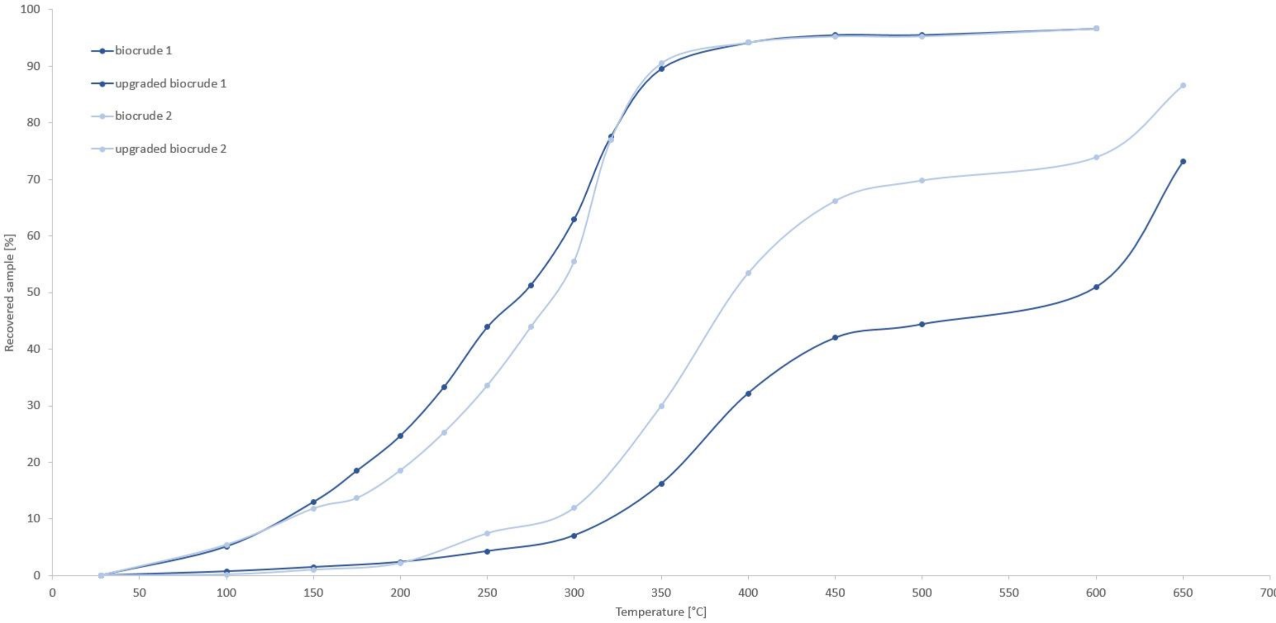

Figure S1: Distillation curves of modelled biocrude and upgraded biocrude for manure 1 and manure 2.

Alternative manure management

Based on [18], emissions for two different manure management scenarios (MIN (solid storage) and MAX (uncovered anaerobic lagoon)) have been calculated. In the following, each part of the derivation will be explained in detail. The calculation starts with regional and livestock specific excretion rates of manure (designated as volatile solids (VS)) and nitrogen, which have been adapted from [18] and are listed in Table S6. Based on the two excretion rates, the ratio of VS to N excretions is calculated. The results are also given in Table S6.

Table S6: VS and N excretion rates for dairy cattle, swine and poultry livestock, distinguished between Western and Eastern Europe.

	VS excretion rates [kg VS/animal/a]		**N excretion rates [kg N/animal/a]**		**VS / N ratio**	
	Western Europe	Eastern Europe	Western Europe	Eastern Europe	Western Europe	Eastern Europe
Dairy cattle	1642.5	1345.0	109.5	84.3	15.0	16.0
Swine	124.8	112.4	18.0	17.7	6.9	6.3
poultry	6.3	6.0	0.5	0.5	12.4	13.1

In a next step, CH_4 emission values based on [18] and expressed as [g CH_4/kg VS] were transformed into emission values expressed as [kg CO_2-Eq./kg VS]. All values are listed in Table S7, relevant values for the MIN and MAX scenario are shown bold.

Table S7: CH_4 emissions adapted from [IPCC] and converted in kg CO_2-Eq. / kg VS. Conversion factor for GWP100 is assumed to be 34. [19–21]

		CH_4 emissions [g CH_4 / kg VS]			**CH_4 emissions [kg CO_2-Eq. / kg VS]**		
High productivity		Cool, moist	Warm, moist	Warm, dry	Cool, moist	Warm, moist	Warm, dry
cattle	**uncovered anaerobic lagoon**	**96.5**	**117.4**	**122.2**	**2.4**	**2.9**	**3.1**
	liquid pit storage	33.8	59.5	65.9	0.8	1.5	1.6
	solid storage	**3.2**	**6.4**	**6.4**	**0.1**	**0.2**	**0.2**
	dry lot	1.6	2.4	2.4	0.0	0.1	0.1

	daily spread	0.2	0.8	0.8	0.0	0.0	0.0
	anaerobic digestion	3.2	3.7	3.7	0.1	0.1	0.1
swine	**uncovered anaerobic lagoon**	**180.9**	**220.1**	**229.1**	**4.5**	**5.5**	**5.7**
	liquid pit storage	63.3	111.6	123.6	1.6	2.8	3.1
	solid storage	**6**	**12.1**	**12.1**	**0.2**	**0.3**	**0.3**
	dry lot	3	4.5	4.5	0.1	0.1	0.1
	daily spread	0.3	1.5	1.5	0.0	0.0	0.0
	anaerobic digestion	6	6.8	6.8	0.2	0.2	0.2
poultry	**uncovered anaerobic lagoon**	**156.8**	**190.7**	**198.6**	**3.9**	**4.8**	**5.0**
	liquid pit storage	54.9	96.7	107.1	1.4	2.4	2.7
	solid storage	**5.2**	**10.5**	**10.5**	**0.1**	**0.3**	**0.3**
	dry lot	2.6	3.9	3.9	0.1	0.1	0.1
	daily spread	0	0	0	0	0	0
	anaerobic digestion	5.2	10.5	10.5	0.13	0.3	0.3

Besides the CH_4 emission values, also N_2O emission values based on [18], expressed as [kg N_2O/kg N], were taken and transformed into emission values expressed as [kg CO_2-Eq./kg N]. All values are listed in Table S8.

Table S8: N_2O emissions adapted from IPCC and converted in kg CO_2-Eq. / kg N. Conversion factor for GWP100 is assumed to be 298. [19–21]

		N_2O emissions [kg N_2O / kg N]			**N_2O emissions [kg CO_2-Eq. / kg N]**		
High productivity		Cool, moist	Warm, moist	Warm, dry	Cool, moist	Warm, moist	Warm, dry
cattle	**uncovered anaerobic lagoon**	**0**	**0**	**0**	**0**	**0**	**0**
	liquid pit storage	0.005	0.005	0.005	1.5	1.5	1.5
	solid storage	**0.01**	**0.01**	**0.01**	**3.0**	**3.0**	**3.0**
	dry lot	0.02	0.02	0.02	6.0	6.0	6.0
	daily spread	0	0	0	0.0	0.0	0.0
	anaerobic digestion	0.0006	0.0006	0.0006	0.2	0.2	0.2
swine	**uncovered anaerobic lagoon**	**0**	**0**	**0**	**0**	**0**	**0**
	liquid pit storage	0.005	0.005	0.005	1.5	1.5	1.5
	solid storage	**0.01**	**0.01**	**0.01**	**3.0**	**3.0**	**3.0**
	dry lot	0.02	0.02	0.02	6.0	6.0	6.0
	daily spread	0	0	0	0.0	0.0	0.0
	anaerobic digestion	0.0006	0.0006	0.0006	0.2	0.2	0.2
poultry	**uncovered anaerobic lagoon**	**0**	**0**	**0**	**0**	**0**	**0**
	liquid pit storage	0.005	0.005	0.005	1.5	1.5	1.5
	solid storage	**0.01**	**0.01**	**0.01**	**3.0**	**3.0**	**3.0**
	dry lot	0.02	0.02	0.02	6.0	6.0	6.0
	daily spread	0	0	0	0	0	0
	anaerobic digestion	0.0006	0.0006	0.0006	0.2	0.2	0.2

Taking into account the VS/N ratio from Table S6 and the emission values of CH_4 and N_2O from Table S7 and Table S8, the total emissions in kg CO_2-Eq. per kilogram of manure can be calculated. The resulting values are listed in Table S9, relevant values for the MIN and MAX scenario are shown bold.

Table S9: Total emissions calculated for cattle, swine and poultry manure, distinguished between Western Europe and Eastern Europe, different climate zones as well as storage method.

		Total emissions [kg CO_2-Eq. / kg manure]					
high productivity		Western Europe			Eastern Europe		
		Cool, moist	Warm, moist	Warm, dry	Cool, moist	Warm, moist	Warm, dry
cattle	**uncovered anaerobic lagoon**	**2.4**	**2.9**	**3.1**	**2.4**	**2.9**	**3.1**
	liquid pit storage	0.9	1.6	1.7	0.9	1.6	1.7
	solid storage	**0.3**	**0.4**	**0.4**	**0.3**	**0.3**	**0.3**
	dry lot	0.4	0.5	0.5	0.4	0.4	0.4
	daily spread	0.0	0.0	0.0	0.0	0.0	0.0
	anaerobic digestion	0.1	0.1	0.1	0.1	0.1	0.1
swine	**uncovered anaerobic lagoon**	**4.5**	**5.5**	**5.7**	**4.5**	**5.5**	**5.7**
	liquid pit storage	1.8	3.0	3.3	1.8	3.0	3.3
	solid storage	**0.6**	**0.7**	**0.7**	**0.6**	**0.8**	**0.8**
	dry lot	0.9	1.0	1.0	1.0	1.1	1.1
	daily spread	0.0	0.0	0.0	0.0	0.0	0.0
	anaerobic digestion	0.2	0.2	0.2	0.2	0.2	0.2
poultry	**uncovered anaerobic lagoon**	**3.9**	**4.8**	**5.0**	**3.9**	**4.8**	**5.0**
	liquid pit storage	1.5	2.5	2.8	1.5	2.5	2.8
	solid storage	**0.4**	**0.5**	**0.5**	**0.4**	**0.5**	**0.5**
	dry lot	0.5	0.6	0.6	0.5	0.6	0.6
	daily spread	0.0	0.0	0.0	0.0	0.0	0.0
	anaerobic digestion	0.1	0.3	0.3	0.1	0.3	0.3

Both N_2O, as well as CH_4 emissions reveal a strong dependence on the duration of storage, whereby the normalized total amount of N_2O is emitted in a significantly shorter timeframe than CH_4. Furthermore, it should be mentioned that based on [22], a threshold temperature of 13.93 °C for CH_4 emissions was introduced. For simplicity, the threshold temperature was adapted for N_2O emissions as well. The total amount of emissions in warmer temperatures (summer) was found to be 44 times (N_2O) and 15 times (CH_4) higher compared to colder temperatures (winter).[22–24]

Overall emissions from manure storage are calculated by taking the individual emission profiles from Figure S2 and adding up emissions for manure that is collected and stored each day. The resulting emissions are accumulating with storage duration as illustrated in

Figure S3 (top). Summation of all daily emissions results in the estimated actual value of 0.60 kg CO_2-Eq./kg manure. Due to the fact, that the estimated value is quite close to the MIN value, the ratio of N_2O to CH_4 emissions from the MIN scenario (solid storage) was used for the calculation. The graph in the bottom of

Figure S3 shows how the potential emission savings in Germany would decrease with increasing storage time. Taking into account the threshold temperature, two different scenarios can be distinguished (cold: blue, warm: orange).

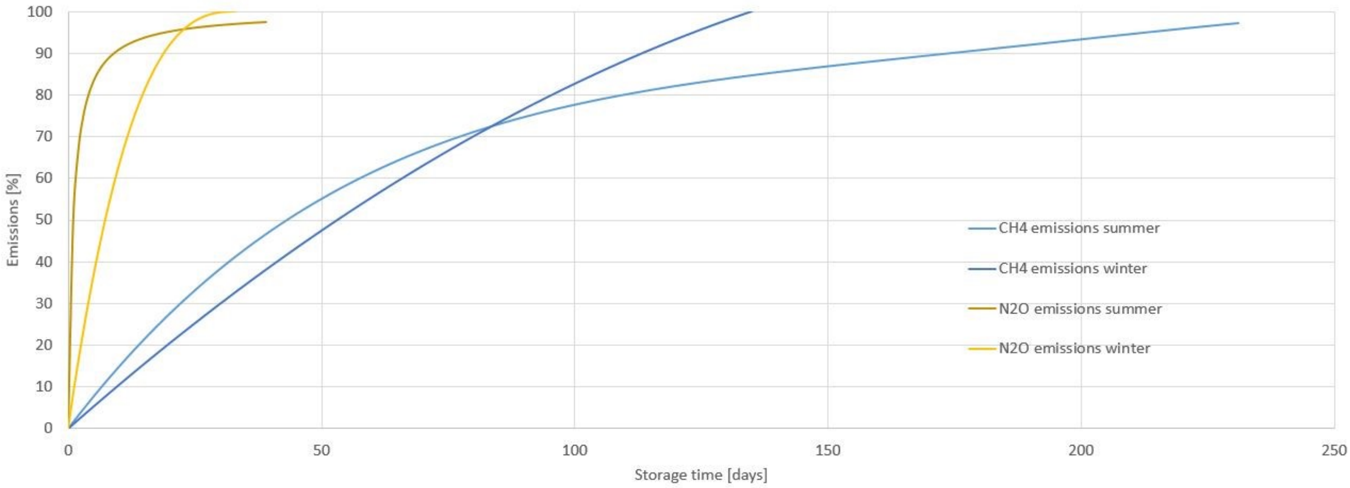

Figure S2: Temperature and time dependent emissions of CH_4 and N_2O based on [22–24].

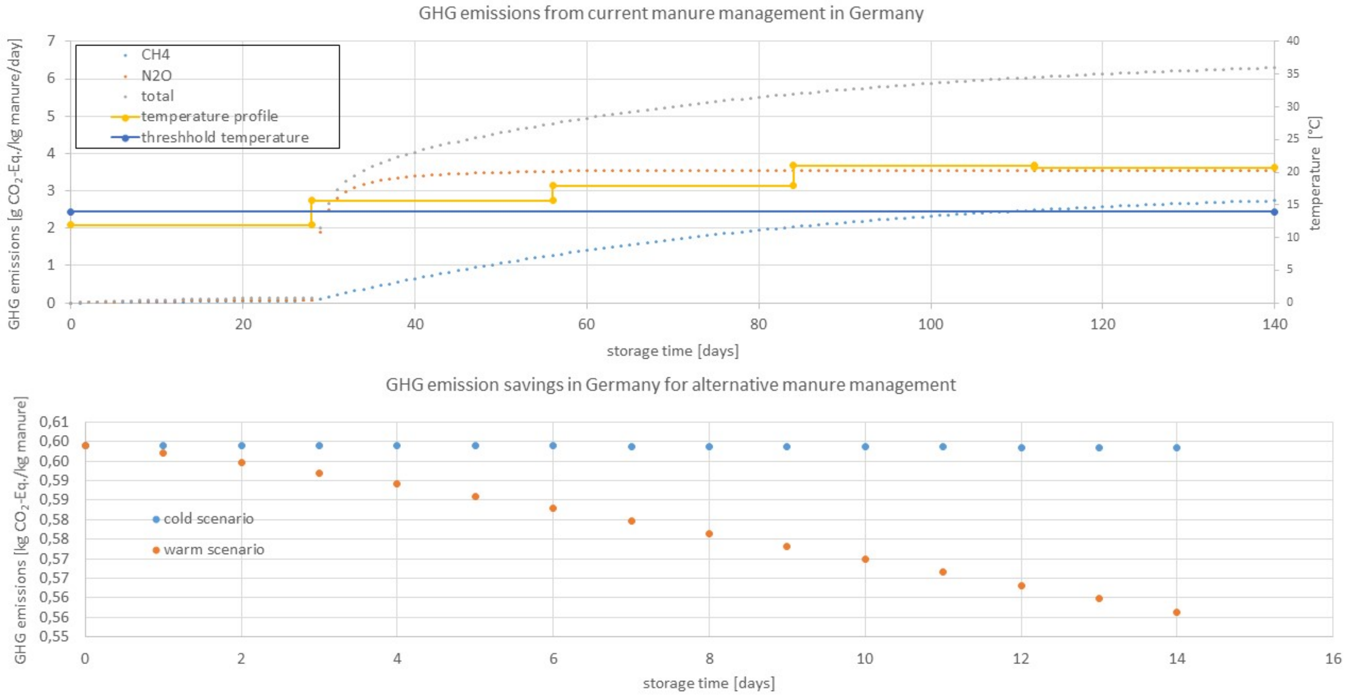

Figure S3: Top: Threshold temperature, monthly average temperature profile and emission profile for the estimated emission value from manure management in Germany. Bottom: Storage time dependent emission savings in Germany.

Results from the LCA model

Table S10: GWP100 results of the 30 investigated scenarios in the cut-off approach. CIs of the electricity inputs are 20, 80 and 410 g CO_2-Eq./kWh for the high wind share, medium renewables and GGM input, respectively.

Scenario	GWP100 [kg CO_2-Eq./$kg_{fuel\ mix}$]		
Electricity input	High wind share	Medium renewables	GGM
AEL-EL-M1	0.22	0.64	2.94
AEL-BG-M1	0.40	0.69	2.25
SMR-EL-M1	0.36	0.56	1.65
SMR-BG-M1	0.56	0.61	0.87

BL-M1	0.87	0.92	1.18
AEL-EL-M2	0.05	0.38	2.19
AEL-BG-M2	0.17	0.41	1.73
SMR-EL-M2	0.17	0.31	1.08
SMR-BG-M2	0.31	0.35	0.53
BL-M2	0.58	0.61	0.80

Table S11: Electricity demands of all process configurations investigated with the two different manure samples as well as electricity demands from literature for other fuel production pathways including PtL – El+RWGS, PtL – Co-El and PBtL.[25] A lower heating value of 43 MJ/kg_{fuel} has been used to convert the literature values.

Process configuration	Electricity demand [MJ_{el}/kg_{fuel}]
BL-M1	2.9
SMR-NG-M1	2.9
SMR-EL-M1	12.0
AEL-NG-M1	17.0
AEL-EL-M1	25.1
BL-M2	2.0
SMR-NG-M2	2.0
SMR-EL-M2	8.4
AEL-NG-M2	14.4
AEL-EL-M2	19.7
PtL – El + RWGS [25]	129.0
PtL – Co-El [25]	77.4
PBtL [25]	68.8

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