Electronic Supplementary Material (ESI) for Energy & Environmental Science.

Supporting Information
The carbon footprint of the carbon feedstock CO_2

Leonard Jan Müller, Arne Kätelhön, Stefan Bringezu, Sean McCoy, Sangwon Suh, Robert Edwards, Volker Sick, Simon Kaiser, Rosa Cuéllar-Franca, Aïcha El Khamlichi, Jay H. Lee, Niklas von der Assen, André Bardow

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SI 1. Literature review current practice “the carbon footprint of the carbon feedstock CO_2"

Author	Year	System boundaries for CO_2 source	Method to solve multifunctionality	Carbon footprint of captured CO_2	Reference
Aresta	1999	CO_2 source included; CO_2 is supplied from natural gas upgrading, ammonia plant, and fossil fuel-fired power plants; Energy demand and MEA loss is identical for all sources;	Allocation based on other relationship (mass and price as criterion) is applied	Not reported/ no calculation possible based on data; According to author: CFP of CO_2 > 0 kg CO_2e per kg CO_2	1
Aresta	2002	System boundaries are unclear; CO_2 is supplied from a fossil-fired thermal power plant;	No method to solve multifunctionality is reported	Not reported / no calculation possible based on data;	2
Clarens	2008	CO_2 source included; CO_2 is supplied from an ammonia synthesis plant; Entire ammonia plant is within system boundaries	Allocation based on other relationship (price as criterion) is applied	CFP of CO_2 > 0.06 kg CO_2e per kg CO_2	3
Falter	2015	CO_2 source included; CO_2 is supplied from direct air capture	No method to solve multifunctionality needed	Not reported/ no calculation possible based on data; According to the author: CFP of CO_2 <0 kg CO_2e per kg CO_2	4
Garcia-Herrero	2016	CO_2 source included; CO_2 is captured from a coal power plant with MEA	Substitution is applied	CFP of CO_2 = -0.86 kg CO_2e per kg CO_2	5

Giesen	2014	CO_2 source included; CO_2 is captured from a coal power plant fired with natural gas or biomass or from ambient air using a direct air capture process;	Multifunctionality does not occur since the entire electricity from the CO_2 source is used for conversion	Not reported/ no calculation possible based on data; CFP of CO_2: For fossil: > 0 kg CO_2e per kg CO_2 For biogenic and direct air capture: < 0 kg CO_2e per kg CO_2	6
Hoppe	2016	CO_2 source included; CO_2 is captured from a biogas purification; CO_2 is separated in biogas purification step and thus, CO_2 capture is not considered as part of the CO_2 based production life cycle	Substitution is applied	CFP of CO_2 = -0.97 kg CO_2e per kg CO_2	7
Al-Kalbani	2016	CO_2 source is excluded; Flue gas from coal power plant enters the system, is purified and serves as CO_2 source;	No multifunctionality problem since power plant is excluded from system boundaries	Not reported/ no calculation possible based on data; Consuming CO_2 lead to negative emissions according to authors; CFP of CO_2 < 0 kg CO_2e per kg CO_2	8
Kim	2000	CO_2 source included; CO_2 is captured from an ammonia plant	Allocation based on other relationship is applied (mass and energy as criteria) at different aggregation level, i.e. a sub-division is applied as far as possible followed by an allocation	CFP of CO_2 ≈ 0.03 kg CO_2e per kg CO_2 (obtained from Fig. 3)	9
Kim	2011	CO_2 source is excluded; An excess amount of pure CO_2 is assumed	No method to solve multifunctionality needed	Not reported/ no calculation possible based on data; Consuming CO_2 leads to negative emissions according to authors: CFP of CO_2 < 0 kg CO_2e per kg CO_2	10
Kongpanna	2014	CO_2 source is excluded; An excess amount of pure CO_2 is assumed	No method to solve multifunctionality needed	CFP of CO_2 = -0.03 kg CO_2e per kg CO_2	11

Luu	2015	CO_2 source is excluded; An excess amount of pure CO_2 is assumed	No method to solve multifunctionality needed;	CFP of CO_2 = -1 kg CO_2e per kg CO_2	12
Matzen	2016	CO_2 source included; CO_2 is supplied from biomass fermentation	Allocation based on other relationship (economic value as criterion) is applied	CFP of CO_2 = 0.08 kg CO_2e per kg CO_2	13
Overcash	2007	CO_2 sources included; CO_2 is supplied from: 1) ammonia plants 2) Hydrogen plants (SMR) 3) Natural deposits 4) Fossil fuel combustion	Allocation based on other relationship (mass as criterion) is applied	Not reported/ no calculation possible based on data; Only allocated life cycle inventories are reported;	14
Parra	2017	CO_2 sources included; 1) Direct air capture (included in system boundaries) 2) Biogas upgrading (supplies waste CO_2)	No method to solve multifunctionality needed for direct air capture; For biogas upgrading, CO_2 is considered as waste with no burdens associated	1) According Fig. 9: CFP of CO_2 > 0 kg CO_2e per kg CO_2 2) CFP of CO_2 = 0 kg CO_2e per kg CO_2	15
Pérez-Fortes	2016	CO_2 source is excluded; Concentrated CO_2, but unpressurized enters system boundaries;	No method to solve multifunctionality needed; Credit is given for avoided emissions + capture emissions	1) CFP of CO_2 = -1 kg CO_2e per kg CO_2 + emission related to compression	16

Reiter	2015	CO_2 sources included; CO_2 is supplied from fossil sources, e.g. fossil fired power plants; Source of CO_2 is excluded from system boundaries; Purification is either part of system boundaries or not	Three cases are distinguished: 1) CO_2 has biogenic origin or is a waste product and would be emitted nearly at reaction conditions ➔ CO_2 is considered to be neutral 2) CO_2 has biogenic origin or is a waste product, but has an additional energy demand for separation ➔ CO_2 carries the burden of separation 3) CO_2 has a fossil origin and would otherwise be stored, utilized in other processes or company would have to pay for emission allowances ➔ allocation is needed	1) CFP of CO_2 = 0 kg CO_2e per kg CO_2 2) CFP of CO_2 = 0 kg CO_2e per kg CO_2 + emissions of seperation/compression 3) CFP of CO_2 > 0 kg CO_2e per kg CO_2	17
Schäffner	2014	System boundaries are gate-to-gate Flue gas enters the system and a pure CO_2 stream is obtained ; CO_2 source is excluded from system boundaries	No method to solve multifunctionality needed; Credit is given for avoided emissions + capture emissions	Not reported/ no calculation possible based on data; Consuming CO_2 lead to negative emissions according to authors; CFP of CO_2 < 0 kg CO_2e per kg CO_2	18
Schakel	2016	CO_2 source included; Steam methane reforming as CO_2 source; CO_2 capture, CO_2 compression train is modeled	System expansion applied to include both hydrogen and DME	Not reported/ no calculation possible based on data;	19
Souza	2013	CO_2 source is excluded; An excess amount of pure CO_2 is assumed	No method to solve multifunctionality needed; Credit is given for avoided emissions	CFP of CO_2 = -1 kg CO_2e per kg CO_2	20
Supekar	2014	CO_2 source included; Various CO2 sources in the U.S. are considered	Allocation by distinguishing between determining and dependent products according to Weideman	Between 0.13 and 2.42 kg CO_2e per kg CO_2	21

Uusitalo	2017	CO_2 source is excluded; CO_2 purification and compression is in system boundaries;	No method to solve multifunctionality needed; However, credit is avoided emissions is given+ capture emissions	Not reported/ no calculation possible based on data; Consuming CO_2 lead to negative emissions according to authors; ➔ CFP of CO_2 < 0 kg CO_2e per kg CO_2	22
Van-Dal	2013	CO_2 source is excluded; CO_2 is captured from a coal power plant; CO_2 purification and compression is in system boundaries; CO_2 balance only covers methanol synthesis (gate-to-gate analysis)	No method to solve multifunctionality needed; However, credit is avoided emissions is given+ capture emissions	Not reported/ no calculation possible based on data; Consuming CO_2 lead to negative emissions according to authors; ➔ CFP of CO_2 > 0 kg CO_2e per kg CO_2	23
Walker	2017	CO_2 source is excluded; CO_2 is taken from biogas plant; CO_2 purification and compression is within system boundaries; CO_2 is used to boost methane production	No method to solve multifunctionality needed;	Not reported/ no calculation possible based on data;	24
Zhang	2017	Two different system boundaries are applied: 1) From cradle-to-gate including CO_2 source (fossil power plant + cement) 2) Gate-to-gate excluding CO_2 source	1) System expansion is applied (FU = km driven + electricity/cement) 2) Allocation using underlying physical relationship (mass as criterion) (FU = km driven)	Not reported/ no calculation possible based on data; CFP of CO_2 > 0 kg CO_2e per kg CO_2 (Fig. 5)	25
Von der Assen	2014	System boundaries are cradle-to-gate; CO_2 is taken from a coal power plant	1) System expansion is applied (FU = 1 kg Polyol + 0.36 kWh electricity) 2) Sensitivity analysis for solving multifunctionality (100% allocation to CCU system, substitution)	CFP of CO_2 = 0.2 kg CO_2e per kg CO_2 (worst case allocation) CFP of CO_2 = -0.8 kg CO_2e per kg CO_2 (substitution) CFP of CO_2 = -1 kg CO_2e per kg CO_2 (substitution with ideal CO_2 source)	

Sternberg	2015	CO_2 source included; CO_2 sources are 1) Ideal CO_2 source 2) Coal power plant 3) CO_2 is used that would otherwise be stored	Substitution is applied	1) CFP of CO_2 = -1 kg CO_2e per kg CO_2 2) CFP of CO_2 = -0.67 kg CO_2e per kg CO_2 3) CFP of CO_2 = 0 kg CO_2e per kg CO_2	26
Thonemann	2019	CO_2 source included; System boundaries are cradle-to-gate Various CO_2 sources are considered	Substitution is applied	1) CFP of CO_2 = -0.8 kg CO_2e per kg CO_2 (near term marginal mix) 1) CFP of CO_2 = -0.34 kg CO_2e per kg CO_2 (long term marginal mix)	27

SI 2. Detailed description of calculation in section 3.1

The carbon footprint (CFP) for CO_2 source with capture can be calculated from the carbon footprint without capture by subtracting the amount of CO_2 that is captured ($m_{captured\ CO2}$) and adding the indirect emission ($GW_{indirect,capture}$) for capture (Figure 1):

$$CFP_{CO2-source+CO2\ supply} = CFP_{CO2-source} - m_{captured\ CO2} + GW_{indirect,capture}$$

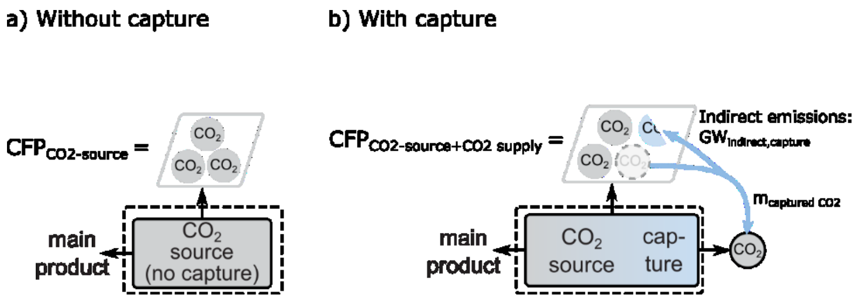


Figure 1: The carbon footprint of a generic CO_2 source with and without capture

In our examples, we only account for greenhouse gas emissions related to energy demand. In practice, however, other indirect emissions may also be relevant, e.g. emissions caused by the construction of the capture units. The indirect emissions for the supply of energy to drive the capture process are calculated from the energy demands per kg of CO_2 captured (Thermal energy q_{th}, electricity W_{el} and combustion of natural gas $q_{natural\ gas}$), the corresponding emission factors per MJ of energy(Thermal energy GW_{th} , electricity GW_{el} and combustion of natural gas $GW_{natural\ gas}$) and the amount of captured CO_2 ($m_{captured\ CO2}$)

$$GW_{indirect,capture} = \left(q_{th} \cdot GW_{th} + W_{el} \cdot GW_{el} + q_{natural\ gas} \cdot GW_{natural\ gas}\right) \cdot m_{captured\ CO2}$$

For our calculation, we use the emission factors provided in Table 1.

Table 1: Emission factors of required energy. The emission factor of natural gas includes the combustion[32]

GW_{th}	$0.057\ \frac{kg\ CO_2\ eq}{MJ_{th}}$
GW_{el}	$0.1325\ \frac{kg\ CO_2\ eq}{MJ_{el}}$
$GW_{natural\ gas}$	$0.0678\ \frac{kg\ CO_2\ eq}{MJ_{natural\ gas}}$

SI 2.1 Ammonia plant

According to the Ecoinvent dataset *"RoW: ammonia production, steam reforming, liquid ecoinvent 3.3"* 1.85 kg CO_2 eq per kg of ammonia ($= \mathrm{CFP_{ammonia}}$) are emitted. An average amount of 1.26 kg CO_2 per kg ammonia is formed and available for capture.[28–31] Consequently, to supply 1 kg as feedstock, 0.8 kg of ammonia is produced. The CO_2 capture is related to 0.401 MJ of electricity and 0.0754 MJ of heat per kg of CO_2 The carbon footprint per 1 kg of CO_2 as feedstock and 0.8 kg ammonia ($\mathrm{CFP_{ammonia+CO2\ supply}}$) is then the sum of the $\mathrm{CFP_{ammonia}}$ for 0.8 kg of ammonia and the indirect emissions caused by CO2 capture minus the amount CO_2 that is captured:

$$\mathrm{CFP_{ammonia+CO2\ supply}} = \mathrm{CFP_{ammonia}} - \mathrm{m_{captured\ CO2}} + \mathrm{GW_{indirect,capture}}$$

The carbon footprint 0.8 kg ammonia and 1 kg of CO_2 ($GW_{ammonia+CO2\ supply}$) are then calculated to:

$$\begin{aligned}\mathrm{CFP_{ammonia+CO2\ supply}} \\ &= 1.85\,\frac{\mathrm{kg_{CO2}\ eq}}{\mathrm{kg_{ammonia}}}\cdot 0.8\ \mathrm{kg_{ammonia}} - 1\ \mathrm{kg\ CO_2} \\ &+ \left(0.401\,\frac{\mathrm{MJ_{el}}}{\mathrm{kg\ CO2}}\cdot 0.1325\,\frac{\mathrm{kg\ CO_2e}}{\mathrm{MJ_{el}}} + 0.008\,\frac{\mathrm{MJ_{heat}}}{\mathrm{kg\ CO2}}\cdot 0.0754\ \frac{\mathrm{kg\ CO_2eq}}{\mathrm{MJ_{heat}}}\right)\cdot 1\mathrm{kg_{CO_2}}\end{aligned}$$

$$\mathrm{CFP_{ammonia+CO2\ supply}} = 0.525\ \frac{\mathrm{kg_{CO2}eq}}{0.8\ \mathrm{kg_{ammonia}} + 1\ \mathrm{kg_{CO2}}}$$

SI 2.2 Fermentation plant

According to Kaliyan, 56.4 g CO_2 eq per MJ of ethanol from cradle-to-gate corn is emitted.[33] The lower heating of ethanol is reported to be 21.5 MJ per dm^{-3}.[33] The density of ethanol is 0.789 kg per dm^{-3}. The CO_2 emission per kg of ethanol calculate as follows:

$$\mathrm{CFP_{ethanol}} = 56.4\,\frac{\mathrm{g\ CO_2eq}}{\mathrm{MJ_{ethanol}}} = 56.4\,\frac{\mathrm{g\ CO_2eq}}{\mathrm{MJ_{ethanol}}}\cdot\frac{21.5\,\frac{\mathrm{MJ_{ethanol}}}{\mathrm{dm^3_{ethanol}}}}{0.789\,\frac{\mathrm{kg_{ethanol}}}{\mathrm{dm^3_{ethanol}}}} = 1{,}54\,\frac{\mathrm{kg\ CO_2eq}}{\mathrm{kg_{ethanol}}}$$

In contrast Kaliyan et al., we include the carbon uptake of plants for the production of ethanol. For each molecule of ethanol, two molecules of carbon dioxide are absorbed from the atmosphere. Consequently, the amount of CO_2 that is absorbed per kg of ethanol calculated as follows:

$$\mathrm{m_{ethanol,CO2\ absorbed}} = \frac{\mathrm{m_{ethanol}}}{\mathrm{M_{ethanol}}}\cdot 2\ \mathrm{M_{CO2}} = \frac{1\ \mathrm{kg_{ethanol}}}{46\,\frac{\mathrm{g_{ethanol}}}{\mathrm{mol}}}\cdot 2\cdot 44\,\frac{\mathrm{g_{CO2}}}{\mathrm{mol}} = 1.913\,\frac{\mathrm{kg\ CO_{2\,absorbed}}}{\mathrm{kg_{ethanol}}}$$

The carbon footprint per kg of ethanol including carbon uptake calculates then to:

$$\mathrm{CFP_{ethanol,incl.\ biogenic}} = 1.54\,\frac{\mathrm{kg\ CO_2e}}{\mathrm{kg_{ethanol}}} - 1.913\,\frac{\mathrm{kg\ CO_2\ absorbed}}{\mathrm{kg_{ethanol}}} = -0.373\,\frac{\mathrm{kg\ CO_2e}}{\mathrm{kg_{ethanol}}}.$$

The ethanol plant produces approximately 0.96 kg of CO_2 per kg ethanol. The CO_2 capture is related to 0.432 MJ of electricity and 0.1325 MJ of heat per kg of CO_2. The total CFP for producing 1 kg of CO_2 and $\frac{\mathrm{1\ kg\ ethanol}}{\mathrm{0.96\ kg\ CO_2}} = 1.04\ \frac{\mathrm{kg\ ethanol}}{\mathrm{kg\ CO_2}}$ calculates to:

$$\begin{aligned} CFP_{\mathrm{ethanol+CO2supply\ ,incl.\ biogenic}} \\ &= -0.373\,\frac{\mathrm{kg\ CO_2e}}{\mathrm{kg_{ethanol}}} \cdot 1.04\ \frac{\mathrm{kg\ ethanol}}{\mathrm{kg\ CO_2}} - 1\ kg\ CO_2 \\ &+ \left(0.432\,\frac{\mathrm{MJ_{el}}}{kg\ CO2} \cdot 0.1325\,\frac{\mathrm{kg\ CO_2e}}{\mathrm{MJ_{el}}} + 0.057\ \mathrm{MJ_{heat}}\ \cdot 0.0754\,\frac{\mathrm{kg\ CO_2e}}{\mathrm{kg\ CO2}}\right) \cdot 1\ \mathrm{kg\ CO_2} \\ &- 1\ \mathrm{kg\ CO_2} \end{aligned}$$

$$e_{ethanol+CO2supply\ ,incl.\ biogenic} = -1.336\ \frac{kg\ CO_2e}{(1\ kg\ CO2 + 1{,}04\ kg\ ethanol)}.$$

SI 2.3 Direct air capture

The Direct air capture process only provides CO_2. The process consumes 1.013 MJ electricity and 4.038 MJ of natural gas per captured kg of CO_2. The carbon footprint for capture CO_2 $\mathrm{CFP_{DAC,CO2\ supply}}$ for direct air capture is then calculated:

$$\mathrm{CFP_{DAC,CO2\ supply}} = -\mathrm{m_{captured\ CO2}} + \mathrm{GW_{indirect,capture}}$$

$$\begin{aligned} \mathrm{CFP_{DAC,CO2\ supply}} &= -1\ \mathrm{kg\ CO_2} \\ &+ \left(1.013\,\frac{\mathrm{MJ_{el}}}{\mathrm{kg\ CO2}} \cdot 0.0678\,\frac{\mathrm{kg\ CO_2\ eq}}{\mathrm{MJ_{el}}} + 4.038\,\frac{\mathrm{MJ_{natural\ gas}}}{\mathrm{kg\ CO2}} \cdot 0.0678\,\frac{\mathrm{kg\ CO_2\ eq}}{\mathrm{MJ_{natural\ gas}}}\right) \cdot 1\ \mathrm{kg\ CO_2} \end{aligned}$$

$$\mathrm{CFP_{DAC,CO2\ supply}} = -0.592\ \frac{\mathrm{kg\ CO_2e}}{\mathrm{kg\ CO2}}$$

SI 3. CO_2 supply

SI 3.1 Supply of biogas in a low-carbon economy

Name	Capacity TWh (E_{total})	Potential CO_2 Supply	Comment	Reference
AEBIOM Technical potential	779	273.2	5% of arable land in EU, Maize monoculture, not environmentally acceptable	[34]
EEA - Environmentally acceptable potential	622	218.2		[35]
Projection 2020 AEBIOM	465	163.1		[34]
IEA - Energy Technology Perspectives 2017	393	137.8	Biogas generation in 2060 in Europe	[36]
Projection Agriculture 2020 AEBIOM	364	127.6		[34]
Biogas beyond 2020, CE Delft	360	126.1	Accelerated 2030 scenario	[37]
ICCT Projection	324	113.6	Selected has upper bound.	[38]
Projection 2020 EU	320	112.1		[39]
EEA - Resource efficient scenario	233	81.8	Biogas from manure and straw Selected has lower bound.	[40]
Biogasproduction in 2015 (Eurostat)	229	80.2	Biogas supply of today	[41]
Average	**409**	**143.4**		

The potential supply of CO_2 from biogas in a low-carbon economy is separated into the anaerobic digesters and the biogas power plant. After biogas formation in the anaerobic digester, we assume that all biogas is upgraded, i.e. the separation of methane and carbon dioxide. Subsequently, methane is fed to the natural gas grid and combusted in power plants. Therefore, the total CO_2 supply from biogas $m_{supply,biogas}$ is:

$$m_{CO_2\,supply,biogas} = m_{CO_2\,supply,upgrading} + m_{CO_2\,supply,combustion}$$

The amount of CO_2 from upgrading at the digesters is calculated as follows: According to AEBIOM, one cubic meter of biogas contains 0.6 m^3 biomethane and the remaining volume is filled with CO_2, the lower heating value of upgraded biomethane $LHV_{biomethane}$ is 46.8 MJ per kg and the densities of biomethane $\rho_{biomethane}$ and CO_2 ρ_{CO_2} is 0.73 kg respectively 1.98 kg per norm cubic meter. Therefore, the available mass CO_2 from biomethane upgrading $m_{CO_2\,supply,upgrading}$ can be calculated from the potential total energy as follow:

$$m_{CO_2\,supply,upgrading} = V_{CO_2\,supply,upgrading} \cdot \rho_{CO_2}$$

$$V_{CO_2\,supply,upgrading} = V_{biogas} \cdot (1 - 0.6)\frac{m^3_{CO_2}}{m^3_{biogas}}$$

$$V_{biogas} = \frac{V_{biomethane}}{0.6\,\frac{m^3_{Biomethane}}{m^3_{biogas}}}$$

$$V_{biomethane} = \frac{m_{biomethane}}{\rho_{biomethane}}$$

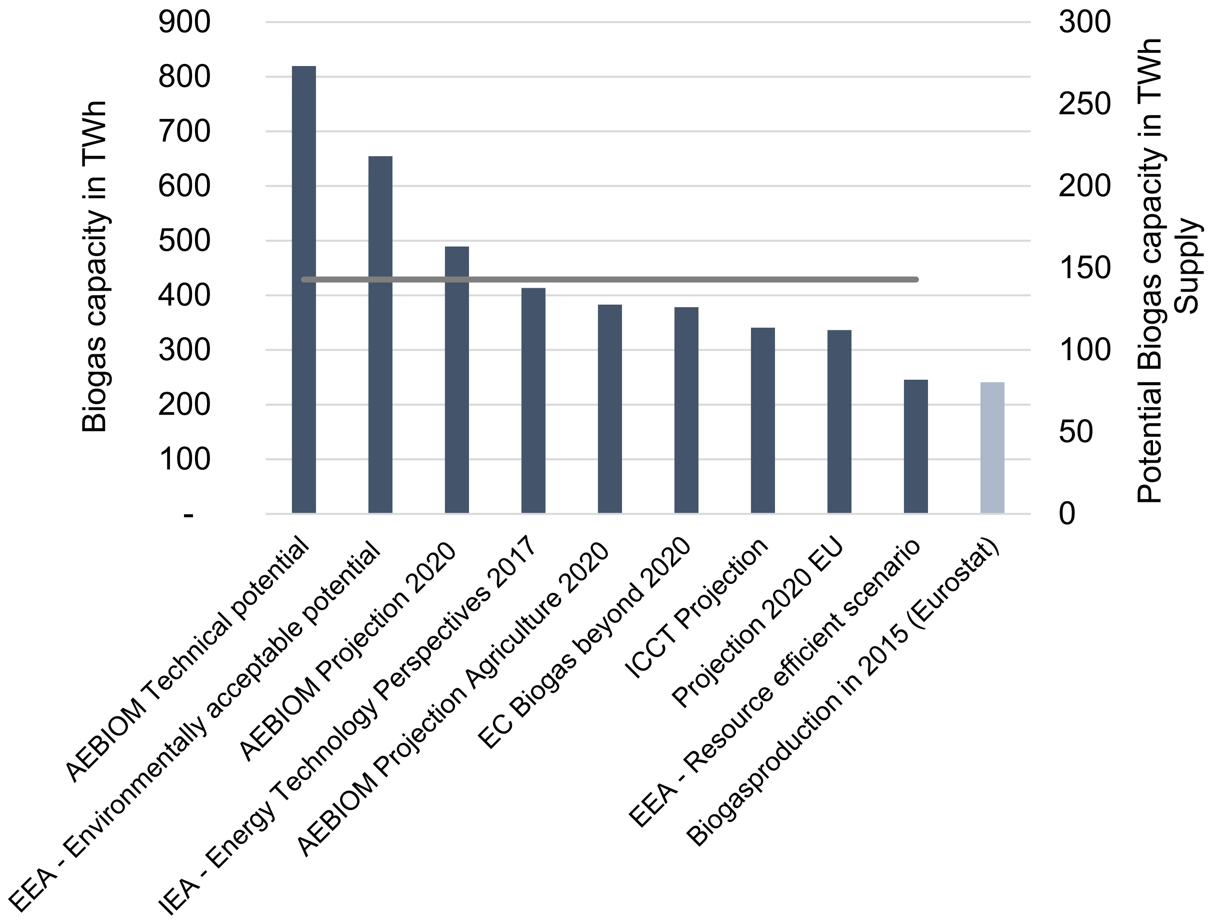
$$m_{biomethane} = \frac{E_{total}}{LHV_{biomethane}}$$

$$\Rightarrow m_{CO_2\,supply,upgrading} = \frac{E_{total}}{LHV_{biomethane}} \cdot \frac{(1 - 0.6)\frac{m^3_{CO_2}}{m^3_{biogas}}}{0.6\,\frac{m^3_{CH_4}}{m^3_{biogas}}} \cdot \frac{\rho_{CO_2}}{\rho_{biomethane}}$$

At the power plants, the available mass of CO_2 from the combustion of biomethane $m_{CO_2\,supply,combustion}$ assumes that 2.75 kg CO_2 is formed per kg of biomethane. Then, the potential CO_2 supply can be calculated from the potential total energy supply by:

$$m_{CO_2\,supply,combustion} = m_{biomethane} \cdot 2.75\,\frac{kg_{CO_2}}{kg_{biomethane}}$$

$$m_{CO_2\,supply,combustion} = \frac{E_{total}}{LHV_{biomethane}} \cdot 2.75 \frac{kg_{CO_2}}{kg_{biomethane}}$$



SI 3.2 Steel and iron mills

Table 2: Process energy demand for iron and steel making

Process	**Blast furnace oven**	**Blast oxygen furnace**	**Direct Reduction of Iron Ore with hydrogen from electrolysis**	**Advanced electric arc furnace**
Product	Pig Iron	Pig iron to crude steel	Sponge iron	Sponge iron to crude steel
Source	[42]	[42]	[43]	[43]
	[GJ per tonne]		[GJ per tonne]	[GJ per tonne]
Coke	10.30	7.26		
Coal dust	4.67	4.39		
Net power demand	0.20		10.13	1.26
Power demand N2 and O2	0.12	1.07		
Natural Gas	0.21	3.59	5.62	0
Coke oven gas	0.28			
Oxygen steel furnace gas	0.17			
Export of blast furnace gas	4.72	1.69		
Net Energy demand	11.24	14.62		
	[kg per tonne]		[kg per tonne]	[kg per tonne]
Use of hydrogen			58.17	

Table 3: Applied GHG emissions for energy sources and efficiency for converting water electrolysis and methanation (Taken from Otto et al. Table 1)[42]

$\mathbf{GW}_{el,fossil}$	160	kg CO_2 eq/GJ
$\mathbf{GW}_{el,renew}$	0	kg CO_2 eq/GJ
$\mathbf{GW}_{NG}$	56	kg CO_2 eq/GJ
$\mathbf{GW}_{Coke}$	105	kg CO_2 eq/GJ
$\mathbf{GW}_{Hardcoal}$	94.2	kg CO_2 eq/GJ
$\mathbf{GW}_{H2,SMR}$	66.64	kg CO_2 eq/GJ
$\mathbf{GW}_{CH4,synthetic,p2g}$	0	kg CO_2 eq/GJ
$\mathbf{GW}_{cokeovengas}$	40	kg CO_2 eq/GJ
$\mathbf{GW}_{BOFgas}$	257.8	kg CO_2 eq/GJ

Efficiency water electrolysis	0.7	kWh hydrogen per kWh electricity
Efficiency methanation	0.84	kWh methane per kWh hydrogen

Table 4: CO_2 supply in a low carbon economy (own calculation using production volumes of Steel Statistical Yearbook 2013)[44]

Total Capacity iron and steel making	169	Mt per year
Direct Reduction of Iron Ore with hydrogen from electrolysis	99	Mt per year
Electrical Arc oven	70	Mt per year
Total GHG Emissions	46	Mt CO_2 eq per year
Total GHG emissions no primary route	14.5	Mt CO_2 eq per year

SI 3.3 Cement plants

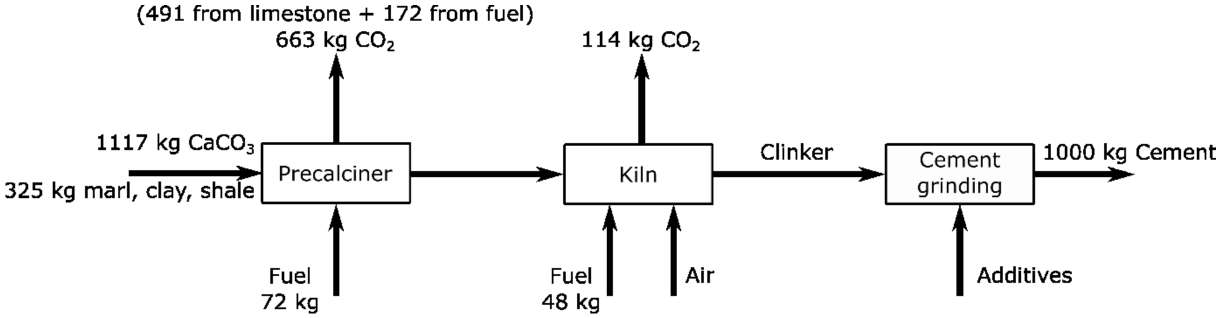


Fig. 1: Carbon flows as reported by Rodriguez et al. [46]

The emissions of the reference related to fuel combustion are $172\,kg\,CO_2 + 114\,kg\,CO_2 = 286\,kg\,CO_2$. Rodriguez et al. report a lower heating value of 25 MJ per kg of fuel.[46] The carbon content is 65 % since 172 kg CO_2 are emitted from 72 kg of fuel.[46,47] Methane has a lower heating value of 50 MJ per kg of methane and a carbon content of 75 %.

The CO_2 formation of using methane instead of the reported fuel calculated after the following equation:

$$e_{CO2,methane} = (72+48)\frac{kg_{fuel}}{t_{Cement}} \cdot \frac{25\frac{MJ}{kg_{fuel}}}{50\frac{MJ}{kg_{Methane}}} \cdot 0.75\frac{kg_C}{kg_{Methane}} \cdot \frac{44\frac{kg_{CO2}}{mol}}{12\frac{kg_C}{mol}}$$

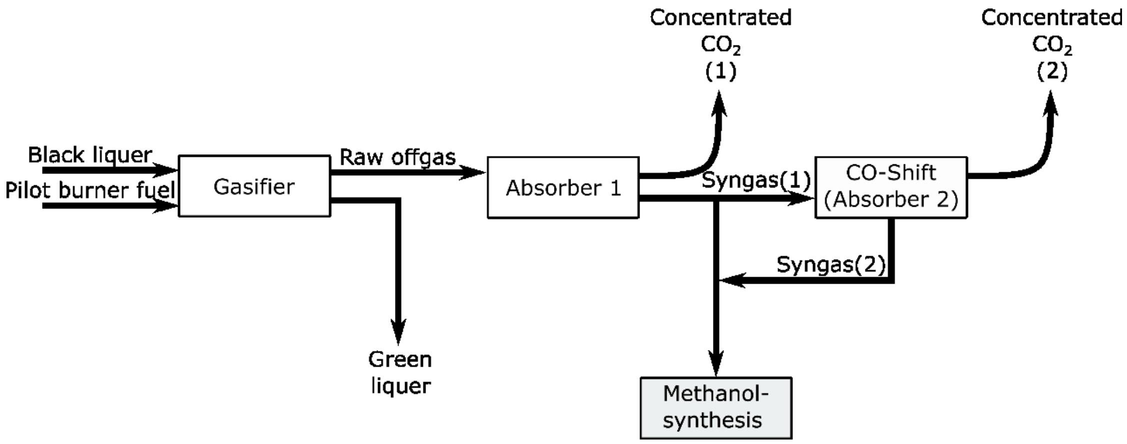
$$e_{CO2,methane} = 165\frac{kg_{CO2}}{t_{cement}}$$

Therefore, greenhouse gas emissions from fuel combustion are reduced by 16% by switching from the reported fuel to methane:

$$relative\ reduction = \frac{e_{total,methane}}{e_{total,fuel}} = \frac{(165+491)\cdot\frac{kg_{CO2e}}{t_{cement}}}{(286+491)\cdot\frac{kg_{CO2e}}{t_{cement}}} \approx 84\%.$$

SI 3.4 Pulp and paper mills

All data is based on Ekbom et al. [45]



Process unit	***Input***				***Output***		
		t/d	CO_2 [t/d]	CH_4 [t/d]		CO_2 [t/d]	t/d
Gasifier	Black Liquor	3,420			Green Liquor		
	Pilot burner fuel				Raw Gas	1,275	
Absorber 1	Raw Gas	3,164	1,275		CO_2 (1)	1,261	
					Syngas (1)	14	
Absorber 2	CO Shift Gas	396	1,025		CO_2 (2)	987	
					Syngas (2)	38	
Methanol synthese	Syngas	1,240	53	33	Methanol		1,183
					Combustible gases		

Greenhouse gas emissions (own Calculations)			
	4477	t CO_2 eq/d	Reference system
	2337	t CO_2 eq/d	With methanol production
	52 %	CO_2 for capturing in comparison to reference system	

SI 4. Total energy demand to capture CO_2 demand in a low-carbon economy

The total energy demand to supply the projected CO_2 demand for chemicals (255 Mt CO_2), fuels (415 Mt CO_2), and chemicals and fuels (670 Mt CO_2) is calculated from the projected supply of each CO_2 source and the corresponding energy demand (see Table 5 for results). In our calculation, we first select the CO_2 source with the lowest carbon footprint of feedstock CO_2 until the CO_2 supply capacity is fully exhausted. Then the next CO_2 source with the lowest carbon footprint is exhausted until the projected CO_2 demand is satisfied.

The total energy demand W_{total} is the sum over the energy that is consumed by the CO_2 source i:

$$W_{total} = \sum_i \left(\left(q_{th,i} + w_{el,i} + q_{natural\ gas,i} \right) \cdot m_{CO2\ supplied,i} \right)$$

In a low-carbon economy, we assume that thermal energy ($q_{th,i}$) is provided by electrode vessel and natural gas ($q_{natural\ gas,i}$) is substituted by synthetic natural gas. Consequently, both the thermal energy and the natural gas demand can be expressed through a demand for electricity. For thermal energy, we assume an energy efficiency of 0.99 ($\eta_{electrode\ vessel}$). Synthetic natural gas is produced from hydrogen, which is produced via electrolysis. We assume an efficiency for electrolysis of $\eta_{electrolysis} = 0.7$ and for the subsequent methanation step $\eta_{methanation} = 0.8$:

$$w_{el,th,i} = \frac{q_{th,i}}{\eta_{electrode\ vessel}} = \frac{q_{th,i}}{0.99}$$

$$w_{el,sng,i} = \frac{q_{natural\ gas,i}}{\eta_{electrolysis} \cdot \eta_{methanation}} = \frac{q_{natural\ gas,i}}{0{,}7 \cdot 0.8}$$

Consequently, the total energy demand is then calculated to

$$W_{total} = \sum_i \left(\left(w_{el,i} + \frac{q_{th,i}}{0.99} + \frac{q_{natural\ gas,i}}{0{,}7 \cdot 0.8} \right) \cdot m_{CO2\ supplied,i} \right).$$

Table 5: Total energy demand to supply sufficient CO_2 for chemicals (255 Mt CO_2), fuels (415 Mt CO_2), and chemicals and fuels (670 Mt CO_2)

	Chemicals			Fuels			Chemicals + Fuels		
CO_2 source	CO_2 supplied [Mt CO_2]	Energy demand [TWh]	Carbon footprint in [MT CO_2 eq]	CO_2 supplied [Mt CO_2]	Energy demand [TWh]	Carbon footprint in [MT CO_2 eq]	CO_2 supplied [Mt CO_2]	Energy demand [TWh]	Carbon footprint in [MT CO_2 eq]
Biogas upgrading	39	5	-39	39	5	-39	39	5	-39
Fermentation to Ethanol	7	1	-7	7	1	-7	7	1	-7
Biogas combustion	59	38	-59	59	38	-59	59	38	-59
Integrated pulp and paper mill	37	24	-30	30	24	-30	30	24	-30
Waste Incineration	41	13	-41	41	13	-41	41	13	-41
Steel and Iron	40	20	-40	40	20	-40	40	20	-40
Cement	32	54	-32	123	207	-122	123	207	-122
Direct Air Capture	0	0	0	69	158	-68	334	740	-320
Total		156	-254		467	-413		1050	-665
Total for direct air capture		583	-252		948	-410		1531	-662
Relative difference		**73 %**	**0.8 %**		**51 %**	**0.6 %**		**31 %**	**0.4 %**

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