Environmental impacts of a chemical looping combustion power plant

Rebecca J. Thorne[a,*], Evert A. Bouman[a], Kyrre Sundseth[a,b], Asuncion Aranda[c], Tomasz Czakiert[d], Jozef. M. Pacyna[a,e], Elisabeth. G. Pacyna[a], Mariusz Krauz[f], Agnieszka Celińska[g]

[a] NILU - Norwegian Institute for Air Research, Department of Environment Impacts and Sustainability, P.O. Box 100, Kjeller 2027 Norway
[b] SINTEF Industry, P.O. Box 4760 Torgarden, NO-7465 Trondheim, Norway
[c] IFE – Institute for Energy Technology, Department of Environmental Industrial Processes, Kjeller, Norway
[d] Częstochowa University of Technology, Institute of Advanced Energy Technologies, Częstochowa, Poland
[e] AGH - University of Science and Technology, Department of Energy and Fuels, 30-059 Krakow, Poland
[f] Institute of Power Engineering Ceramic Department (CEREL), Boguchwała, Poland
[g] National Centre for Nuclear Research, Nuclear Facilities Operations Department, 05-400 Otwock-Świerk, Poland

ABSTRACT

Chemical Looping Combustion (CLC) is a promising CO_2 capture option since it inherently separates CO_2 from other flue components, theoretically with low energy penalty. Here, a Life Cycle Assessment model was developed of a theoretical hybrid CLC (HCLC) power plant facility utilising experimental data for CuO based oxygen carrier (OC) production and oxygen capacity. Power plant models with and without post-combustion CO_2 capture, recognised as the most mature capture technology, acted as environmental performance targets. Results show that when OC is produced at lab-scale without optimisation, almost all (> 99.9%) lifecycle impacts per kWh electricity from an HCLC plant derive from the specific OC material used, giving a total of ˜700 kg CO_2eq/kWh. This is related to high electrical input required for OC processing, as well as high OC losses during production and from plant waste. Only when processing parameters are optimised and OC recycling from plant waste is implemented - reducing fresh OC needs – is the environmental impact lower than the conventional technologies studied (e.g. 0.2 kg CO_2 eq/kWh vs. ˜0.3-1 kg CO_2 eq/kWh, respectively). Further research should thus focus on identifying OCs that do not require energy intensive processing and can endure repeated cycles, allowing for recycling.

1. Introduction

As part of a European transition to a greener society, increasingly stringent emission targets are being set. This includes the EU 2020 climate and energy package, specifying a 20% reduction of greenhouse gas (GHG) emissions by 2020 with respect to the 1990 baseline (EC, 2018a) and the 2030 climate and energy framework extending the reduction to at least 40% by 2030 (EC, 2018b). Despite a large recent increase in the use of renewable energy, a significant proportion of electricity still derives from coal (IEA, 2017). The typical boiler combustion technology for coal energy generation is subcritical, although high-efficiency, low-emissions technologies - such as fluidised bed combustion - are increasingly implemented (World Energy Council, 2016). A subset of fluidised bed combustion, Circulating Fluidised Bed (CFB) is a combustion technology utilising relatively low temperatures that results in low NO_x emissions (IEA CCC, 2013). Despite these improvements, the only way to significantly lower CO_2 emissions and

meet GHG reduction targets whilst still utilising coal resources is by implementing carbon capture and storage (CCS).

CCS refers to a set of technologies that can reduce emissions of CO_2 with the trade-off that it requires additional energy and resources. Where implemented in power plants, this implies a lower power plant net efficiency since extra fuel needs to be combusted to produce the same net power output. Post-combustion CO_2 capture using amines, i.e. separating CO_2 from the flue gas with a solvent after combustion has taken place, is currently the most mature capture option with several large scale projects capturing > 1 Mt annually (Global CCS Institute, 2017). Since the CO_2 level in combustion flue gas is usually relatively low, the energy penalty and associated costs for the capture unit are elevated. This results in a typical loss in power plant efficiency of between 9% and 15% (Clarens et al., 2016; IEA, 2013; IPCC, 2013; Koornneef et al., 2008; Pehnt and Henkel, 2009; Spath and Mann, 2001; Zapp et al., 2012). CO_2 capture efficiency is generally around 90%.

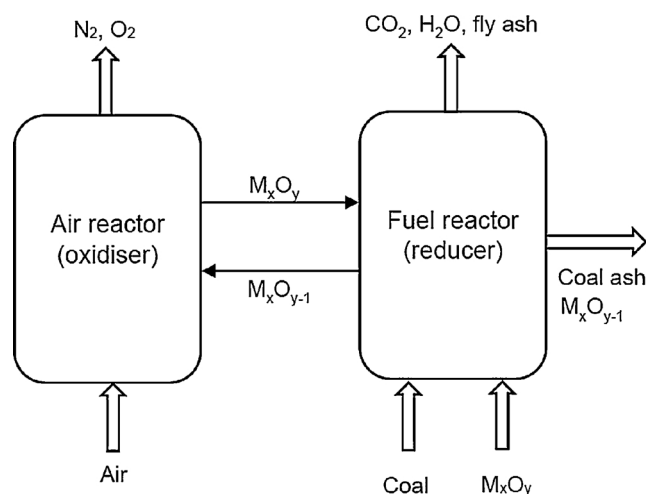Chemical Looping Combustion (CLC) is a promising pre-capture

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Fig. 1. Chemical Looping Combustion (CLC) schematic. M_xO_y and M_xO_{y-1} are the oxidised and reduced oxygen carrier (OC) states, respectively.

alternative to post-combustion CO_2 capture since it inherently separates CO_2 from other flue gas components *during* combustion (Linderholm et al., 2016; Lyngfelt, 2014). Consequentially, no additional energy or equipment is directly needed for gas separation itself. The technology typically employs a dual CFB system with two interconnected reducer and oxidiser reactors, where a solid stream of oxygen carrier (OC) mixed with a certain amount of ashes circulates between them (Fig. 1). The OC is normally a metal-bearing solid able to provide in-situ oxygen for fuel combustion, which re-oxidises at certain operation conditions in cyclic redox reactions. The reaction between OC and solid fuel may involve an intermediate gasification step (in-situ gasification Chemical Looping Combustion, or igCLC), or in the case of Chemical-Looping with Oxygen Uncoupling (CLOU), may directly lead to theoretically higher conversion rates (Lyngfelt and Leckner, 2015). Hybrid CLC (HCLC) aims to combine these processes into a complementary process to make the technology efficient and flexible to solid fuel types. In CLOU, oxygen is released from the OC due to favourable reactor conditions affecting the conversion of OC in its oxidised and reduced form. For example, the equilibrium oxygen concentration between Cu_2O and CuO is 2% at 1186 K (Lyngfelt, 2014). This means that, in the fuel reactor, where oxygen concentrations are low, CuO is reduced to Cu_2O thus releasing oxygen to react with the solid fuel particles. Alternatively, igCLC requires gasification of the solid fuel to syngas which subsequently reacts with the (solid) OC. HCLC aims to allow for both processes to occur simultaneously as a strategy for improvement of efficiency, as well as to allow for flexibility in solid fuel types. Associated energy penalty is around 4–5 %, with 1–2 % associated mostly with the energy for OC re-oxidation and the rest with CO_2 compression (Fan et al., 2012; Gauthier et al., 2016; Lyngfelt and Leckner, 2015). Effective CO_2 capture efficiencies are process dependent, varying from e.g. 88% (Fan et al., 2017) to almost 100% (Mukherjee et al., 2015).

In the fuel reactor (reducer), fuel and OC material react to produce mostly steam and CO_2, which may then be separated by condensation. The CO_2 is subsequently purified, compressed and transported for storage or utilisation. In the air reactor (oxidiser), depleted OC material regenerates (i.e., re-oxidises) through air contact at a suitable temperature.

The critical element of CLC technology is the OC material, which must have high oxygen capacity, fast reaction kinetics, high fuel combustion efficiency and at the same time have high mechanical stability over many redox cycles. As well as reacting with oxygen in the air reactor, OC for HCLC also needs to release oxygen through decomposition in the fuel reactor at high temperature. The cheap natural mineral ilmenite, $FeTiO_3$, has received much attention for use with a solid fuel in-situ gasification stage, but does not evolve oxygen at high temperatures

and cannot be used for Chemical-Looping with Oxygen Uncoupling processes (Azis et al., 2010; Lyngfelt and Leckner, 2015). Metal oxide systems such as CuO/Cu_2O, Mn_2O_3/Mn_3O_4 and Co_3O_4/CoO have suitable equilibrium oxygen partial pressures at temperatures of interest for combustion (Adanez-Rubio et al., 2011; Keller et al., 2014; Skulimowska et al., 2017). Copper oxide (CuO) is one of the most interesting options since it has a high ability to release oxygen at combustion temperatures, is not sensitive to sulphur, possesses fast decomposition kinetics (at the process temperature), and has few health and safety challenges associated with its use. Since sintering during operation and mechanical weakness is the most challenging issue of using unsupported OC, CuO may be supported on waste materials such as fly ash (Skulimowska et al., 2017). Limitations of CuO are related to the relatively low melting point of metallic Cu (1358 K) (Skulimowska et al., 2017), thus limiting high temperature operation, as well as the relative high costs associated with the material. In order to reduce the quantity of fresh material required, research has shown that recycling and reuse of OC is technically feasible (Garcia-Labiano et al., 2007). Using a CuO OC (supported on waste materials) and implementing OC recycling is therefore of high interest.

The environmental performance of power plants with CCS has been extensively assessed in the literature using Life Cycle Assessment (LCA). LCA is a tool used to assess environmental impacts of a technological system over the full lifecycle and by incorporating the full value chain. By assessing all parts of the CCS lifecycle, the environmental benefits of CO_2 reduction can then be weighed up against the environmental impacts related to the extra resources, emissions and processes required. Most LCA CCS studies conclude that the reduction of GHG emissions (measured by the 'Global Warming Potential' or GWP) is not as large as the CO_2 capture rate initially suggests, and that other impact indicators such as human toxicity potential are often increased regardless of capture technology, time horizon, or fuel considered (Corsten et al., 2013; Koornneef et al., 2012; Singh et al., 2012; Zapp et al., 2012). This is primarily due to an increase in upstream and downstream emissions, as a result of a decrease in net plant efficiency and additional resources required for CCS.

There are currently relatively few LCA studies regarding CLC technology. Petrescu et al. (2014) and Petrescu and Cormos (2017) produced some of the first LCA studies of CLC technology for both industrial and power plant applications, showing the CLC options to reduce environmental impacts across selected impact categories compared to conventional technologies (e.g. 0.34 kg CO_2-eq/kWh vs. 0.92 kg CO_2-eq/kWh, respectively for power plants). These studies utilised ilmenite as OC, and included standardised (regular) ilmenite production and concentration within the modelled system boundaries. The relationship between lifecycle GHG emissions and key CLC power plant parameters was recently investigated by Fan et al. (2018), including the type of OC, the lifetime of the OC, the GWP of the OC (based on the manufacture of its metal content), and the thermodynamic power plant performance. They found that the OC (and its associated GWP and lifetime) can have a major influence on the overall lifecycle GHG emissions of the electricity produced using CLC (which at designed conditions was calculated at 0.063 kg/kWh). As with Wang et al, (2018); Fan et al. (2018) approximated the lifecycle impacts of OC production by considering the production of its primary metal content. However, to the authors' knowledge, use of actual experimental data (such as material losses or energy input) relating to the additional processing necessary for OC specific use has not yet been implemented in an LCA. Although using experimental data for OC processing does not reflect the higher efficiencies of large-scale production, various approaches for the scale-up of processes from laboratory to industrial scale may be made (Caduff et al., 2011, 2014; Piccinno et al., 2016, 2018).

In this article, the lifecycle environmental impacts of HCLC electricity generation (using a CuO OC) are quantified, with special focus on the contribution of OC processing that has not been fully included
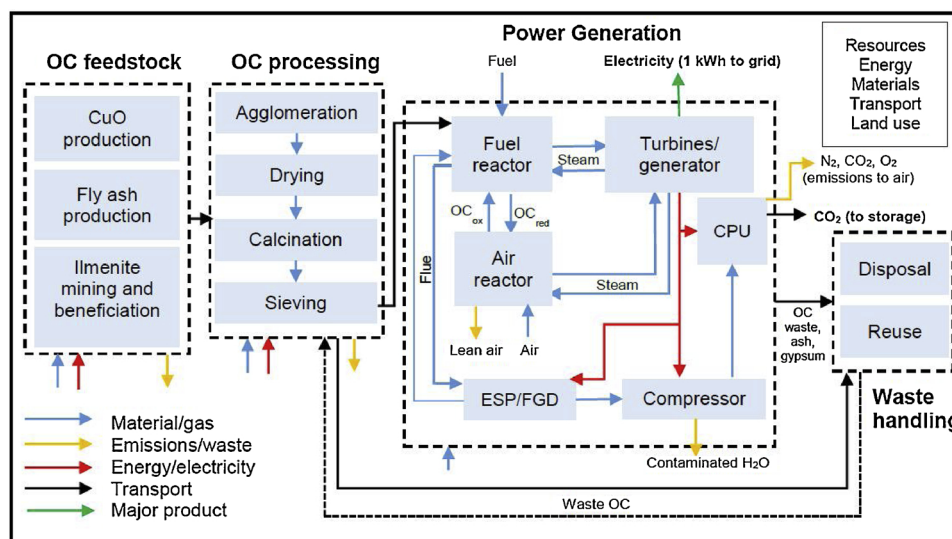
Fig. 2. Process specific foreground map for a hybrid Chemical Looping Combustion (HCLC) system. Only major inputs/outputs are labelled. CPU – CO_2 Purification Unit; ESP – Electrostatic Precipitator; FGD – Flue Gas Desulphurisation.

previously. Since CLC technology is at an early development stage and the study is based on un-optimised lab-scale data, the primary aim of the article is to pinpoint environmental hotspots and provide an optimisation guide for the development of environmentally competitive HCLC technology. The experimental output resulting from OC development, processing for use and testing (Idziak et al., 2017; Krzywanski et al., 2017) is fully integrated and was used as a starting point for the study. Power plant models without CO_2 capture or with post-combustion capture effectively act as environmental performance targets, which the HCLC system must reach in order to be a competitive technology.

2. Methods

Lifecycle environmental impacts of a theoretical HCLC plant were calculated using a process based LCA model carried out according to ISO 14040/14044:2006. In the following subsection the goal and scope definition of the LCA is described (2.1), followed by a presentation of the key parameters for the HCLC and comparative power plant models (2.2). Finally, parameter variations associated with the incremental optimisation of key process elements are described, resulting in a range of analyses (2.3).

2.1. LCA goal and scope

The primary goal of the LCA was to identify the environmental impact of HCLC, and compare it two mature power production technologies, i) a coal-fired power plant without CO_2 capture ('Reference case') and ii) a coal-fired power plant with post-combustion CO_2 capture ('MEA case'), which act as benchmark performance targets. The starting point for the modelling was the lab-scale production of OC (5 kg batches) combined with theoretical HCLC parameters. Lab-scale data with no process/equipment optimisation and solids reprocessing included is not representative of industrial scale production and is not comparable to other mature technologies. Thus, step-wise improvements were made leading to a theoretically optimised HCLC case that could be compared to the other plants. Optimising the model from the lab-scale data starting point first required analysis and identification of 'environmental hotspots', i.e. those parts of the lifecycle that contributed most to environmental impacts. Thus, a secondary goal of the LCA was to act as a guide and show where further research is necessary to lower the environmental impact of the technology.

The functional unit of analysis was defined as 1 kWh of net electricity delivered to grid. The geographical reference for all work was an existing coal-fired Polish coal power plant using simulated Polish industrial conditions, with all relevant impacts included in the assessment.

The LCAs were modelled using SimaPro Analyst v8.1.1.16, and aimed to include the contributions from all relevant lifecycle processes within the system boundaries identified in Section 2.2. Data on material/energy input, emissions and waste was collected directly for foreground processes (experimental or extrapolated results with experimental basis) or from the literature. For background processes or where there was no primary data, standardised data available in the Life Cycle Inventory database ecoinvent v3 was used, containing all the input resources and energy, emissions and waste data resulting from production of different commodities. Background processes were attached to the foreground processes to build the cases (normalising all inputs and outputs per the functional unit). The ReCiPe Midpoint (hierarchist) method was selected for analysing the impact of lifecycle emissions. Impact results are presented for four categories. These impact categories, and their corresponding impact indicators, are: climate change (GWP_{100} in kg CO_2-eq), particulate matter formation (in kg PM_{10}-eq), terrestrial acidification (in kg SO_2-eq) and human toxicity (in kg 1,4 DB-eq). These were selected (out of a possible 18 categories) since they reflect key issues and give comparability with much other LCA literature, as well as avoiding the additional uncertainty associated with endpoint categories.

2.2. Model descriptions and system boundaries

2.2.1. HCLC plant

Fig. 2 shows a process-specific map of the foreground processes for the HCLC power plant system. For simplicity, full descriptions of material input and output are not given in the system boundary figures. The LCA foreground included OC production (OC feedstock and OC processing), power generation (including CO_2 capture and compression, flue gas treatment and a CO_2 purification unit) and waste management. Transport and storage of CO_2 were defined outside the scope of the work.

Key modelled parameters for the HCLC plant are shown in Table 1. Due to the early stage of testing, most key parameters are theoretical. The plant was based on a supercritical 460 MW_e CFB boiler running on Sobieski (hard) coal, with a net electrical efficiency of 43.3%. This

Table 1

Key theoretical power plant parameters. Note: For the HCLC case, [a]Parameter varies when OC recycling from waste is implemented. Emissions were modelled as [b]total sulphur, and [c]total nitrogen, to water.

Parameter		Reference case (no CO_2 capture)	MEA case	HCLC case
General	Power output (MW)	460	460	460
	Technology	CFB	CFB + MEA unit	CFB + HCLC
	Net plant efficiency (%)	43.3	33.3	38.3
	PM capture (ESP) (%)	99	99	99
	SO_2 capture (FGD) (%)	90	90	90
CO_2 capture	Total energy penalty for CO_2 capture and compression (%-points)	–	10	5
	Energy for compression (kWh/kg CO_2)	–	0.103	0.103
	CO_2 capture efficiency (%)	–	90	99
	SO_2 capture from CO_2 unit (%)	–	90	–
	NO_2 capture from CO_2 unit (%)	–	25	–
	PM capture from CO_2 unit (%)	–	50	–
Key material input requirements	Coal (kg/kWh)	0.35	0.46	0.40
	MEA (kg/kWh)	–	0.0025	–
	OC (kg/kWh)	–	–	6.86[a]
Key direct emissions	CO_2 (kg/kWh)	0.80	0.10	0.01
	SO_2 (kg/kWh)	9.85E-4	3.20E-5	1.11E-3[b]
	NO_x (kg/kWh)	1.61E-3	1.57E-3	1.82E-3[c]
	NH_3 (kg/kWh)	–	2.20E-4	–
	MEA (kg/kWh)	–	1.47E-5	–
Waste	Coal ash (kg)	0.030	0.038	0.033
	Solvent sludge (kg/kWh)	–	2.52E-3	–
	OC waste (kg/kWh)	–	–	6.86[a]

efficiency and size is analogous to the CFB boiler unit at Łagisza Power Station in Poland (Jäntti, 2011). With CFB technology dry flue gas desulphurisation (FGD) may be utilised, but here a wet FGD was modelled (capture efficiency of 90%) assuming requirements of new, large and 'clean' units where the dry method may be insufficient. In addition, an electrostatic precipitator (ESP) unit was modelled. The modelled plant was adapted for a HCLC reactor unit utilising a copper based OC, which was based on a CO_2 capture efficiency of 99% and associated energy penalty of 5%points. Net plant efficiency was thus reduced from 43.3% to 38.3%.

Emissions and coal ash waste were calculated using a mass balance approach based on the quantity of coal required per kWh (calculated from the net electrical efficiency and energy content) and the coal elemental composition (Table A3). Emission species depend on the specific OC in question, operating conditions, and gaseous environment (Linderholm et al., 2016). In the present work it was assumed that full carbon and sulphur oxidation occur, and only 20% of coal nitrogen oxidises to form NO_x. Due to the pressurised flue-gas train in CLC systems, oxidation of NO to NO_2 is favoured. After gas treatment, remaining gases in the CO_2 stream are vented at the CO_2 Purification Unit (CPU). Since the solubility of NO_2 and SO_2 in water is high, absorption is known to occur forming sulphuric and nitric acids (Ajdari et al., 2015, 2016; Normann et al., 2013). Thus, NO_x and SO_x emission output were modelled to water and not to air. Other inputs, characteristics of the ESP and FGD units, emissions and wastes were based on pre-existing processes in the ecoinvent database. Key wastes include gypsum from the FGD, fly ash from the ESP, and bottom ash and spent OC from the fuel reactor.

The modelled OC was composed of commercial CuO supported on enriched ilmenite (from Titania A/S, Norway) and waste material (fly ash) in a 60:20:20 wt% ratio. To obtain usable OC, feedstock materials were combined, agglomerated, dried, calcined and sieved to fraction 80–335 μm. Key estimated processing parameters (processing energy and processing losses) are shown in Table 2 and are based on the experimental results of the production of 5 kg OC batches at lab-scale. An OC composed of 100% ilmenite (as used in other studies), which only required sieving and drying steps, was also produced and is shown for comparison in Table 2. Processing 'losses' account for both solid material losses and humidity mass losses due to dehydration. The demand for energy was calculated by taking into account furnace power, residence time of the samples in the furnace and the mass of the furnace

Table 2

Experimentally derived oxygen carrier (OC) processing parameters, at a 5 kg batch production scale. Data on the required OC:coal ratio, based on fuel-OC interaction studies, is also shown here.

Parameter		Cu-based OC	Ilmenite OC
Composition (wt%)	CuO	60	0
	Fly ash	20	0
	Ilmenite	20	100
	Mn tailings	0	0
OC:coal ratio		17.10:1 (10% O_2 excess)	43.20:1 (5% O_2 excess)
Processing losses (wt%)	Agglomeration	40	NA
	Drying	2	2
	Calcination	2	NA
	Sieving	6	3
	Total	50	5
Processing energy (kWh/kg)	Agglomeration	10	NA
	Drying	8	6
	Calcination	25	NA
	Sieving	5	2
	Total	48	8
OC waste		Landfill	Landfill

charge, although it should be noted that the power consumption of lab devices would not scale-up linearly for commercial large-scale production. The enriched ilmenite and commercial CuO feedstocks were modelled in the LCA using existing ecoinvent processes. The ilmenite feedstock process included both mining operation and beneficiation, whilst the CuO was principally based on primary Cu production from copper(II)oxide by hydrometallurgy. The analysis was extended to CuO produced from secondary Cu (sourced from metal scraps). For the LCA purposes, waste materials were considered burden free. Assumed material transport distances are given in Figure A1.

OC requirement per kWh at the HCLC plant was calculated based on the required OC:coal ratios for combustion (shown in Table 2). This is a fundamental material property determined by the oxygen carrying capacity and the OC performance, experimentally derived for each OC material. It was assumed that the OC reacts with coal and is purged from the system (either continuously or batch-wise) from the reactor together with the coal ash, as there is no in-situ separation of ash and OC. Instead, both materials are purged from the fuel reactor after cycling due to assumed mass flow properties. For continuous operation,

Table 3

Individual LCAs regarding HCLC electricity production.

Section	LCAs	Parameter variation				
		OC Processing losses (wt%)	OC Processing energy (kWh/kg)	OC recycling efficiency from plant waste (wt%)	Source of Cu feedstock	OC processing energy type
3.1.1	Theoretical HCLC baseline	50	48	0	Primary Cu	Polish grid electricity
3.1.2	Effect of improving OC production and use parameters	1	2 – 0.1	95 – 99	Primary Cu Secondary Cu	Polish grid electricity
3.1.3	Other potential industrial scale optimisations (OC processing energy type)	1	2 – 0.1	95 – 99	Secondary Cu	Electricity integration at HCLC plant

the flow of OC material exiting the reactor should thus equal the OC make-up flow entering it. The starting point for modelling 'fresh' OC requirement is consequently in contrast to other CLC LCA studies where the OC is modelled with a lifetime defined only by particle attrition and reactivity losses. For example, Fan et al. (2018) defined the duration of the OC by hours (up to 10,000), whilst Petrescu et al. (2017) removed a small fraction (1%) of the solid flow from the air reactor to account for OC deactivation. In the described system, the stoichiometry between OC, fuel and ash leads to a significant flow of OC leaving the reactor. However, this does not imply that the OC is deactivated and cannot be recycled. It is likely to assume that ash and OC can be suitably separated and OC can be reintroduced into the reactor system via an external recycle. Therefore, the introduction of OC recycling is one of the steps towards process optimisation discussed in this article.

2.2.2. Benchmark plants

To provide targets for the environmental optimisation of the HCLC electricity production, two benchmark (theoretical) lifecycle models were developed. This included a coal-fired power plant with no CO_2 capture (referred to as the 'Reference case'), and a power plant with post-combustion CO_2 capture using monoethanolamine (referred to as the 'MEA case'). Process-specific maps of the foreground processes for the Reference and MEA cases are shown in Figure A2 and Figure A3 of the supplementary information, respectively. Key modelled parameters of the Reference and MEA cases are listed in Table 1.

The Reference case (no CO_2 capture) was modelled on a supercritical 460 MW_e CFB boiler, running on coal with a net electrical efficiency of 43.3%. Emissions, coal ash waste, wet FGD and ESP were modelled as for the HCLC case. CO_2, NO_x and remaining SO_x were assumed vented to air. Of the NO_x produced, around 5% was assumed as NO_2, 5% as N_2O, and 90% as NO (Mittal et al., 2012; TSI, 2004).

The MEA case was based on the Reference case, adapted for a retrofitted post-combustion CO_2 capture unit utilising MEA solvent. The CO_2 capture unit (including NH_3/MEA input requirements and MEA/NH_3 emissions) was modelled according to Koornneef et al. (2008), together with a CO_2 capture efficiency of 90% and associated energy penalty of 10%points. This reduced the net plant electrical efficiency from 43.3% to 33.3%. Additional spent solvent waste (sludge) is also generated, as described by Koornneef et al. (2008). MEA production was modelled using the process available in ecoinvent v3. Waste disposal of the capture process was modelled using a generic waste treatment process using values from Koornneef et al. (2008).

Infrastructure and decommissioning of the facilities were included in the LCA analysis (sourced from ecoinvent v3) although differences in required construction materials between the different plant types (per kWh) were considered negligible.

2.3. Analyses

2.3.1. Incremental optimisation of HCLC plant

Areas where the main reductions in environmental impact may be made were identified through a series of contribution analyses combined with variation of key operational plant parameters. The parameter variation reflects example potential improvements expected by performing process optimisation and upscaling from lab-scale (kg) to industrial-scale (t) OC batches, as well as other optimisations. OC processing parameters (processing losses and processing energy) were focused upon, as well as use of secondary Cu and on-site integration of the OC production process plant with the HCLC power plant. In addition, a proportion of the purged OC was assumed regenerated and fed back into the fuel reactor ('OC recycling'), as technically demonstrated by Garcia-Labiano et al. (2007). Determining realistic levels of improvement of OC processing parameters with scale-up is challenging. One way to scale up these processes is by modelling a specific industrial-scale processing plant and all the equipment/processes it contains (Piccinno et al., 2016), but scaling models based on a relationship between capacity and key properties may also be applied (Caduff et al., 2014). With an increase in OC batch size of around three orders of magnitude (kg to t scale), it is realistic to expect that economy of scale effects may improve respective OC processing parameters between one and two orders of magnitude. The latter approach was taken in this work, and OC processing parameters were varied within these limits. A more thorough approach was beyond the scope of this article. It should be taken into account that the optimization presented in this article can also be read as optimization targets required to decrease the environmental impacts associated with HCLC electricity generation.

The LCAs and the corresponding section of the results they are described in are shown in Table 3. Where not detailed, all parameters are as described in Table 1 and 2. Aside from the HCLC baseline case, the LCAs are sets where two parameters (e.g. OC processing energy and OC recycling efficiency from plant waste) were varied together through a range of values. Each range was split into five values, resulting in 25 total analyses per set.

Table 4

Individual LCAs regarding HCLC electricity production.

Parameter	HCLC baseline	HCLC 1	HCLC 2	HCLC 3	HCLC 4	HCLC 5 ('Optimised case')
OC processing losses (wt%)	50	1				
OC processing energy (kWh/kg)	48		1			
OC recycling efficiency from plant waste (wt%)	0			99		
Source of Cu feedstock	Primary Cu				Secondary Cu	
OC processing energy type	Polish grid electricity					Electricity integration at HCLC plant

2.3.2. Comparisons of optimised HCLC plant model with benchmark plants

Based on Section 2.3.1, six specific HCLC LCAs were carried out (Table 4), with cumulative step-wise degrees of optimisation from the 'HCLC baseline' (the starting point of lab-scale production of OC combined with theoretical HCLC parameters). The final 'optimised case' (HCLC 5) was compared in more detail to the coal-fired power plants with and without post-combustion CO_2 capture.

3. Results and discussion

For a newly developed technology to be competitive it should ultimately perform better than existing (competing) technologies. However, it is not realistic to compare a theoretical HCLC plant based on lab-scale experimental data, without any assumed improvement associated with the upscaling to an industrial process. Although the magnitude of improvements that would be expected to occur when scaling up OC production from lab (kg) to industrial (t) scale are unknown, optimisation of certain equipment and methodologies would certainly result in a large degree of process and equipment optimisation, as well as recovery and reuse of waste.

In the following sections, lifecycle environmental impacts of HCLC power generation are thus first discussed in detail in relation to improvement of key OC processing parameters (3.1). Subsequently, the overall comparative analysis between the theoretically optimised HCLC power plant and a reference CFB power plant with and without post-combustion CO_2 capture is presented (3.2). In addition, a sensitivity analysis is provided discussing uncertainties from a broader perspective (3.3).

3.1. Incremental optimisation of HCLC plant

3.1.1. Theoretical HCLC baseline

When the experimental lab-scale OC production data is used directly in a theoretical HCLC plant, impacts are high with lifecycle GHG emissions of over 700 kg CO_2-eq/kWh, almost all of which (> 99.9%) relate to OC production. Other lifecycle impacts were also high at over 4 kg SO_2 eq/kWh, 1 kg PM_{10}-eq/kWh and 800 kg 1,4 DB-eq/kWh. The case included 0% OC recycling efficiency, 50 wt% OC processing mass losses and an OC processing energy requirement of 48 kW h/kg. The OC CuO feedstock contained primary Cu. These results clearly show a necessary improvement potential of three orders of magnitude from lab-scale experimental data to become comparable to a conventional coal-fired power station with and without post-combustion capture.

Contribution analysis (Fig. 3) shows that impacts here are dominated by the Polish grid electricity required for OC processing, which has high lifecycle emissions since it is largely derived from hard coal and lignite combustion. Some contribution also derives from the OC feedstock material production, with the importance of this varying depending on the impact category in question. Whilst the OC feedstock material did not contribute greatly towards lifecycle GHG emissions (~3%), around 35% of human toxicity derives from the OC feedstock. Over > 99% of this can be attributed to the CuO feedstock, which stems from the primary Cu production and associated mining.

When looking at the contribution of individual OC production processes towards impact (and taking lifecycle CO_2-eq emissions as an example), most lifecycle CO_2eq emissions derive from calcination (44%) and agglomeration (33%) since these processes require most energy and/or are where most mass losses are incurred. Contributions from the fly ash and Mn tailings are almost zero, since the materials were treated as burden-free waste with only transport emissions accounted for. OC processing is therefore the key environmental hotspot to optimise.

It is challenging to compare these baseline results to other studies due to changes in the LCA scope and system boundaries. Regardless, it is apparent that by including the processing losses and processing energy required for OC production, and modelling the OC material as a
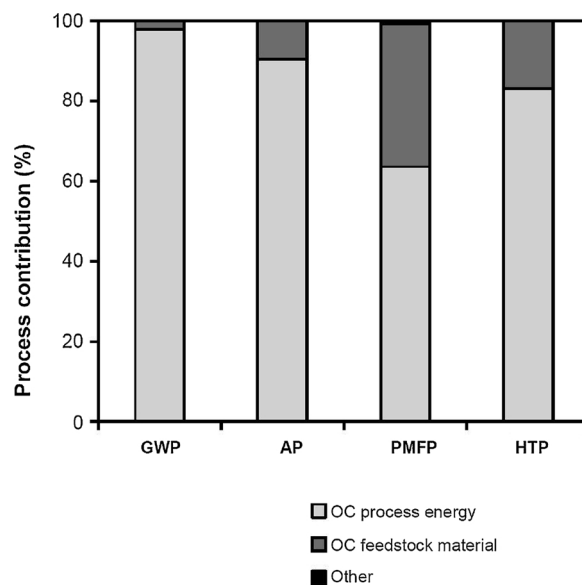
Fig. 3. Key contributing processes towards selected lifecycle impacts, per kWh produced at the HCLC plant (baseline HCLC case). GWP = Global Warming Potential, AP = Acidification Potential, PMFP = Particulate Matter Formation Potential and HTP = Human Toxicity Potential.

consumable rather than as defined by its lifetime/looping ability, impacts can be expected to be much larger than reported in literature. For example, per kg OC produced alone, lifecycle GHG emissions are calculated here as 100 kg CO_2-eq/kg OC, in comparison to 1–2 kg CO_2-eq/kg Fe based OC and 11 kg CO_2-eq/kg Ni based OC used by Fan et al. (2018) and Wang et al. (2018). As a result, other studies report much lower lifecycle GHG emissions for HCLC electricity production, e.g. 0.063 kg/kWh with a Ni based OC (with a lifetime of around 4000 h) as used by Fan et al. (2018), and 0.34 kg/kWh with an ilmenite OC as used by Petrescu et al. (2017). Nevertheless, in common with this study, these studies also determined that the OC contributes significantly towards total impacts per kWh.

3.1.2. Effect of improving OC production and use parameters

Optimising OC processing and use means reducing processing losses and processing energy (kWh/kg), as well as implementing OC recycling from waste to effectively increase OC lifetime. Reductions in OC processing losses and energy per kg OC may be achieved by ensuring recovery and re-processing of waste material, utilising equipment with higher efficiency, and generalised process optimisation.

From the previous section it follows that a reduction in processing energy (from 48 kW h/kg OC to processing energies in the order of 1 kW h/kg OC) should be the key focus to reduce environmental impacts. Reducing processing losses from 50 wt% by one order of magnitude may also give significant reductions in climate change potential, but is dependent on processing energy. When a certain percentage of OC material is lost during production an extra amount equal to that quantity must be processed to replace it and this comes with higher associated environmental impacts when the required processing energy is high. However, when processing energy is already low, the improvement from optimising processing losses is negligible for all impacts aside from for human toxicity, where there is a remaining significant contribution from Cu mining. Due to the domination of the electricity contribution to environmental impacts, variation in other OC production parameters did not lead to significant changes in impacts.

For further analysis we therefore set OC processing losses at 1 wt% and vary OC processing and OC recycling efficiency between 2 kWh/kg OC and 0.1 kWh/kg OC, and 95 wt% and 99 wt%, respectively. Fig. 4 shows the impact indicator results for climate change, acidification,
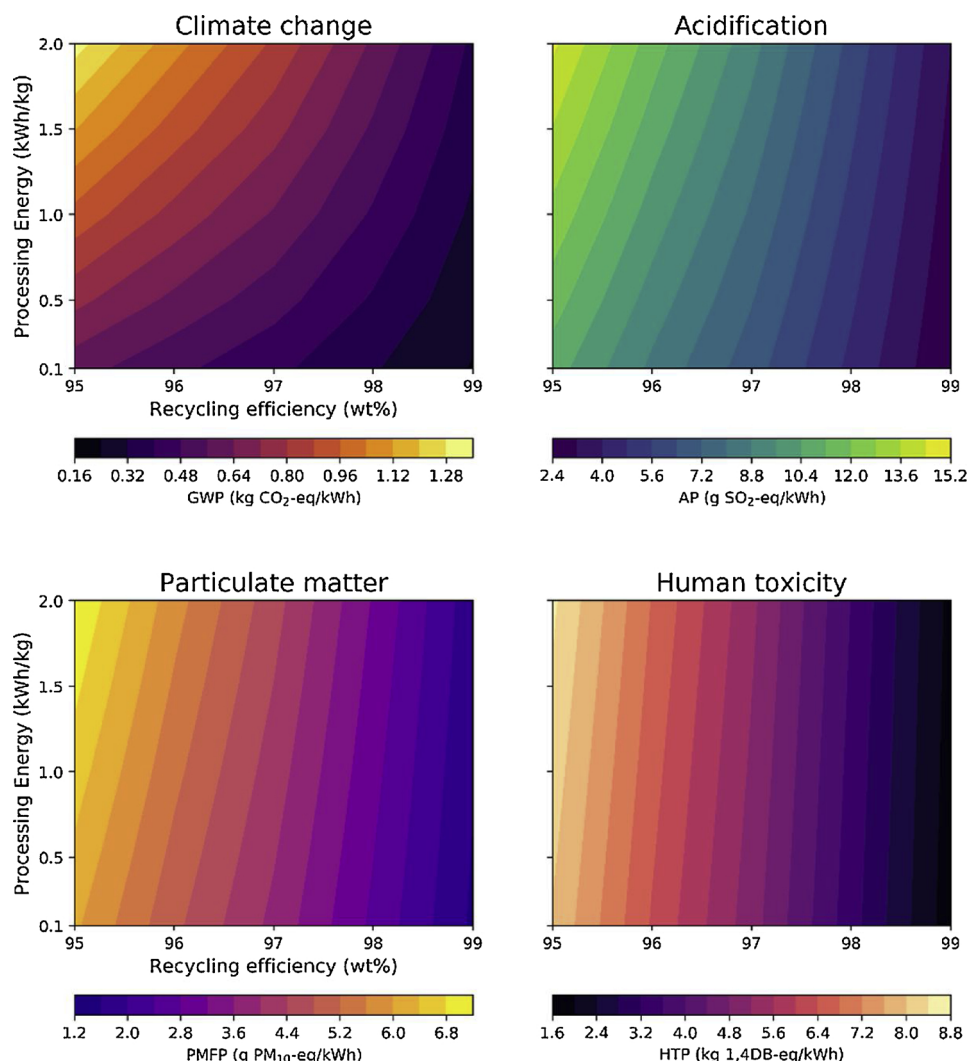
Fig. 4. Effect of improving processing energy and OC recycling efficiency on selected lifecycle impacts, per kWh produced at the HCLC plant. GWP = Global Warming Potential, AP = Acidification Potential, PMFP = Particulate Matter Formation Potential and HTP = Human Toxicity Potential.

particulate matter and human toxicity. It should be noted that implementation of recycling in the model did not include energy and resource requirements for OC re-processing. Results show that both increasing the recycling efficiency, as well as decreasing OC processing energy, decreases impacts to varying degrees across all four investigated categories. This is since these factors affect both the 'net consumption' of OC and quantity of electricity input required. The relationship found here with recycling efficiency effectively corresponds with findings from Fan et al. (2018), who performed a similar analysis by incrementally increasing the lifetime of the OC. Nonetheless, whilst e.g. climate change impacts are similarly dependent on both recycling efficiency and OC processing energy combined, as expected human toxicity is much more dependent on OC recycling efficiency alone. This is due to the changes in the quantity of fresh OC required in the reactor, and the relationship of human toxicity with Cu mining.

For an example case from the figure with 99% OC recycling efficiency, 1 wt% OC processing mass losses and an OC processing energy requirement of 1 kW h/kg (HCLC 3 in Table 3), lifecycle impacts are around 0.3 kg CO_2-eq/kWh, 2.8 g SO_2-eq/kWh, 1.6 g PM_{10}-eq/kWh and 1.8 kg 1,4 DB-eq/kWh. This is a reduction of between two to three orders of magnitude from the HCLC baseline. Approximately 4% of total lifecycle GHG emissions per kWh are direct, deriving mostly from the CPU of the plant where 1% of the 'captured' CO_2 stream is assumed emitted.

Despite improvements to OC production and use, lifecycle emissions remain dominated by the production and processing of the OC. Although the contribution of indirect emissions from electricity generation remain significant, a greater contribution of impacts now derive from the production of the feedstock materials (> 99% of which stems from the CuO feedstock as a result of primary Cu production) compared to the HCLC baseline. Much of the remaining contribution ('other' processes) derives from the contribution of the hard coal feedstock combusted at the plant, i.e. emissions occurring during hard coal mining.

Switching the source of OC Cu to recycled scrap –and thus avoiding primary Cu mining – results in a large reduction to all impact categories but climate change (Fig. 5). For the analysis, other parameters in the figure were set at 99% OC recycling efficiency, 1 wt% OC processing mass losses, an OC processing energy requirement of 1 kW h/kg. Human toxicity potential is reduced from 1.8 kg 1,4 DB-eq/kWh to 0.4 kg 1,4 DB-eq/kWh, terrestrial acidification potential is reduced from 2.8 g SO_2-eq/kWh to 1.1 g SO_2-eq/kWh and particulate matter formation potential is reduced from 1.6 g PM_{10}-eq/kWh to 0.6 g PM_{10}-eq/kWh. Lifecycle GHG emissions are not significantly reduced since the feedstock contribution is less significant. In addition, although the secondary Cu material was treated as burden-free waste, re-processing energy (and thus indirect GHG emissions deriving from fossil fuel combustion) is associated. Using secondary Cu is important not just for
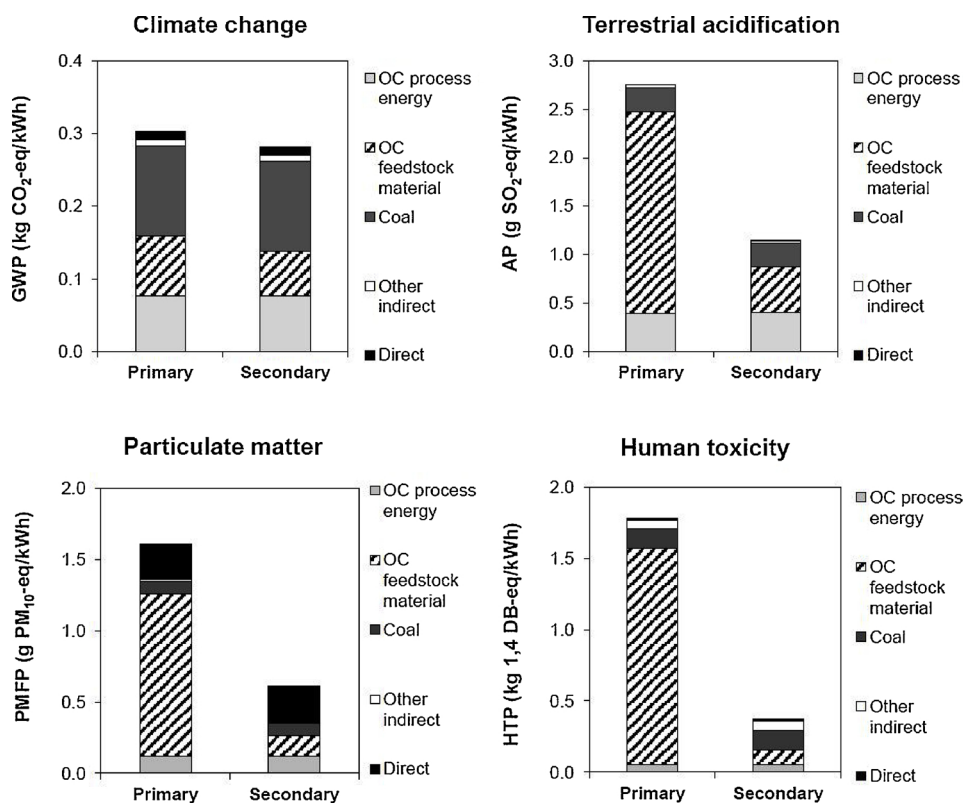
Fig. 5. Key contributing processes towards selected lifecycle impacts, per kWh produced at the HCLC plant (for HCLC 3 with primary Cu and HCLC 4 with secondary Cu from recycled scrap). GWP = Global Warming Potential, AP = Acidification Potential, PMFP = Particulate Matter Formation Potential and HTP = Human Toxicity Potential.

reducing environmental impact associated with primary Cu use, but also for sustainable resource use.

The primary way to decrease impacts further is by decreasing the quantity of Cu required per kWh. This may be achieved by either changing the OC composition (which may be in conflict with the required oxygen carrying capacity of the OC and the OC/fuel ratio needed in the reactor, among other factors), or by increasing the level of OC recycling.

3.1.3. Other potential industrial scale optimisations

Previous sections show that reduction of OC processing losses, OC processing energy, implementing OC recycling with high efficiency and use of secondary Cu in the CuO feedstock are important improvement parameters for reducing the lifecycle environmental impacts associated with electricity production via HCLC. An additional process improvements that may occur with scale up is OC processing on the HCLC plant site using electricity produced on-site (rather than the grid mix).

To illustrate the effects of integrating electricity use for OC processing, the relative change to lifecycle GHG emissions as a function of OC processing energy (2 kW h/kg OC to 0.1 kW h/kg OC) and OC recycling efficiency (95 wt% to 99 wt%) is shown in Fig. 6. OC processing losses were set for this analysis at 1 wt% (and the OC CuO feedstock contained secondary Cu). As can be seen, there is no benefit to using HCLC electricity for OC processing when recycling efficiencies are relatively low and processing energy is high (top left corner). When processing energy is lowered or recycling efficiency increases, additional benefits in terms of a reduction in GWP appear upon integrating electricity use. The highest reduction is calculated when both OC recycling efficiencies and OC processing energy are high (top-right corner). This situation can be explained as follows. Where OC processing is integrated, the net plant efficiency is decreased. For the HCLC 5 case, calculations show that implementing OC processing at the HCLC site reduces the net efficiency from 38.3% to 35.1% (extra penalty of 3% points). Not only does this require more coal to be processed, but more importantly, additional OC to be processed, per net kWh
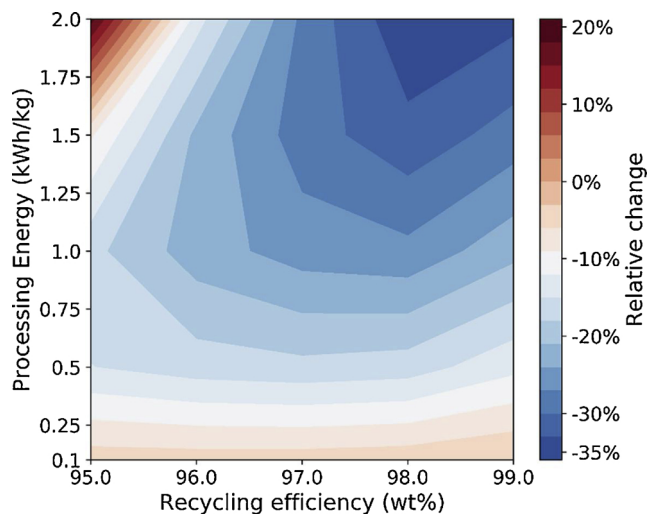

Fig. 6. Relative change to global warming potential (GWP, CO_2-eq) by switching from grid electricity for OC processing to electricity produced on-site at the HCLC plant.

produced (i.e. lifecycle CO_2-eq emissions from upstream processes increase). Since the additional OC processing decreases the net plant efficiency further, a feedback effect occurs resulting in a rise in lifecycle CO_2-eq with increasing processing energy demand or lack of recycling. Conversely, at high recycling rates and high processing energy costs, the emissions reduction gains from integrating are considerable. While improvements from integration may be high at this point, it should be noted that absolute per kWh GHG emissions are still lowest at high recycling and low processing energy. Therefore, where conditions are favourable, the situation with OC processing integrated at the HCLC plant site with high OC recycling rates potentially represents a mature development scenario for HCLC technology.
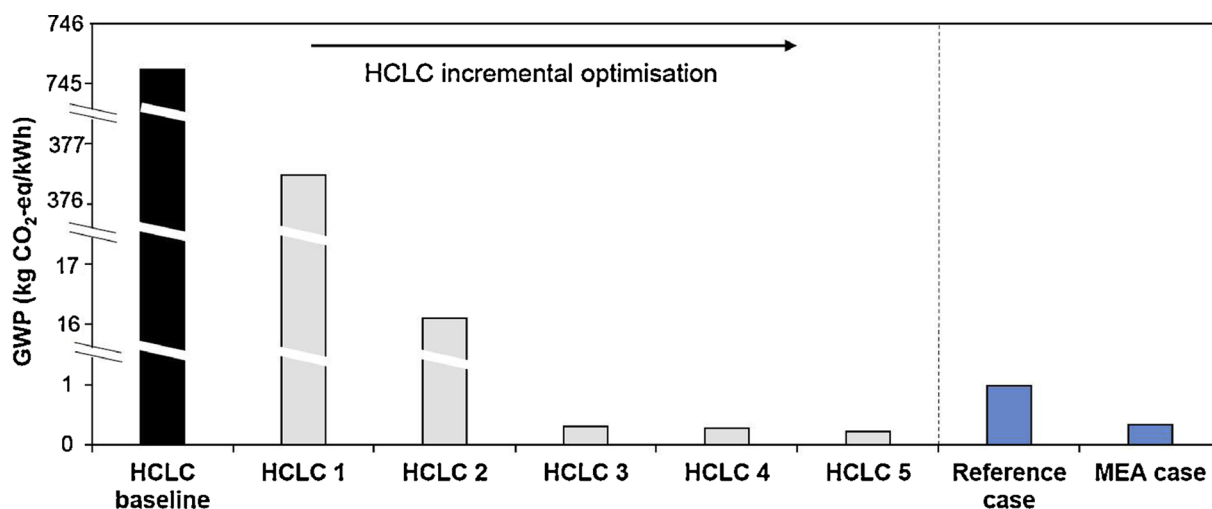
Fig. 7. Optimisation of HCLC environmental performance, compared to the Reference and MEA cases, by implementing additive improvements (see Table 3 for definition of the HCLC cases). The y-axis is broken in three places to allow for the large range of improvements. GWP = Global Warming Potential.

3.2. Comparisons of optimised HCLC plant model with coal-fired power plant with and without post-combustion CO_2 capture

Fig. 7 shows the effects of step-wise (cumulative) performance improvements of the HCLC power plant according to the optimisations presented in Section 3.1, and compares them to the comparative environmental targets set by the Reference and MEA cases. Table 4 should be consulted to obtain the specific parameters for the six HCLC cases.

In the figure, GWP is used as an indicator; results are thus presented in terms of kg lifecycle CO_2-eq (GHG) per kWh electricity. The difference between the baseline case (lab-scale OC without recycling) and 'optimised' case (HCLC 5) is several orders of magnitude. The final optimised case in the figure (HCLC 5) - used further in this section for discussion - includes 1 wt% OC processing losses, 99% OC recycling from plant waste, 1 kW h/kg OC processing energy, use of secondary Cu (scraps) in the OC feedstock and processing integrated at HCLC plant, and has a GWP reduction of 99.9% from the HCLC baseline. Results thus show that HCLC technology has the potential to be environmentally competitive with other carbon capture technologies.

In addition, the Reference and MEA case provide a comparative benchmark for other lifecycle environmental impact categories. Comparative details of other environmental impacts of the optimised industrial HCLC 5 case against the Reference and MEA cases are shown in Fig. 8. As can be seen, lifecycle GHG emissions of the MEA and HCLC 5 cases are respectively 66% and 78% lower than the Reference coal fired power plant on a per kWh produced basis. The HCLC 5 case also has a 38% lower terrestrial acidification potential than the Reference case, whilst the particulate matter formation potential is similar. However, the human toxicity potential of the HCLC 5 case is greater compared to both other cases by around a factor of two. HCLC 5 thus performs better than the MEA case in terms of GHG emissions and acidification, but worse in terms of particulate matter emissions and human toxicity.

Fig. 8 distinguishes between direct and indirect emissions. Direct emissions are those emitted directly during OC production and processing whilst indirect emissions occur either upstream or downstream along the value chain, such as those emitted during the production of grid electricity that is used for OC processing. The impact of direct emissions is most significant with the Reference case, which is to be expected due to the fact that CO_2 is not captured, but emitted directly at the power plant. Indirect emissions make up most of the lifecycle impacts of the HCLC case as a significant amount of emissions is associated with the energy requirements for producing the OC as well as the OC feedstock from secondary Cu metal. In all cases, the hard coal feedstock

is a significant source of indirect emissions per kWh produced. Quantitative comparisons of these results with other studies are difficult due to differences in technology and LCA system boundaries, but trends are similar (Corsten et al., 2013; Cuellar-Franca and Azapagic, 2015; Zapp et al., 2012).

3.3. Sensitivity analysis and uncertainties

The key uncertainty in this study is the question of how OC production and HCLC plant parameters would change for an industrial-scale case. For the work, OC processing energy and losses were considered improved based on the expectation of typical economies of scale effects, and not based on specific modelling of industrial processes. Despite these uncertainties, this article nonetheless does not intend to act as a predictive target, and choices made here are merely intended to act as a guide as to how the environmental impact of HCLC technology can be reduced (with an emphasis on trends, rather than absolute values for specific conditions). Nevertheless, the OC processing energy type, a key parameter towards impacts, was varied here by means of sensitivity analysis.

The electricity type primarily used for OC processing in the LCA model (Polish grid) was varied to simulate the effect of OC production in different regions, and also reflects the uncertainty associated with the electricity mix in LCA models (Marriott et al., 2010). The extent of the impact of this change varied depending on the degree of model optimisation and therefore the contribution of OC processing energy towards total lifecycle impacts. For the example case using secondary Cu (HCLC 4), where OC processing energy is not a significant contributor towards total lifecycle GHG emissions, results are 0.28 kg CO_2eq/kWh, 0.25 kg CO_2eq/kWh and 0.21 kg CO_2eq/kWh with Polish, average World or Norwegian grid electricity, respectively. However, for the theoretical HCLC baseline case where OC processing energy is significant, results are 745 kg CO_2eq/kWh, 468 kg CO_2eq/kWh and 31 kg CO_2-eq/kWh with Polish, average World or Norwegian grid electricity, respectively. This reflects the varying share of renewable energy production towards total electricity production, and hence the quantity of lifecycle GHG emissions from fossil fuel combustion, in each region; According to EIA data (2018), the contribution of renewable energy sources towards total net electricity production in the year 2015 for Poland, the average World, and Norway was 14.8%, 24.0% and 98.6%, respectively. Although this demonstrates that environmental impacts vary depending on the location of OC processing, there is still a requirement in all regions for processing energy to be reduced in order to make all impacts competitive to competitor technologies.
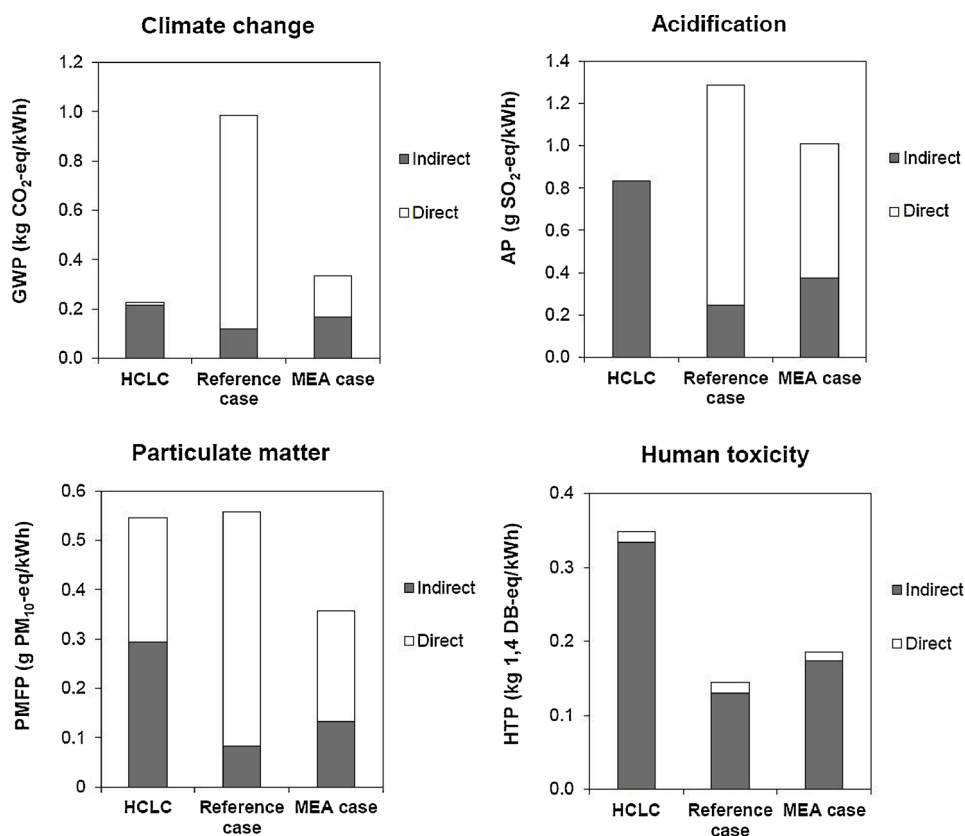
Fig. 8. Key environmental impact indices for the Reference, MEA, and optimised HCLC (HCLC 5) cases, relating to direct and indirect emissions. GWP = Global Warming Potential, AP = Acidification Potential, PMFP = Particulate Matter Formation Potential and HTP = Human Toxicity Potential.

4. Conclusions

In this article the environmental impacts for electricity production with HCLC are presented and compared with a reference coal fired power plant with and without post-combustion CO_2 capture. The analysis in the article departs from the experimental data to synthesise and produce the OC material at lab-scale, and scales this up to the scale of an industrial process. The LCA enables the identification of environmental hotspots and the comparative analysis with other (more mature) technologies allows for theoretically optimising those parts of the process to meet a performance target, indicating how OC production and associated HCLC technology may evolve to convey the required reduction in environmental impacts.

Though direct GHG emissions from an HCLC plant were found to be negligible due to high CO_2 capture efficiencies, lifecycle GHG emissions of an HCLC plant where OC is processed using the experimental lab values were much higher than the other cases, per kWh produced (over 700 kg CO_2eq/kWh compared to < 1 kg CO_2eq/kWh). Results indicated that total GHG emissions were dominated by the electricity required for OC processing. This is largely due to the upstream use of fossil fuels for the electricity production. Consequentially, the reduction of OC processing energy and mass losses (from a lab-scale baseline of 48 kW h/kg OC and 50 wt% losses down to 1 kW h/kg OC and 1 wt% losses, respectively) resulted in significant reduction in lifecycle GHG emissions for OC production. Implementation of OC recycling from plant waste reduced the quantity of fresh OC required and effectively decreased the contribution of OC to the lifecycle impacts. For the parameters used, an upper limit to OC processing energy of just under 2 kW h/kg OC was established for favourable integration of OC production at the HCLC plant. Optimisation of OC processing losses, OC processing energy and OC recycling rate reduced lifecycle GHG emissions such that the primary contributions derived from (primary) Cu mining. Thus, utilising a recycled Cu as feedstock for the OC production further reduced many environmental impacts such as human toxicity potential, and is also important for sustainable resource use.

Comparing the environmental impacts resulting from the production of 1 kW h from the 'optimised' HCLC case model to a coal fired power plant with no CO_2 capture (Reference case) and a coal-fired power plant with post-combustion CO_2 capture (MEA case), showed that potential improvements could be made with climate change and acidification potential, but increases made to other impacts such as human toxicity potential.

HCLC technology thus has potential as an energy production method with low environmental impact only if the whole OC production and waste processing chain can be streamlined. In particular, the energy requirements for OC processing should be minimised through technological and process optimisation approaches. Since processing requirements are particularly dependent on the OC selected, a great effort is required to identify an OC type that has a high oxygen carrying capacity (and hence a low coal-to-OC ratio) coupled with a low environmental impact, per kg OC produced. In practice this most likely requires use of waste or recycled materials that additionally have a low contribution from their feedstock.

Results also indicate that it is important to develop and implement technology for OC recycling of plant waste, to reduce the quantity of fresh OC required. The OC should consequently be able to endure a large number of total cycles. The number of OC cycles undergone in the reactor depends on the OC inventory and the circulation rate, and in practice, it is the quantity of OC purged that determines the effective total cycling number of OC in the reactor. To calculate this, we need to know the purge rate of OC (out), the total inventory of OC in the system (both reactors), and the time for one OC cycle (calculated from the flow velocity). These components are not defined for this theoretical system, but should be investigated in future work.

Future LCA work should focus on collecting increased primary data for OC feedstock and processing to reduce uncertainties associated with use of data contained in standard lifecycle inventory databases. Due to the significance of the OC, including all inputs and outputs for OC production and use is of key importance for LCA studies. In addition, an effort should be extended to the collection of primary data for any OC re-processing (effectively re-use from plant waste), directly coupled to experimental work.

Acknowledgements

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.ijggc.2019.04.011.

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