# Life cycle assessment of optimised chemical looping air separation systems for electricity production

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## Abstract

Chemical looping air separation (CLAS) is as a very promising technology for the production of pure oxygen through the cyclic reduction and oxidation of a solid material at elevated temperatures. This study focused on the environmental potential of electricity and $CO_2$ production through oxyfuel combustion of lignite. First, an attributional LCA assessed the operations which mainly contribute to the total environmental impacts for two different scenarios at the limits of the operating window (100% and 25% active material). Then, this study analysed the potential of electricity and pure $CO_2$ production through CLAS when compared with conventional power production technologies from renewable and fossil alternatives, including electricity from hydro power, electricity from wind power, electricity from nuclear, electricity from photovoltaic, electricity from biogas, electricity from biomass, electricity from waste, electricity from hard coal and electricity from natural gas. Overall the results, analysed per MJ of electricity produced, showed how the chemical looping technology consistently performs better than the other technologies, especially thanks to the recovery of the pure $CO_2$ stream used for industrial purposes, which avoids the production of $CO_2$ from fossil resources. However, the cleaning of the flue gas of the oxyfuel combusted lignite strongly limits the toxicities indicators.

## Key words

Chemical looping air separation, life cycle assessment, energy production systems, carbon capture and utilization

## 1.Introduction

In 2013, the UK energy supply sector, including the production and use of electricity and natural gas, was responsible for 47% of the total domestic greenhouse gas emissions. A well-defined relationship between the world's energy use and the increase of carbon dioxide in the atmosphere with the associated temperature increase is established (Shah et al., 2013). The

Intergovernmental Panel on Climate Change (IPCC) has advised limiting the temperature rise well below 2°C (IPCC, 2007a) compared to pre-industrial levels to limit the threats (McMichael, 2013) of global warming. In 2009 and 2014, the EU set binding targets to cut the greenhouse gas emissions by at least 20% and 40% (from 1990 levels) by 2020 (The European Parliament And The Council Of The European Union, 2009) and 2030 (European Council, 2014), respectively. The UK has strengthened its commitment to tackle climate change under the international agreement with the Climate Change Act (UK Government, 2008). With interim budgets, this act reflects the UK contribution to cut the global carbon emissions of 50% by 2050 (compared to the 1990 level) corresponding to an increase in temperature of 2°C by 2100.

Therefore, political support is given to the development of advanced low carbon energy technologies to meet emission reduction targets. A combination of a) reduction of energy consumption by increasing energy conversion efficiencies and b) novel technologies based on less carbon intensive fuels including renewable energy sources (biofuel, wind power, etc.) is the key to reduce the environmental impacts of the energy sector. However, especially in the case of energy produced from fossil resources, these solutions need to be complemented with carbon capture (CC), which is a suitable pathway for the reduction of $CO_2$ emissions to the atmosphere when associated with $CO_2$ utilization (CCU) or storage (CCS) (Adanez et al., 2012). According to UK National Grid (National Grid, 2014), carbon capture is essential for the production of clean energy and is projected to fully develop in the near future. CC technologies produce a pure and concentrated stream of $CO_2$ from industrial and energy-related sources, which can be used in industrial synthesis as a substitute of alternatively produced $CO_2$ or transported to and stored into underground deposits to prevent it from being emitted to the atmosphere for a long period of time.

Three main approaches are available for $CO_2$ capture in energy production technologies (Toftegaard et al., 2010): i) post-combustion - separating the $CO_2$ in the flue gas from the other components, ii) pre-combustion - converting the fuel for the power plant into $CO_2$ and a carbon-free combustible, e.g. hydrogen, and then separating $CO_2$ from the hydrogen - and iii) oxy-fuel combustion - separating the combustion air into nitrogen and oxygen and then combusting the fuel in pure $O_2$ to obtain a mixture of $CO_2$ and water vapour as flue gas, from which a pure $CO_2$ stream is easily separated. However, most of these technologies, especially the oxyfuel systems, are associated with a high energy penalty, which results in a reduction of the overall energy efficiency of the energy production process. Pure oxygen is commonly produced at industrial scales through air separation using either cryogenic-distillation (CASU) or adsorption systems (ADS) (Moghtaderi, 2010). Even though conventional cryogenic air separation methods are mature technologies, they are capital intensive and typically lead to 3–4% energy penalty in oxyfuel operations, resulting in revenue losses, increased fuel utilization per unit of electricity generated and indirectly, increased environmental impacts of the oxy-fuel coal fired power generation (Shah et al., 2013). At the same time, oxygen based adsorption systems remain expensive due to installation, integration and maintenance and the specific power consumptions of these plants are not much lower than their cryogenic counterparts (Shah et al., 2013).

Thus, great efforts have been carried out recently to develop new low-cost carbon capture oxyfuel technologies and chemical looping air separation (CLAS) has been identified (Adanez et al., 2012) as a very promising technology for pure oxygen production thanks to the cyclic reduction and oxidation of a solid material serving as an oxygen carrier. The system usually consists of two fluidized bed reactors in which continuous recirculation of metal oxide particles occurs. In the two interconnected reactors, oxidation (eq 1,) and reduction (eq 2) of carrier particles take place. Air is fed into the oxidation reactor, so that the incoming reduced carrier particles can be regenerated to a higher oxidation state. The regenerated carrier particles, in turn, are transported back to the reduction reactor, where oxygen decoupling occurs in the presence of a gas with a negligible oxygen partial pressure, such as steam. Hence, the production of a pure oxygen stream is achieved. This oxygen stream can be used for producing $CO_2$ of high purity - 85-99.94 vol% (Wetenhall et al., 2014) - through fuel combustion. The low energy requirement of the CLAS system, compared to the CASU and to the ADS, is due to the redox reactions (equations 1 and 2) which balances heat requirements and production. Furthermore, CLAS has the potential for heat integration with both the combustion process and the steam cycle.

Oxidation $Me_xO_{y-2}(s)+O_2(\text{in air}) \rightleftarrows Me_xO_y(s)$ eq (1)

Reduction $Me_xO_y(s) \rightleftarrows Me_xO_{y-2}(s)+O_2(g)$ eq (2)

The life cycle assessment methodology (LCA) is a comprehensive tool that can be used to compare the environmental burdens of alternative technologies and to identify process chains which mostly contribute to the total environmental burden of the technology under analysis. There are no current evaluations available on the environmental impacts of CLAS although few studies are available more generally on carbon capture and storage technologies (CCS). For example, Akai et al (Akai et al., 1997) analysed the greenhouse gas emissions of different pre- and post-combustion capture technologies based on physical and chemical absorption. Spath et al. (Spath and Mann, 2001) assessed the carbon emissions of a conventional power plant with post-combustion capture. Lombardi et al. (Lombardi, 2003) performed an exergetic life cycle assessment and a classical environmental life cycle assessment of a semi-closed gas turbine combined cycle with $CO_2$ reduction from the exhausts by means of amine solution chemical absorption and an integrated gasification combined cycle with $CO_2$ reduction from the synthesis gas by means of amine solution chemical absorption and $O_2/CO_2$ cycle. Van Gijlswijket al. (Van Gijlswijk et al., 2006) studied the post combustion capture in a natural gas combined cycle and a pulverized coal steam cycle plant, and pre-combustion capture in a coal gasification combined cycle plant and were alone in analysing other environmental impacts than the global warming potential. Viebahn et al. (Viebahn et al., 2007) explained how per unit of energy produced, CCS reduced $CO_2$ emissions from combustion but the emissions from the supply chain rose, due to the increased coal demand. Pehnt et al. (Pehnt and Henkel, 2009) presented a LCA of $CO_2$ capture and storage for several lignite power plant technologies, including post-combustion, pre-combustion and oxyfuel capture as well as subsequent pipeline transport and storage of the separated $CO_2$ in a depleted gas field. The results showed an increase in the cumulative energy demand and a substantial decrease in greenhouse gas (GHG) emissions for all $CO_2$ capture approaches in

comparison with power plants without CCS. Finally, Schreiber et al. (Schreiber et al., 2009) studied coal-based power plants with and without $CO_2$ capture via mono-ethanolamine (MEA) and assessed the comparison of their environmental effects based on the life cycle assessment methodology.

CLAS is in its early age of development and, as shown, its environmental potential, when integrated with fuel combustion, still needs to be assessed. Therefore, the purpose of this study is to perform an attributional life cycle assessment of electricity production through the chemical looping air separation technology and identify the hot spots of the process. Two different scenarios and related mass and energy balanced are considered. Furthermore, the CLAS for electricity production is compared to renewable and fossil alternatives. The global warming potential of other $CO_2$ post-combustion and oxyfuel technologies is also considered for comparison.

## 2. Life cycle assessment methodology

Life cycle assessment is one of the most developed and widely used environmental methodology for comparing alternative processes or services. Life cycle assessment systematically analyses the entire life cycle of goods and services from raw material extraction to the product final disposal, including manufacturing, transport, use, re-use, maintenance and recycling, i.e. all flows to and from nature are assessed under a 'cradle to grave' perspective (Baumann and Tillman, 2004). Moreover, it helps to determine the ''hot spots'' in the system, that are those activities that have the most significant environmental impact and should be improved as the first priority, thus enabling identification of more environmentally sustainable options (Clift, 2006).

The LCA methodology consists a four very distinct phases. In the goal and scope definition the purpose of the study is primarily defined but also the following points should be addressed: i) what political or technical decision will depend on the results of the study; ii) what are the system boundaries for the study iii) what is the basis for comparison between different alternatives (i.e. which is the functional unit). During the inventory phase a life-cycle model of the product of interest is built up and all the environmentally relevant inputs and outputs of the process are listed. The inputs and outputs of each unit operation in the model are quantified and identified as either resource use or emissions (emissions to soil, water and air). In the impact assessment phase, the energy and mass flows are translated into potential impacts (referred to as environmental indicators) to the environment. According to its mass flow each environmental intervention is transformed into an environmental burden through a common unit, specific for the environmental category. Normalization and weighting are also included in this phase. The last phase includes the analysis of the results and the assessment of the conclusions based on the points reported in the goal and scope definition.

In LCA, a multifunctional process is defined as an activity that fulfils more than one function, such as a waste management process dealing with waste and generating energy (Ekvall and Finnveden, 2001). It is then necessary to find a rational basis for allocating the environmental burdens between the functions. The problem of allocation in LCA has been the topic of much

debate (Clift et al., 2000; Heijungs and Guinée, 2007). The ISO standards (ISO 14040, 2006) recommend that the environmental benefits of recovered resources should be accounted for by broadening the system boundaries to include the avoided burdens of conventional production (Eriksson et al., 2007). This approach is applied in this study.

Following the methodological approach of Clift et al. (Clift et al., 2000) a distinction is made between Foreground and Background, considering the former as 'the set of processes whose selection or mode of operation is affected directly by decisions based on the study' and the latter as 'all other processes which interact with the Foreground, usually by supplying or receiving material or energy'. The burdens are evaluated under three categories (Clift et al., 2000): direct burdens, associated with the use phase of the process/service; indirect burdens, due to upstream and downstream processes (e.g. energy provision for electricity or diesel for transportation); and avoided burdens associated with products or services supplied by the process (e.g. energy or secondary material produced by the system).

When translating the inventory data to environmental impacts, two general approaches are available, the so-called mid-point or end-point (Clift, 2013). In this study the mid-point approach is used and inputs are expressed in terms of their contribution to a set of impact mid-point categories. The standard mid-point impacts used in this study are those defined by Guinée (2002) and reported in Table 1. The results are commented only for the most relevant categories but all data are reported in the supplementary information.

| **Impact categories** | **Impact Indicator** | **Acronym** | **Characterisation model** | **Units** |
|---|---|---|---|---|
| Climate change | Global warming potential | GWP | CML 2001 baseline (IPCC, 2007b) (Apr. 2015) | kg $CO_2$eq |
| Acidification | Acidification potential | AP | CML 2001 baseline (Hauschild and Wenzel, 1998) (Apr. 2015) | kg $SO_2$eq |
| Resources depletion (fossil) | Abiotic depletion Fossil | ADP fossil | CML 2001 baseline (Guinée, 2001) (Apr. 2015) | MJ |
| Resources depletion (element) | Abiotic depletion Element | ADP element | CML 2001 baseline (Guinée, 2001) (Apr. 2015) | kg Sb eq |
| Eutrophication | Eutrophication potential | EP | CML 2001 baseline (Hauschild and Wenzel, 1998) (Apr. 2015) | kg phosphate eq |
| Photochemical ozone formation | Photochemical ozone creation potential | POCP | CML 2001 baseline (Jenkin and Hayman, 1999) (Apr. 2015) | kg ethane eq |
| Ecotoxicity (freshwater) | Fresh water aquatic ecotoxicity potential | FAETP | USEtox model (Rosenbaum et al., 2008) (Apr. 2015) | kg DCB[1] eq |
| Ecotoxicity (terrestrial) | Terrestric ecotoxicity potential | TETP | USEtox model (Rosenbaum et al., 2008) | kg DCB eq |
| Ecotoxicity (marine) | Marine aquatic ecotoxicity | MAETP | USEtox model (Rosenbaum et al., | kg DCB eq |

| | | | | |
|---|---|---|---|---|
| | potential | | 2008) | |
| Human toxicity | Human toxicity potential | HTP | USEtox model (Rosenbaum et al., 2008) | kg DCB eq |
| Ozone depletion | Ozone layer depletion potential | ODP | CML 2001 baseline (World Meteorological Organization, 2010) (Apr. 2015) | kg $R11^2$ eq |

Table 1. Impact indicators (Thinkstep, 2015).

Currently more than thirty software packages exist to perform LCA analysis, with differing scope and capacity: some are specific for certain applications, while others have been directly developed by industrial organisations (Manfredi and Pant, 2011). In this study GaBi 6 has been used (Thinkstep, 2015). GaBi 6 contains databases developed by Thinkstep, it incorporates industry organisations' databases (e.g. Plastics Europe, Aluminium producers, etc.) and also regional and national databases (e.g. Ecoinvent, Japan database, US database, etc.).

## 3. Goal and scope definition and system boundary

Figure 1 shows the system boundary analysed in this study. The foreground includes the CLSA system with electricity production which is compared to conventional electricity production from renewable and fossil resources. The entire life cycle of the systems under analysis has been considered in the modelling approach. Indirect activities of energy, chemicals and water production, and recovery and final disposal of waste material are included in the background. In addition, avoided burdens are allocated to valuable substances production and recovery. However, avoided burdens are not allocated to the production of electricity as for the comparative analysis, the displaced system would have accounted the same for all the scenarios analysed. In fact, the results are reported according to a functional unit of 1 MJ of electricity produced. Datasets used in the analysis are country-specific and refer to the UK.

The power production systems considered in the study are as follows.

- From renewable sources:
    - Electricity from hydro power,
    - Electricity from wind power,
    - Electricity from nuclear,
    - Electricity from photovoltaic,
    - Electricity from biogas,
    - Electricity from biomass,
    - Electricity from waste.
- From fossil resources:
    - Electricity from chemical looping air separation (lignite),
    - Electricity from hard coal,

- Electricity from natural gas.

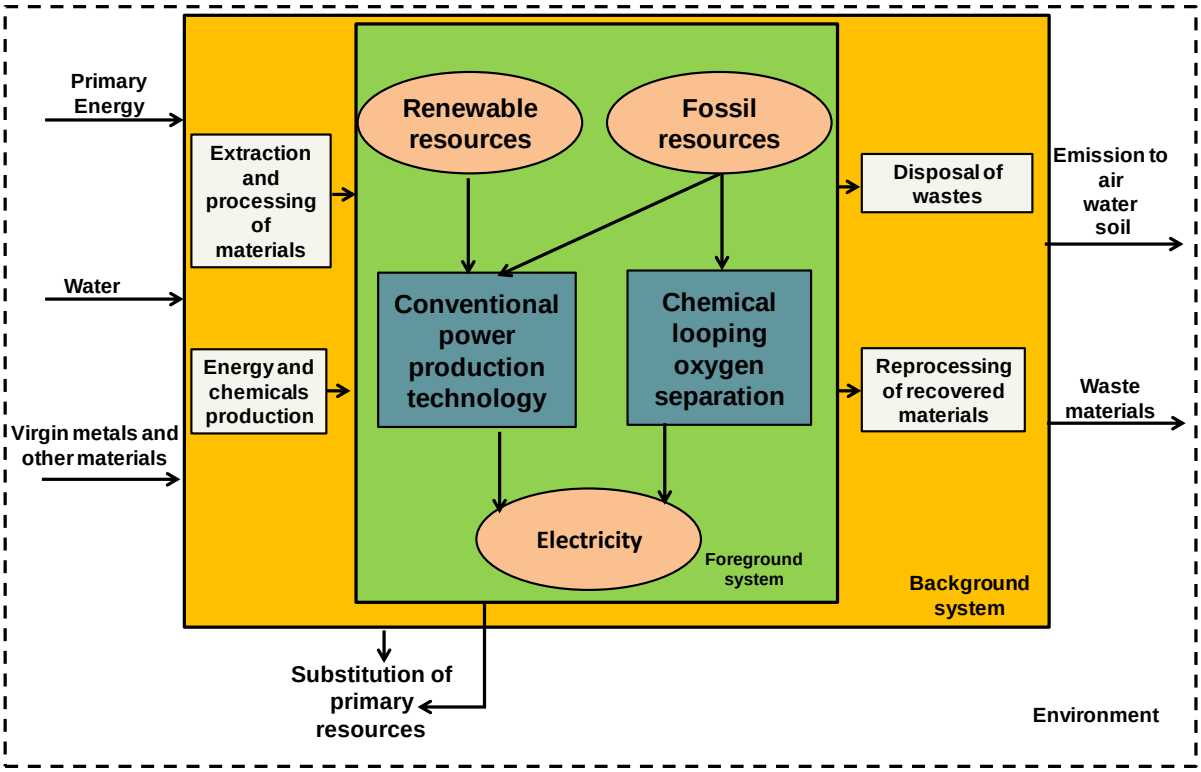



Figure 1. System boundary.

## 4. Life cycle inventory

### 4.1 Chemical looping air separation for electricity production

Figure 2 shows the flowsheets for the electricity production using the CLAS oxyfuel combustion system for two different scenarios as described below.

The chemical looping process is characterized by the cyclic reduction and oxidation of a solid material serving as an oxygen carrier and it usually consists of two interconnected fluidized beds, viz. the oxidizer and reducer (Moghtaderi, 2010). In the chemical looping process for air separation, by cycling the oxygen carrier material between the oxidizer and reducer, continuous oxygen production is achieved in the latter. In the environmental model, the amount of oxidised and reduced solid circulating between the chemical looping reactors (500 tonnes and 250 tonnes, respectively) is based on data reported in the literature (Lyngfelt and Leckner, 2015). To account for the production and disposal processes, the catalyst is assumed to be replaced three times a year (Francisco García-Labiano et al., 2007) and the plant to operate for 7446 h/year for 20 years. The thermodynamic properties of the oxygen carrier material modeled resemble those of SrFeO3 which was considered in the environmental assessment. The amount of Sr and Fe required for the oxidised and reduced solids are based on stechiometric relations. Iron ore and celestite (SrSO4) are used to produce the active material SrFeO3. The model for the primary production of celestite is based on the literature (Nuss and Eckelman, 2014) whereas model for iron production is based on the Gabi 6

database (Thinkstep, 2015). The active material of the oxygen carrier solid is supported by alumina ($Al_2O3$) whose production process is modelled according to the data reported in the Ecoinvent database (Swiss Centre for Life Cycle Inventories, 2014). The catalyst is disposed to landfill as no technology is currently available for recycling of the active material (U.S. Geological Survey, 2015) and its disposal does not create particular hazard (Agency for Toxic Substances and Disease Registry (ATSDR), 2016).

As the reducing reactor requires the introduction of a gas flow with a low oxygen partial pressure, the captured carbon dioxide is partially recycled to this reactor in addition to steam from the steam cycle. The steam cycle and the associated electricity production in a steam turbine are mainly driven by the heat recovered from the depleted air exiting the oxidising reactor. Electricity for export is recovered only from the steam cycle because the heat produced from fuel combustion is used in the reducing reactor, as the decomposition of $SrFeO3$ to $Sr_2Fe_2O5$ is endothermic, making the exothermic oxidation the major source of heat available to the steam cycle.

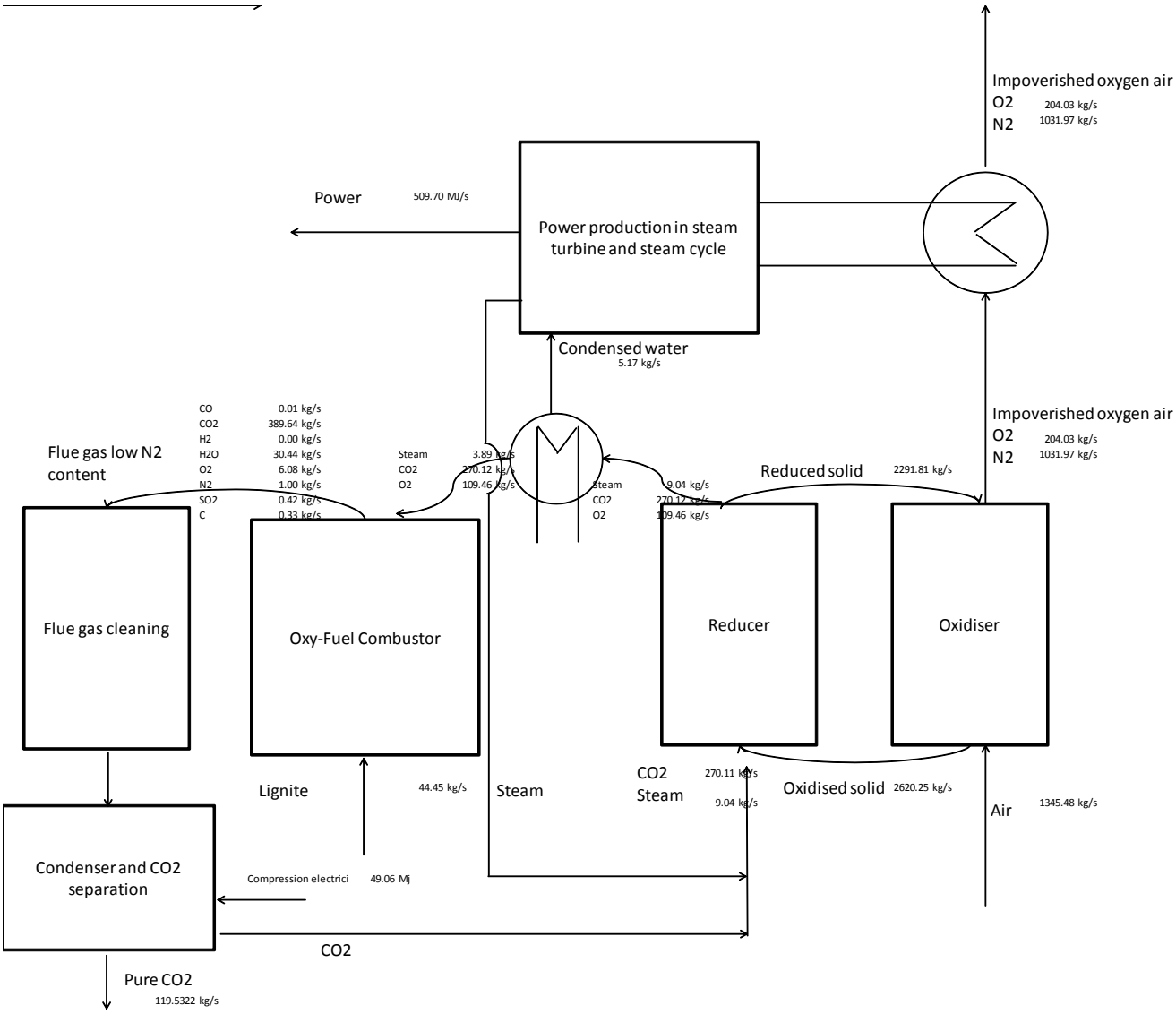
The oxy-fuel combustor requires a molar fraction of oxygen of around 0.35 when a dry recycle is used (in oxy-dry combustion water vapour is removed from the recycled flue gas) (Wall et al., 2009). The fuel chosen for oxy combustion and production of a pure $CO_2$ stream is lignite. This is representative also of other types of fuel such as hard coal and natural gas owing to a similar molar low heating value (LHV) - the amount of fuel required to stoichiometrically combust with one mol of oxygen. In fact, the molar LHV ranges from 352 kJ/mol $O_2$ to 422 kJ/mol $O_2$ for lignite (as received - not dried or processed) and natural gas, respectively. Consequently, the net electrical efficiency changes less than 2 percentage points using different types of fuel. The entire life cycle of lignite production has been modeled according to the Gabi database (Thinkstep, 2015). The data set considers the whole supply chain from lignite mining, lignite upgrading, transport and distribution.

The flue gas cleaning is based on the Ecoinvent model (Swiss Centre for Life Cycle Inventories, 2014) for power production from lignite (UCTE). This model has been adjusted to consider the flue gas cleaning process only, which includes wet scrubbers for SOx removal, a Selective Catalyst Reduction for NOx removal and removal and disposal of ashes and retentions.

In the carbon dioxide separation unit, a high purity carbon dioxide stream is produced; this is compressed to 150bar. The power requirement for compression taken from the literature is 0.114 kWh/kg of $CO_2$ (Fu and Gundersen, 2013). It is assumed that the pure carbon dioxide is sold to the market and this reduces the requirements of the carbon dioxide production from the conventional HABER-BOSCH process. Hence, avoided burdens are allocated to $CO_2$ production according to the Gabi database (Thinkstep, 2015).

To represent a wide range of possible operating conditions of the chemical looping reactors and to analyze the behavior of two thermodynamically identical oxygen carriers (one which does not require support material and one which does in order to exhibit the required mechanical strength and resistance against sintering), the two following scenarios have been modeled.

- Scenario 1 (S.1) is the base scenario and is further compared with the other energy production technologies. Figure 2 a) reports the process flow diagrams and the mass and energy balances for this scenario. Here, the model assumes that the solid in the dual circulating fluidized bed consists of active material only, i.e. no support material is added to the oxygen carrier material (Taylor et al., 2016). The net electrical efficiency, excluding the flue gas cleaning requirements, is 38.9 %.
- Scenario 2 (S.2). Figure 2 b) reports the process flow diagrams and the mass and energy balances for this scenario. It is assumed that the oxygen carrier material requires the addition of inert support material in order to achieve the appropriate mechanical strength and resistance against sintering. The ratio active material/(support material+active material) is 25% and this leads to endothermic operations even though the thermodynamic properties of the active phase are the same as S.1. As a consequence of adding inactive thermal mass to the solid in the CLAS unit, only around 70 % of the heat demand of the chemical looping air separation unit is met by the oxy combustor using the entire amount of oxygen produced by the CLAS unit itself. This means that the oxygen carrier material does not provide oxygen in sufficient quantity to allow for the combustion of the amount of fuel required to meet the entire heat demand of the CLAS unit. The overall net efficiency of 36.9% is therefore reduced by the additional external requirement of pure oxygen produced by a cryogenic air separation unit (CASU). This additional oxygen used in the oxy-combustor allows for the production of the remaining 30% of the heat demand of the chemical looping unit. For S.2, the production of oxygen from cryogenic air separation is considered according to the Gabi database (Thinkstep, 2015).


Impoverished oxygen air
O2 204.03 kg/s
N2 1031.97 kg/s
Power 509.70 MJ/s
Power production in steam turbine and steam cycle
Condensed water
5.17 kg/s
Impoverished oxygen air
O2 204.03 kg/s
N2 1031.97 kg/s
Flue gas low N2 content
CO 0.01 kg/s
CO2 389.64 kg/s
H2 0.00 kg/s
H2O 30.44 kg/s
O2 6.08 kg/s
N2 1.00 kg/s
SO2 0.42 kg/s
C 0.33 kg/s
Steam 3.89 kg/s
CO2 270.12 kg/s
O2 109.46 kg/s
Steam 9.04 kg/s
CO2 270.12 kg/s
O2 109.46 kg/s
Reduced solid 2291.81 kg/s
Flue gas cleaning
Oxy-Fuel Combustor
Reducer
Oxidiser
Lignite 44.45 kg/s
Steam
CO2 270.11 kg/s
Steam 9.04 kg/s
Oxidised solid 2620.25 kg/s
Air 1345.48 kg/s
Condenser and CO2 separation
Compression electrici 49.06 Mj
CO2
Pure CO2
119.5322 kg/s

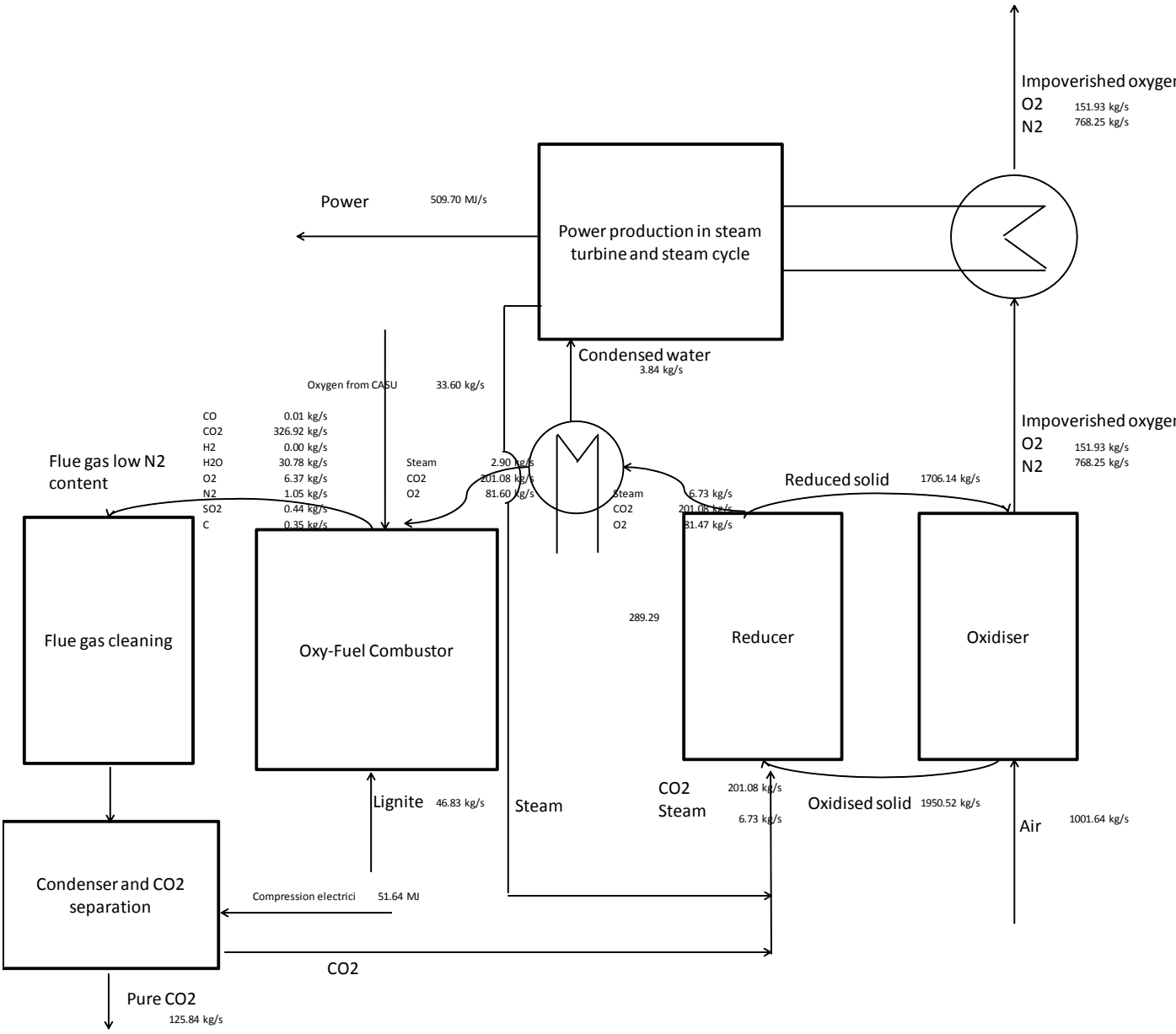



Figure 2. Process flow diagram and mass and energy balance for a) S.1 and b) S.2.

### 4.2 Conventional renewable energy technologies

These technologies are modelled according to the Gabi database (Thinkstep, 2015) and are all UK-country specific. The background system of all the following technologies includes the production of electricity and heat requirements as well as transport of material and production of refinery products.

#### 4.2.1 Electricity from hydro power

The foreground system includes construction, installation, operation, dismantling and removal of electrical parts of the hydro power plant. End-of-life of infrastructure like

concrete foundations or earth dams is not taken into consideration. Operational life time of the hydro power models are 60 years. Maintenance is included as well as the change of service material like oil for the generators. Greenhouse gas emissions (GHG) from biomass degradation in reservoirs are also included.

### 4.2.2 Electricity from wind power

The UK specific share of onshore and offshore electricity generation and annual full load hours are taken into account. The foreground system includes production, transportation, installation, operation, dismantling and removal of the wind turbines. Main components modeled are the rotor (spinner + three blades), the nacelle, the tower and foundation for the turbines. The wind converter system also includes the transformer station, the electrical gear, internal cables which connect the turbines to the transformer station and external cables which connect the wind park to the existing electricity grid. Losses in the cables and transformer station are included and calculated to approximately 5%.
The onshore model is based on a 300 MW wind park, operating 182 wind turbines with 1.65 MW each. The rotor blade length is 82m. The capacity factor is 40.8 (efficiency of the electricity generation in relation to a theoretical maximum of electricity generation). Operational life time of the wind turbines and cables is 20 years.
The offshore model is based on a 300 MW wind park, operating 100 wind turbines with 3.00 MW each. The rotor blade length is 90m. The capacity factor is 54.2. Operational life time of the wind turbines and cables is 20 years.
Maintenance is included as well as the change of service material like oil for the generator.

### 4.2.3 Electricity from nuclear power

The model comprises the production of electricity from a UK specific mix of pressurized water (PWR) and boiling water (BWR) reactors. It also includes the infrastructure of the power plant and the end-of-life of the auxiliary buildings, e.g. cooling tower. The following main steps are considered: uranium mining and milling, conversion of the uranium oxide into uranium hexafluoride, isotopes enrichment, fuel production, use of the uranium in reactor, end-of-life of the spent fuel and waste.

### 4.2.4 Electricity from photovoltaic

The manufacturing and operation life cycle phases of the photovoltaic technology are considered. End-of-life of the panels is not included since there is no common technology to reuse/ recycle them. Operational life times of the panels are modeled with 20 years. The photovoltaic model is based on the mix of different photovoltaic technologies installed. All technologies are modeled individually. The following average efficiencies per technology are used: Mono-Silicon 14.0 %, Multi-Silicon 13.2%, Cadmium-Telluride (CdTe) 9.0%, Amorphous-Silicon 5.5 %, Ribbon-Silicon 11.2 %, Copper-Indium-Gallium-Diselenide 11.0 %. A global average share of different Photovoltaic (PV) technologies is considered: Mono-Silicon 47.7 %, Multi-Silicon 38.3%, Cadmium-Telluride (CdTe) 6.4 %, Amorphous-Silicon 5.1 %, Ribbon-Silicon 1.5 %, and Copper-Indium-Gallium-Diselenide 1.0 %. It is assumed

that the assembly of the photovoltaic panels takes place in Germany but UK specific annual irradiation values are taken into account.
The system efficiencies are 75% for slanted roof installation and 80 % for ground mounted into consideration. The share of slanted roof installation is 90 %.

### 4.2.5 Electricity from biogas and biomass

The biomass (solid) and biogas supply chains consider the whole supply chain of the energy carrier from production, processing and transport of the fuels to the power plants. Flue-gas desulphurisation, NOx removal and de-dusting are also considered. The emissions of the power plants are based on a mix of national inventory reports and thermodynamics calculation as reported in the Gabi database (Thinkstep, 2015). Combustion residues from biomass solid fuel burning, such as gypsum, bottom ash or fly ash are assumed to be reused e.g. in construction work. Waste treatment for these substances is therefore not considered.

### 4.2.6 Electricity from waste

The share of the waste fractions and the elementary composition of the incinerated municipal solid waste (MSW) represent the average European waste treated thermally. The MSW composition does not include separate collection of certain waste fractions like paper or glass. The waste fraction share and the elementary composition of the MSW are reported in the Gabi database (Thinkstep, 2015). The model represents the generation of electricity in an incineration plant for the thermal treatment of municipal solid fitted with a grate and a steam generator. Dry flue gas cleaning and selective non-catalytic reduction (SNCR) for NOx-removal to meet the legal requirements are considered as well as the disposal of air pollution control residues and metal recycling from bottom ash. The NOx reducing agent, ammonia, is directly injected into the furnace and reacts with the NOx to nitrogen and water. The flue gas is conditioned, adsorbents added and filtered with fabric filters. Lime milk and small parts of hearth furnace coke are used as adsorbents; a part of the adsorbents is re-circulated. The fly ash together with the adsorbent is mixed together with the boiler ash. The produced bottom ash after metal recovery and ageing is re-used as construction material. The APC residues including boiler ash, filter cake and slurries are disposed in underground deposits salt mines.
Emission are modeled according to the mean emission values per cubic meter of cleaned flue gas published by the EU Commission (European Commission, 2006).

### 4.3 Conventional fossil energy technologies: electricity from hard coal and natural gas

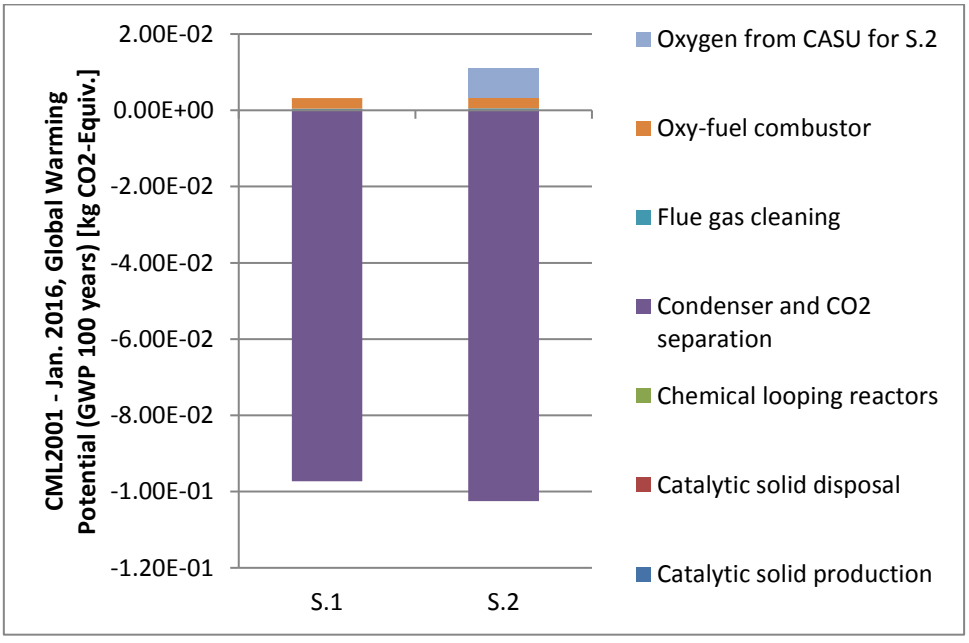
The coal and natural gas power plants are modeled as already described for the biomass power plant but emissions and efficiencies are specific for coal and natural gas, respectively and reported in the Gabi database (Thinkstep, 2015).

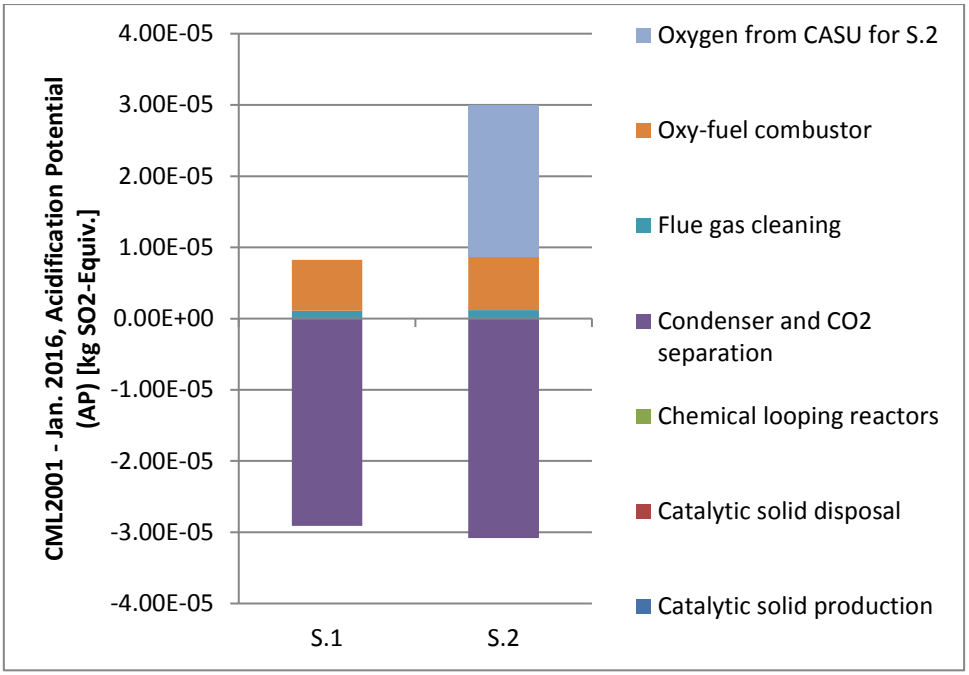
## 5. Results

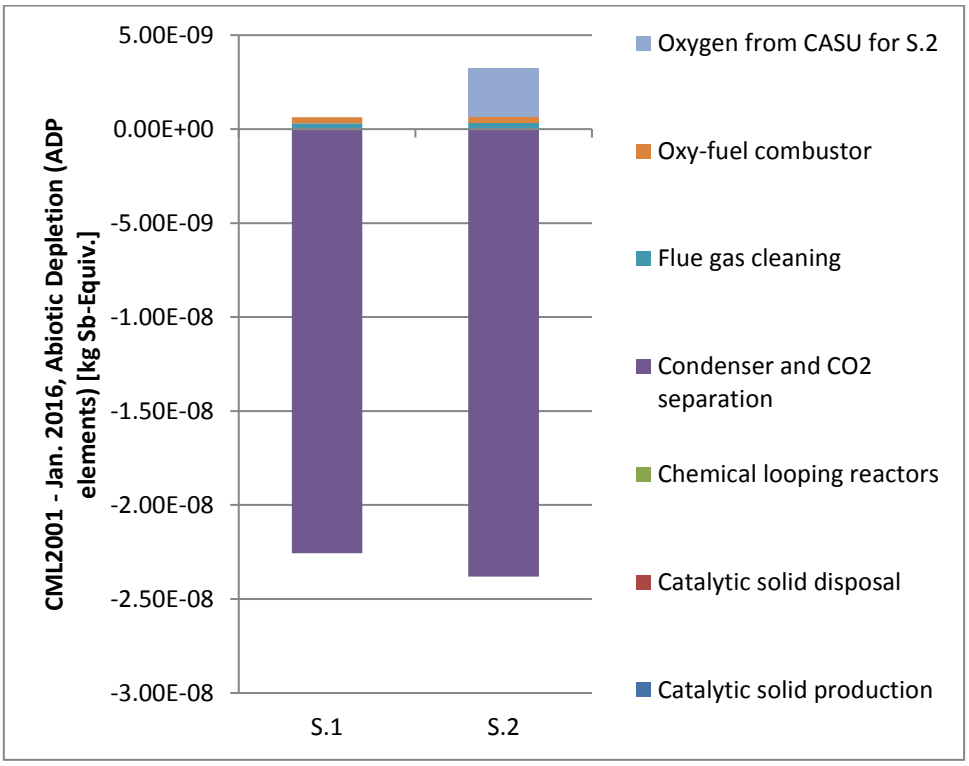
### 5.1 Electricity from CLAS – hot spot analysis

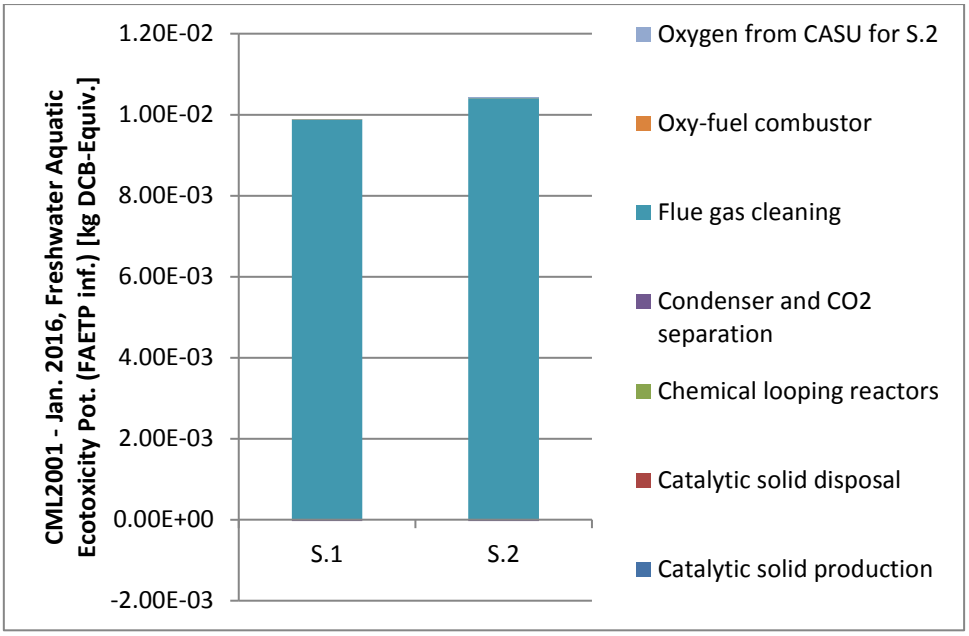
Figure 3 shows the hot spot analysis of S.1 and S.2 compared. Results are reported per functional unit (1 MJ of electricity produced) and no avoided burdens are allocated to electricity production as this would be the same for the two scenarios.

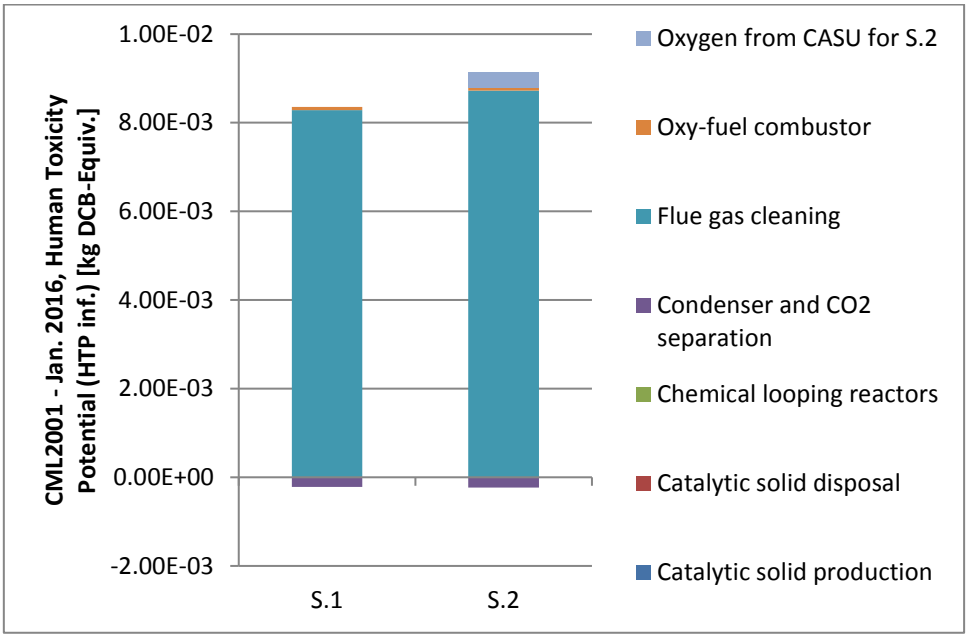
For both scenarios, the GWP is strongly affected by the avoided burdens associated to the avoided production of conventional $CO_2$ from fossil resources –for S.2 these are higher given the higher amount of pure $CO_2$ produced per unit of power. Furthermore, greenhouse gas emissions are associated to the mining and processing of the lignite used in the oxyfuel combustion but this accounts for only $2.56*10^{-3}$ kg of $CO_2$ eq in both scenarios. The penalty of external oxygen requirements for S.2 is shown as an increase in greenhouse gas emissions equivalent to $7.7*10^{-3}$ kg of $CO_2$ eq. A very similar trend is also shown for the AP; however, in this case the penalty associated to the external requirement of oxygen for S.2 reaches about 70% of the total positive contributions. The POCP and TEPT report very similar results to the GWP and AP and are therefore reported in the supplementary information. As opposed to what is reported in literature on the life cycle of production processes in which precious metal are used as catalysts (Tagliaferri et al., 2016), the production and disposal of the active (oxygen carrier) and support materials do not significantly increase the ADP element (Figure 3 c). A negligible impact is then associated to the oxygen carrier. The toxicities – fresh water, human (Figure 3 d) and e)) and marine (see the supplementary information) - and the ODP are almost entirely affected by the cleaning of the flue gas after the oxy combustor. This is especially due to the assumption adopted by the Ecoinvent database (Swiss Centre for Life Cycle Inventories, 2014): the lignite ash is disposed of as mine backfill and the recycled part is not inventoried. The disposal of the lignite ash to landfill affects the EP only marginally as also for this case the avoided burdens allocated to the production of the pure $CO_2$ have the biggest impact on the total ($-4.45*10^{-5}$ kg of phosphate eq.).




CML2001 - Jan. 2016, Acidification Potential (AP) [kg SO2-Equiv.]
4.00E-05
3.00E-05
2.00E-05
1.00E-05
0.00E+00
-1.00E-05
-2.00E-05
-3.00E-05
-4.00E-05
S.1
S.2
Oxygen from CASU for S.2
Oxy-fuel combustor
Flue gas cleaning
Condenser and CO2 separation
Chemical looping reactors
Catalytic solid disposal
Catalytic solid production


CML2001 - Jan. 2016, Abiotic Depletion (ADP elements) [kg Sb-Equiv.]
5.00E-09
0.00E+00
-5.00E-09
-1.00E-08
-1.50E-08
-2.00E-08
-2.50E-08
-3.00E-08
S.1
S.2
Oxygen from CASU for S.2
Oxy-fuel combustor
Flue gas cleaning
Condenser and CO2 separation
Chemical looping reactors
Catalytic solid disposal
Catalytic solid production


CML2001 - Jan. 2016, Freshwater Aquatic Ecotoxicity Pot. (FAETP inf.) [kg DCB-Equiv.]
1.20E-02
1.00E-02
8.00E-03
6.00E-03
4.00E-03
2.00E-03
0.00E+00
-2.00E-03
S.1
S.2
Oxygen from CASU for S.2
Oxy-fuel combustor
Flue gas cleaning
Condenser and CO2 separation
Chemical looping reactors
Catalytic solid disposal
Catalytic solid production


CML2001 - Jan. 2016, Human Toxicity Potential (HTP inf.) [kg DCB-Equiv.]
1.00E-02
8.00E-03
6.00E-03
4.00E-03
2.00E-03
0.00E+00
-2.00E-03
S.1
S.2
Oxygen from CASU for S.2
Oxy-fuel combustor
Flue gas cleaning
Condenser and CO2 separation
Chemical looping reactors
Catalytic solid disposal
Catalytic solid production

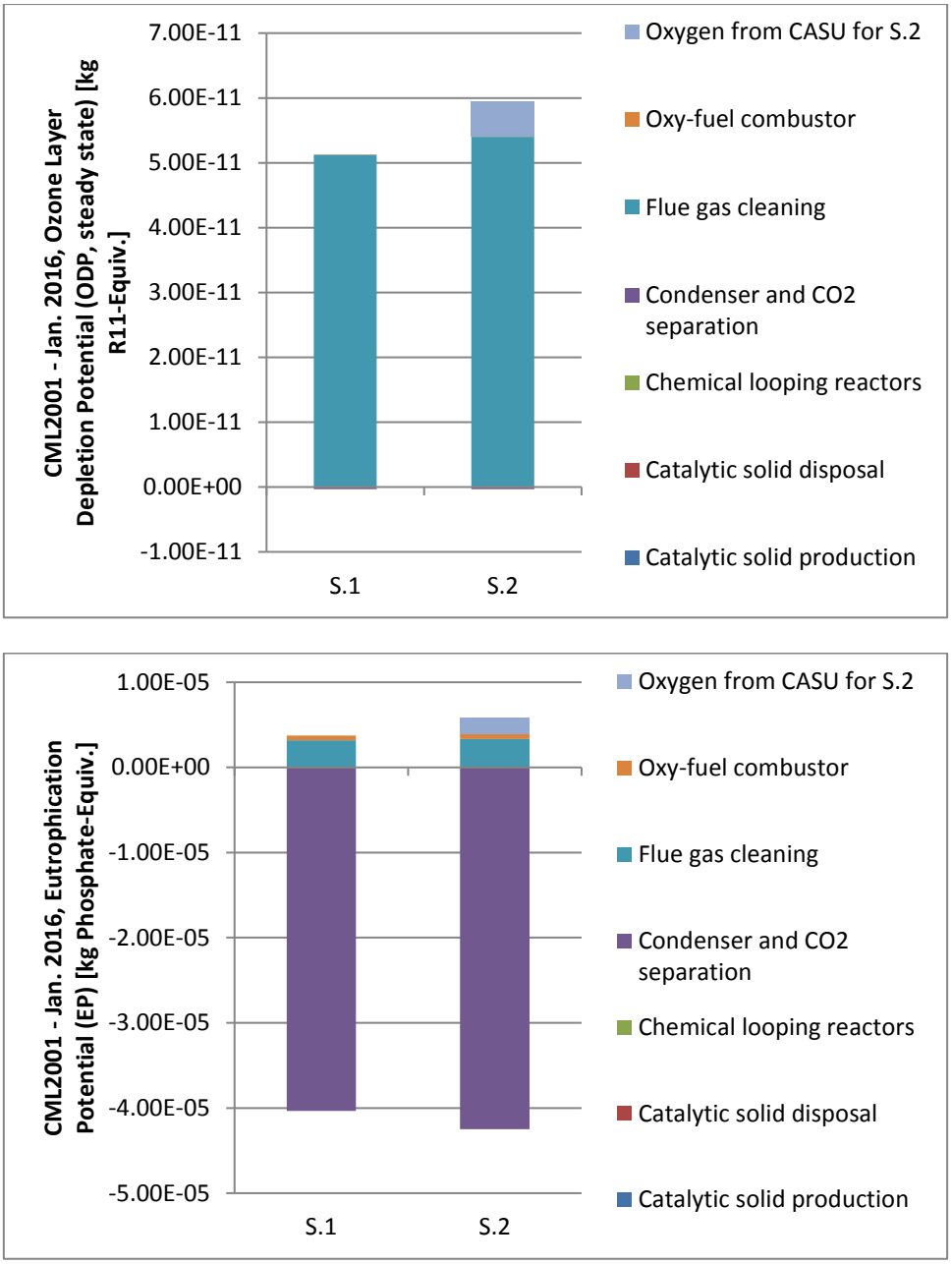



Figure 3. Hot spot analysis of the CLAS system (S.1). a) GWP. b) AP. c) ADP elementl. d) FAETP. e) HTP. f) ODP. g) EP.

Overall, a change in the ratio active material/support does not strongly affect the environmental results except for the case in which additional heat needs to be supplied through external oxygen requirements.

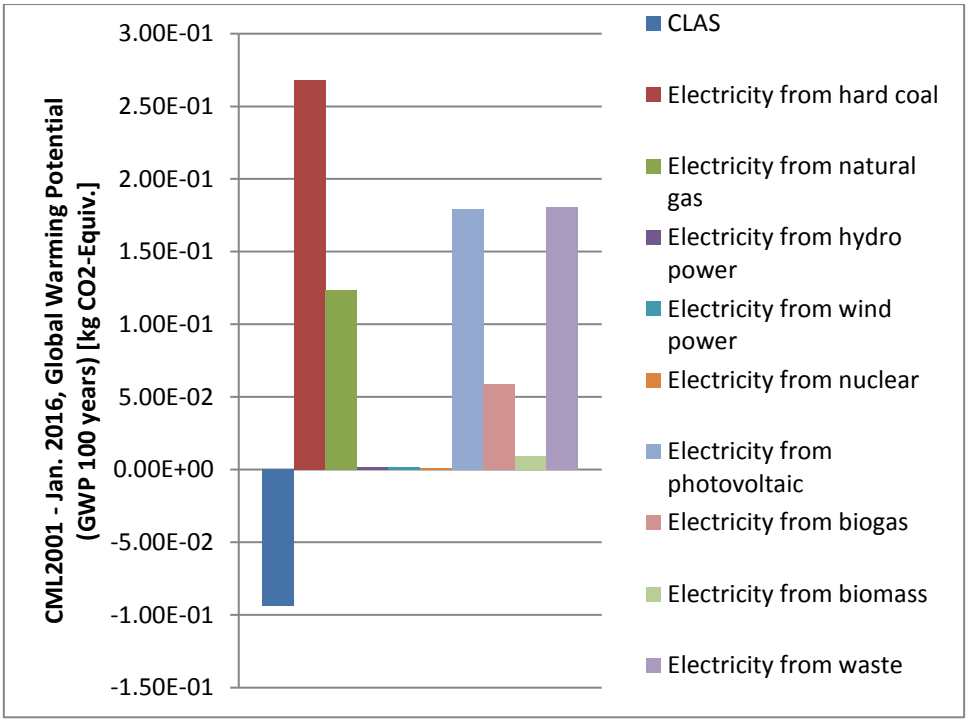
### 5.2 Comparison of electricity production technologies

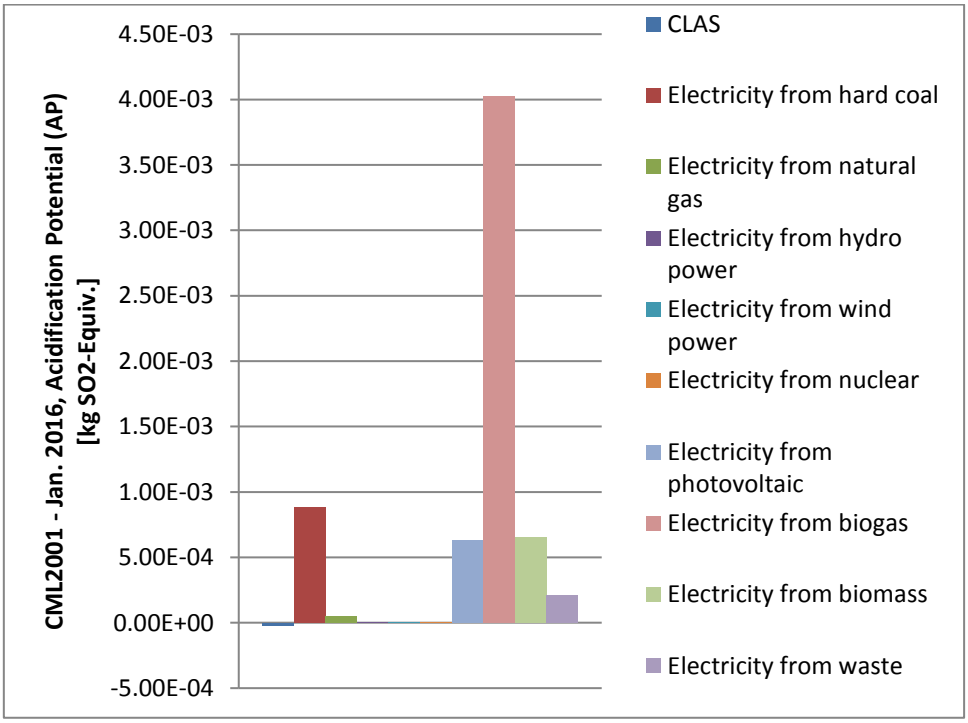
Figure 4 compares the results of S.1 to the burdens of the other energy technologies analysed in this study. Results are reported per functional unit (1 MJ of electricity produced) and no avoided burdens are allocated to electricity production to avoid double counting.

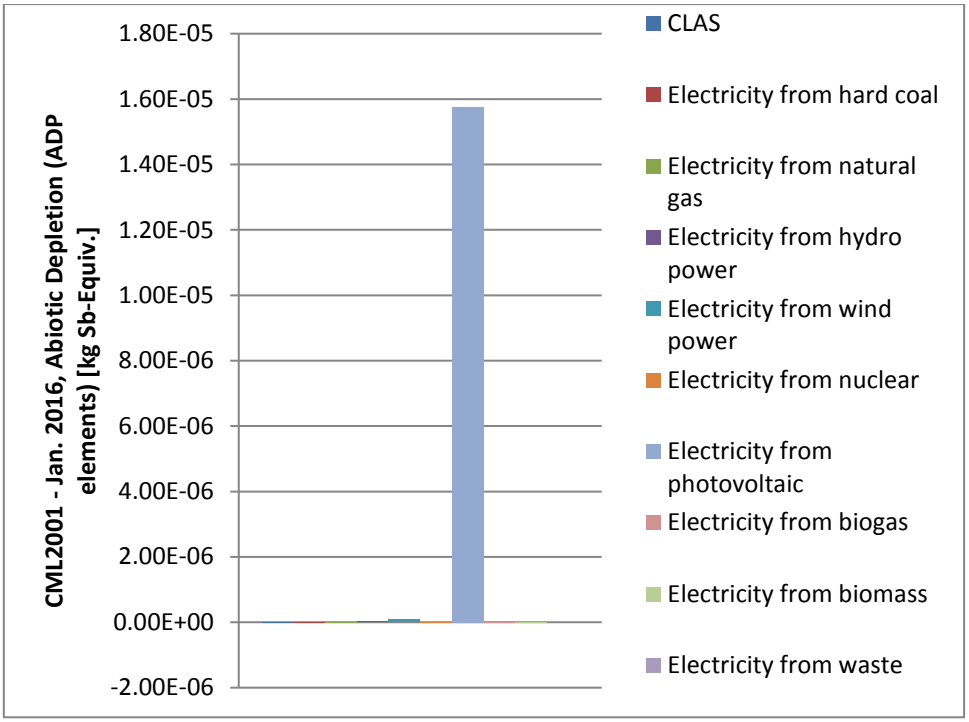
The production of electricity through CLAS shows the lowest (and the only negative) GWP thanks to the associated capture and use of a pure $CO_2$ stream (Figure 4 a)). Conversely, electricity production from fossil resources (hard coal and natural gas) as well as from waste shows the highest GWP impact which is due to fossil $CO_2$ emissions. The GWP of electricity production from photovoltaic is higher than that from natural gas ($1.79*10^{-1}$ vs $1.24*10^{-1}$ kg of $CO_2$ eq) and this is due to the production process of the photovoltaic systems. No direct $CO_2$ emissions are associated to hydro and wind power and the GWP of these technologies is mainly associated to the production processes of the systems. Figure 4 b) shows that the biogas fired power plants cause an AP of $4*10^{-3}$ kg $SO_2$ eq. compared to the $-6.9*10^{-5}$ kg of $SO_2$ eq of S.1. The AP of the biogas fired power plant is also higher than that of the hard coal power plant ($4*10^{-3}$ kg $SO_2$ eq. vs $8*10^{-4}$ kg $SO_2$ eq.) although $SO_2$ emissions per unit mass of fuel are lower than those for hard coal and biomass (Table 2) (Thinkstep, 2015) The higher AP is explained by the lower overall plant efficiency as reported in Table 2. To produce 1MJ of electricity more fuel is need in the case of biogas compared to the case of hard coal, biomass and natural gas and hence, the emissions per unit of power produced, are higher. The ADP element and the HTP of all scenarios become negligible when compared to the impacts of the photovoltaic technology and this is again associated to the production process of the solar panels. S.1 performs worst in the FAETP and ODP categories and as previous stated this is due to the assumption that lignite ash is disposed of as mine backfill. Figure 4 g) shows that the ADP fossil of S.1 is the lowest thanks to the avoided $CO_2$ production from conventional sources. However, if these avoided burdens had not been considered, the ADP fossil of S.1 would have been $7.7 *10^{-1}$ MJ. This is lower than the ADP fossil of electricity from hard coal, natural gas and photovoltaic (2.86 MJ, 2.11 MJ and 2.3 MJ, respectively) but higher than the ADP fossil of the electricity from renewable sources. Finally, the POCP and EP of power plants fed with waste, biomass, biogas and hard coal as well as the photovoltaic technology show the highest impact associated to emissions to air and the production process, respectively. The results of the TEPT are reported in the supplementary information.

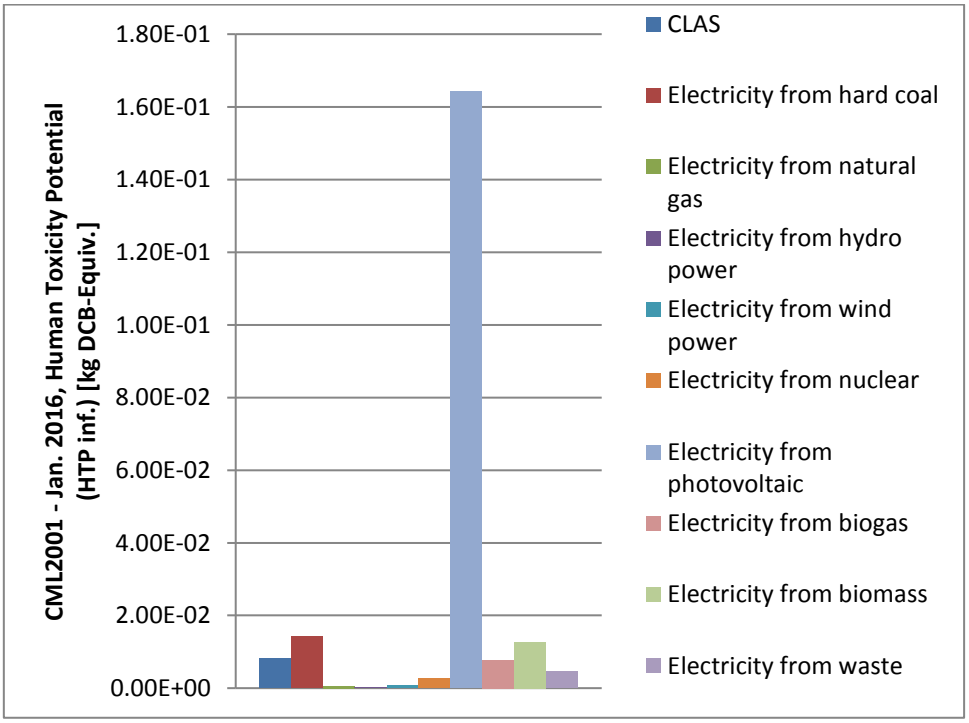
| **Energy carrier specific power plants [data are specific for UK]** | **Natural gas** | **Biogas** | **Biomass** | **Hard coal** |
|---|---|---|---|---|
| $CO_2$ [kg/TJ fuel input] | 56.4 | 101.2 | 101.11 | 87.9 |
| CO [kg/TJ fuel input] | 19 | 150 | 300 | 24.6 |
| $SO_2$ [kg/TJ fuel input] | 0.4 | 75.5 | 97.7 | 166.9 |
| NOx [kg/TJ fuel input] | 30.6 | 100 | 91.1 | 172.8 |
| Efficiency electricity plant [%] | 52 | 29.1 | 38.8 | 39 |

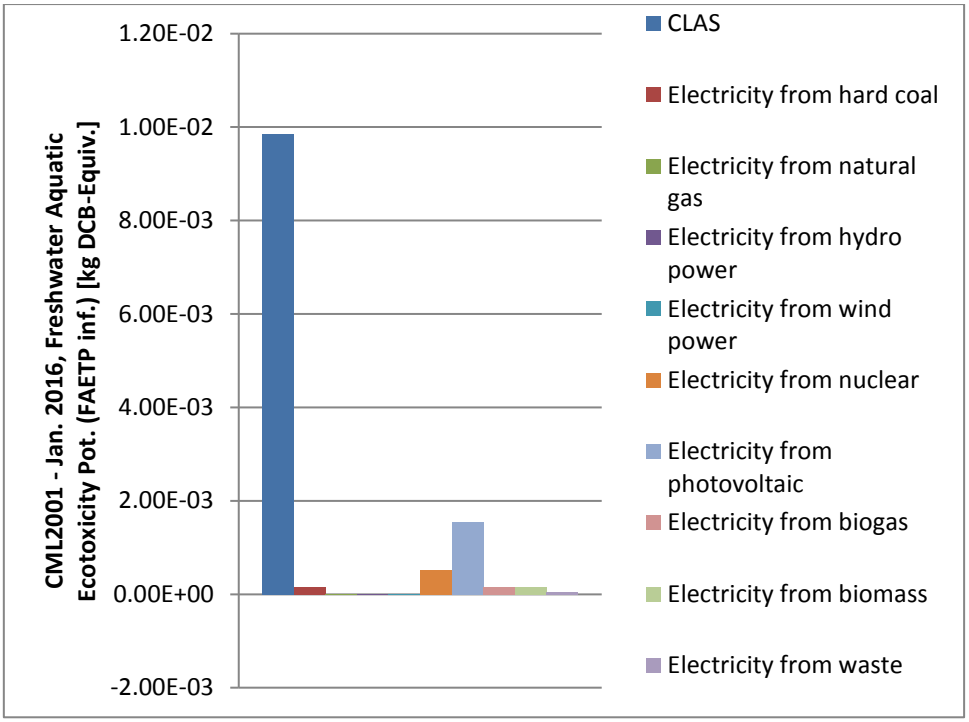
Table 2. Emission factors for power plants > 50 MW (Thinkstep, 2015).

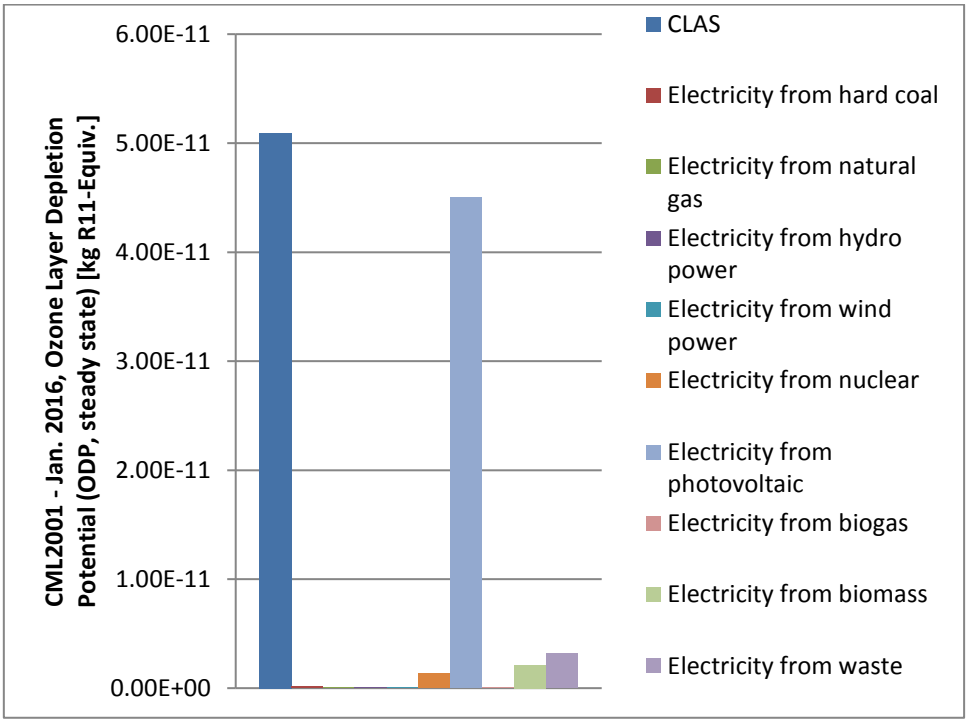

CML2001 - Jan. 2016, Global Warming Potential (GWP 100 years) [kg CO2-Equiv.]
3.00E-01
2.50E-01
2.00E-01
1.50E-01
1.00E-01
5.00E-02
0.00E+00
-5.00E-02
-1.00E-01
-1.50E-01
CLAS
Electricity from hard coal
Electricity from natural gas
Electricity from hydro power
Electricity from wind power
Electricity from nuclear
Electricity from photovoltaic
Electricity from biogas
Electricity from biomass
Electricity from waste

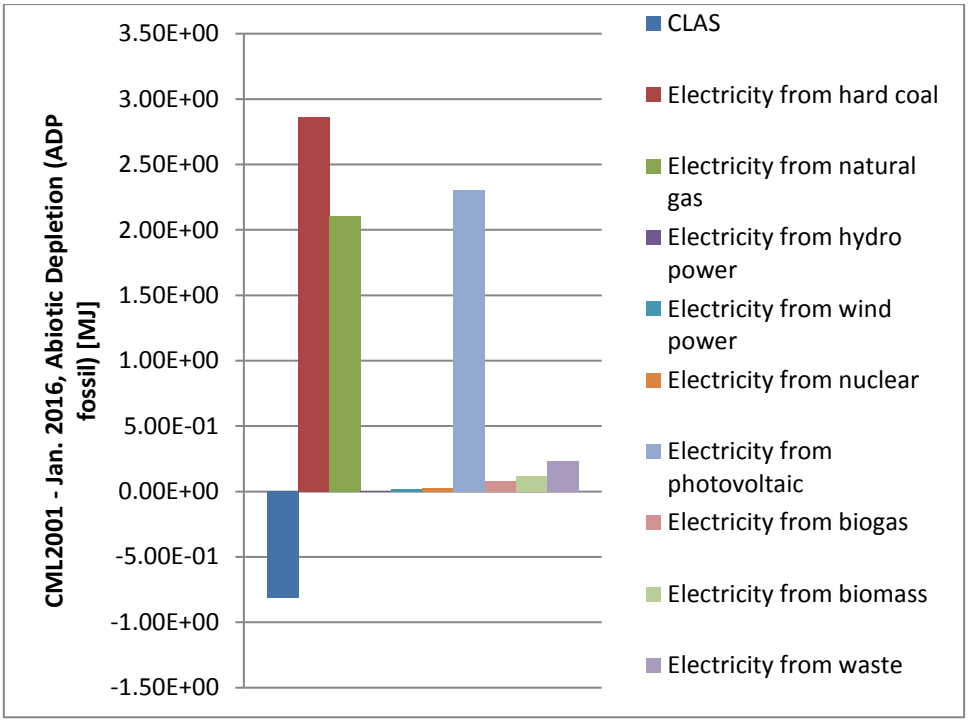

CML2001 - Jan. 2016, Acidification Potential (AP) [kg SO2-Equiv.]
4.50E-03
4.00E-03
3.50E-03
3.00E-03
2.50E-03
2.00E-03
1.50E-03
1.00E-03
5.00E-04
0.00E+00
-5.00E-04
CLAS
Electricity from hard coal
Electricity from natural gas
Electricity from hydro power
Electricity from wind power
Electricity from nuclear
Electricity from photovoltaic
Electricity from biogas
Electricity from biomass
Electricity from waste

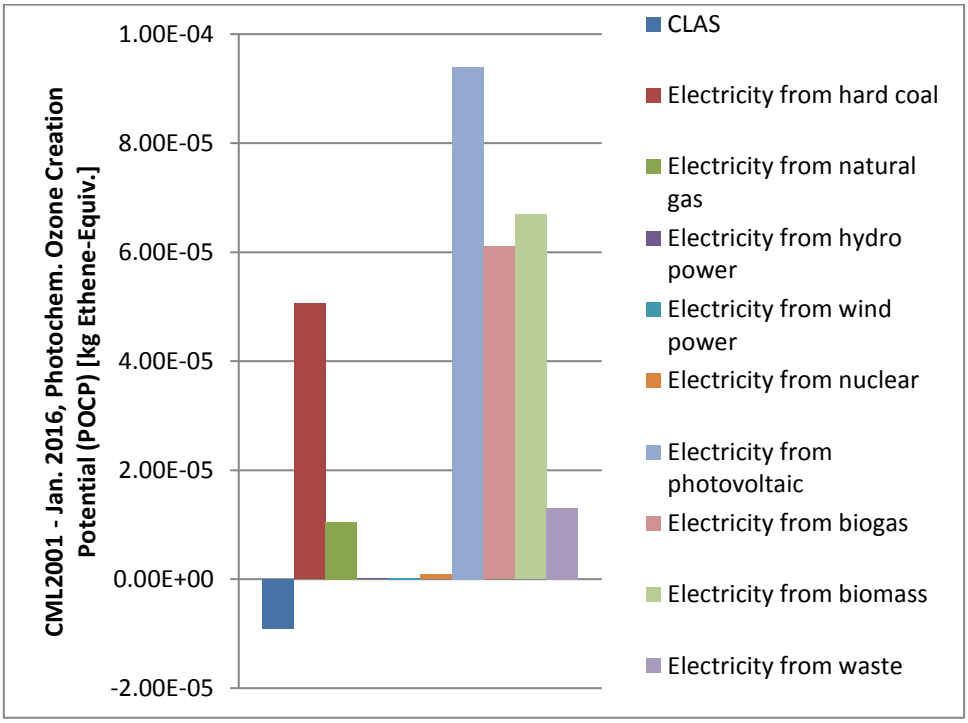

CML2001 - Jan. 2016, Abiotic Depletion (ADP elements) [kg Sb-Equiv.]
1.80E-05
1.60E-05
1.40E-05
1.20E-05
1.00E-05
8.00E-06
6.00E-06
4.00E-06
2.00E-06
0.00E+00
-2.00E-06
CLAS
Electricity from hard coal
Electricity from natural gas
Electricity from hydro power
Electricity from wind power
Electricity from nuclear
Electricity from photovoltaic
Electricity from biogas
Electricity from biomass
Electricity from waste


CML2001 - Jan. 2016, Human Toxicity Potential (HTP inf.) [kg DCB-Equiv.]
1.80E-01
1.60E-01
1.40E-01
1.20E-01
1.00E-01
8.00E-02
6.00E-02
4.00E-02
2.00E-02
0.00E+00
CLAS
Electricity from hard coal
Electricity from natural gas
Electricity from hydro power
Electricity from wind power
Electricity from nuclear
Electricity from photovoltaic
Electricity from biogas
Electricity from biomass
Electricity from waste


CML2001 - Jan. 2016, Freshwater Aquatic Ecotoxicity Pot. (FAETP inf.) [kg DCB-Equiv.]
1.20E-02
1.00E-02
8.00E-03
6.00E-03
4.00E-03
2.00E-03
0.00E+00
-2.00E-03
CLAS
Electricity from hard coal
Electricity from natural gas
Electricity from hydro power
Electricity from wind power
Electricity from nuclear
Electricity from photovoltaic
Electricity from biogas
Electricity from biomass
Electricity from waste


CML2001 - Jan. 2016, Ozone Layer Depletion Potential (ODP, steady state) [kg R11-Equiv.]
6.00E-11
5.00E-11
4.00E-11
3.00E-11
2.00E-11
1.00E-11
0.00E+00
CLAS
Electricity from hard coal
Electricity from natural gas
Electricity from hydro power
Electricity from wind power
Electricity from nuclear
Electricity from photovoltaic
Electricity from biogas
Electricity from biomass
Electricity from waste


CML2001 - Jan. 2016, Abiotic Depletion (ADP fossil) [MJ]
3.50E+00
3.00E+00
2.50E+00
2.00E+00
1.50E+00
1.00E+00
5.00E-01
0.00E+00
-5.00E-01
-1.00E+00
-1.50E+00
CLAS
Electricity from hard coal
Electricity from natural gas
Electricity from hydro power
Electricity from wind power
Electricity from nuclear
Electricity from photovoltaic
Electricity from biogas
Electricity from biomass
Electricity from waste


CML2001 - Jan. 2016, Photochem. Ozone Creation Potential (POCP) [kg Ethene-Equiv.]
1.00E-04
8.00E-05
6.00E-05
4.00E-05
2.00E-05
0.00E+00
-2.00E-05
CLAS
Electricity from hard coal
Electricity from natural gas
Electricity from hydro power
Electricity from wind power
Electricity from nuclear
Electricity from photovoltaic
Electricity from biogas
Electricity from biomass
Electricity from waste

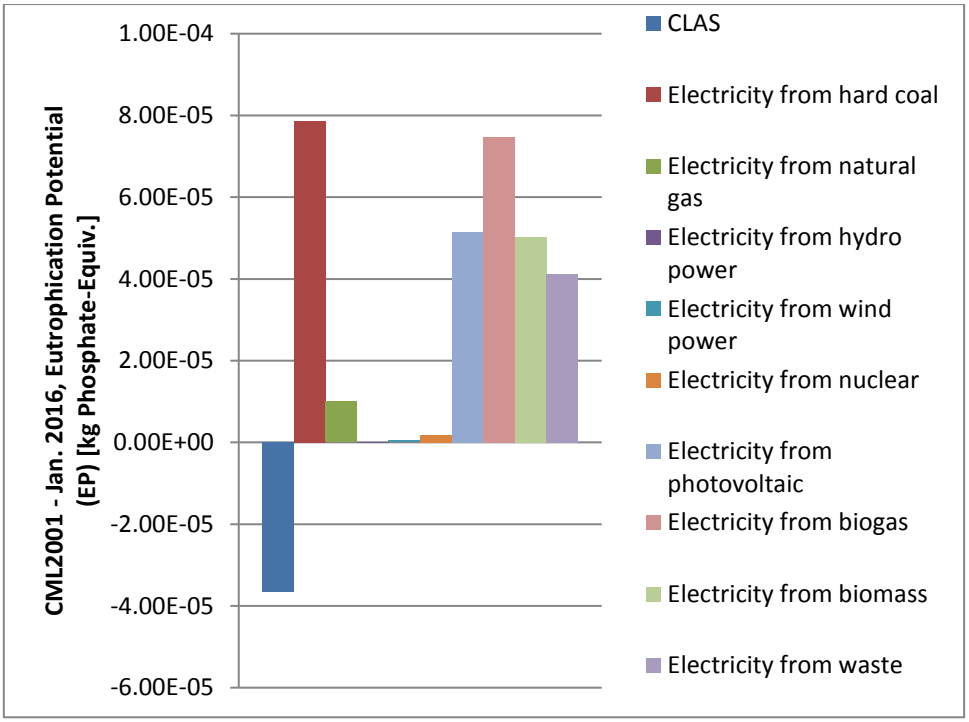



Figure 4. Comparison of alternative electricity production technologies. S.1 is considered for the CLAS technology. a) GWP. b) AP. c) ADP element. d) HTP. e) FAETP. g) ADP fossil.

Overall, electricity production through the CLAS system shows significantly reduced environmental burdens when compared to both fossil and renewable alternatives. Conversely, the environmental burdens of electricity production through photovoltaic systems are strongly limited by the production process of the solar panels.

## 6. Discussion

To facilitate the comparison across impact categories, normalized results have been analysed in Figure 5. The ADP element has not been considered (it is reported in the supplementary information) because the results were entirely dominated by the effect of the photovoltaic power production and the variation among the different indicators could not be appreciated. Figure 5 shows that the burden is equally distributed among the toxicities, the GWP, AP and ADP. Therefore, there is no one impact that should be addressed first by legislations but they are all equally important.

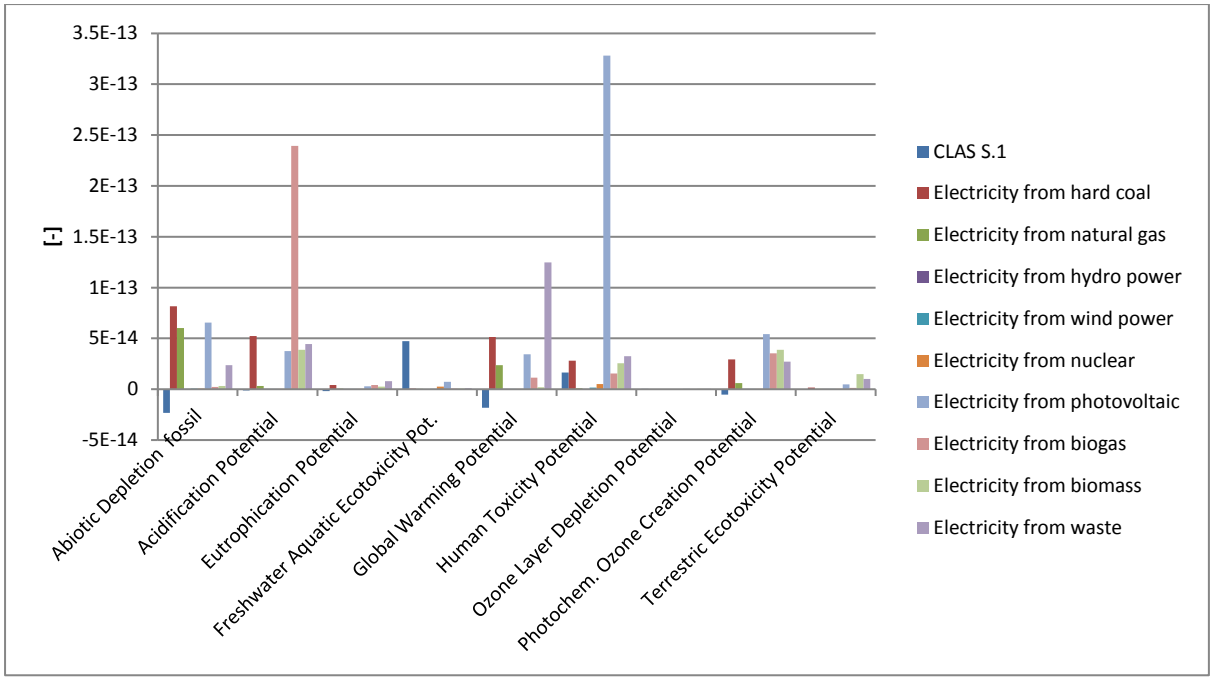



Figure 5. Normalised results. The normalisation is done based on CML, IPCC, ReCiPe (region equivalents), EU25+3, year 2000 (Thinkstep, 2015).

As previously shown, electricity production through chemical looping performed better than the other renewable and fossil energy technologies analysed. However, none of the technologies analysed consider carbon capture. Therefore, in Figure 6, the GWP of S.1 is compared to other electricity production technologies from lignite which consider carbon capture (and storage depending on the scenario). The data collected are based on the literature (Bauer et al., 2008; Marx et al., 2011, 2010; Pehnt and Henkel, 2009; Schreiber et al., 2009). Schreiber et al. (Schreiber et al., 2009) also consider past, present and future power plants and CCS systems up to 2020.

To directly compare the GWP of S.1 to the data found in the literature, the avoided burdens allocated to the utilization of the pure $CO_2$ stream produced are not considered in Figure 6. In addition, plants processing lignite only are analysed. Electricity production through CLAS is the best option, especially when compared to CCS. This is due to the energy penalty associated with the pre and post combustion technologies (circa 7%-18.2% increasecompared to the no capture case (Marx et al., 2011)). For the CCS cases, electricity requirements and hence, the GWP are even higher than the no CCS cases because of pumping before storage. Electricity production through CLAS can be more directly compared to oxyfuel combustion (using CASU) but also for this case Bauer et al. (Bauer et al., 2008) reports values about 4 times higher than the values of S.1. It is worth pointing out that all the data collected from the literature refer to a country specific situation (mainly Germany) and therefore, upstream and downstream processes and the analysed technologies do not match those modelled in this study. Furthermore, background data and assumption of literature studies are often of difficult interpretation.

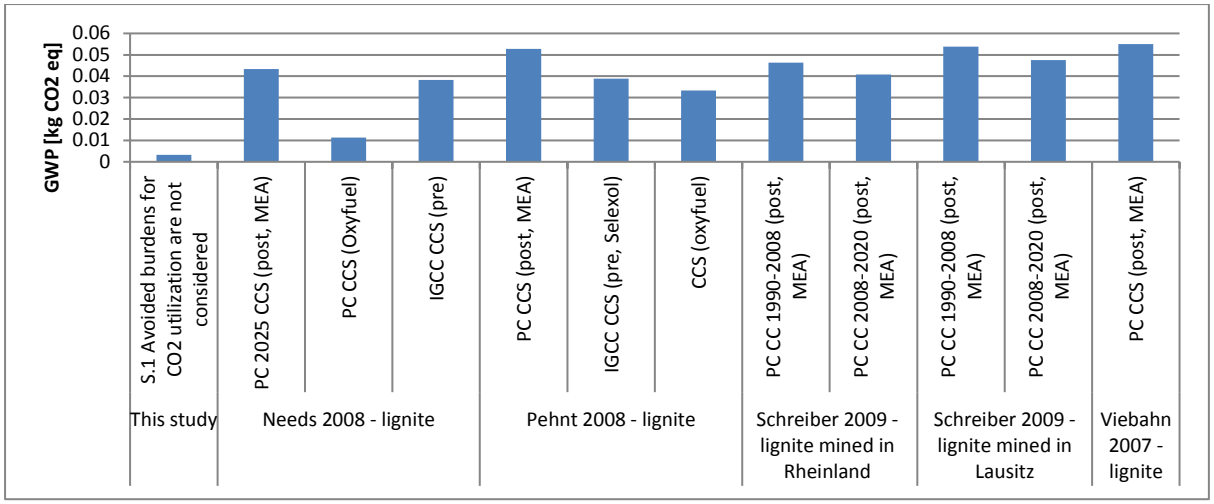



Figure 6. Comparison of S.1 with other capture technologies reported in literature (data is reported according to the functional unit). CC=carbon capture; CCS=carbon capture and storage; pre=pre combustion capture; post= post combustion capture; MEA=monoethanolamine; PC=pulverised coal technology; IGCC=integration gasification combined cycle.

Finally, the total world consumption and production of $CO_2$ must be considered when assuming that the pure $CO_2$ stream is sold to market (and avoided burdens are allocated to it). The potential supply of anthropogenic $CO_2$ is very much larger than potential demand (Global CCS Institute and PB Parsons Brickerhoff, 2011). The global $CO_2$ reuse market currently amounts to approximately 80 million tonnes/year, and is dominated by enhanced oil recovery (EOR) demand in North America. EOR accounts for approximately 50 million tonnes of demand annually, of which around 40 million tonnes is supplied annually from naturally occurring $CO_2$ reservoirs at prices generally in the order of US$15–19/tonne. It is estimated that globally around 500 million tonnes of low-cost (<US$20/tonne) high concentration $CO_2$ is available annually as a by-product from natural gas processing, fertiliser plants and some other industrial sources. At a much higher cost (US$50–100/tonne), around 18,000 million tonnes could also be captured annually from the dilute $CO_2$ streams currently emitted by power, steel and cement plants. Therefore, the utilization of captured $CO_2$ in novel power plant technologies might be strongly limited by the market. For this reason the results of Figures 2 and 3, without the allocation of avoided burdens to the $CO_2$ production from conventional sources (for S.1 and S.2), have been reported in the supplementary information for comparison. The GWP, ADP fossil, AP, POCP and EP are significantly affected as, for this case, the environmental burdens S.1 range between the renewable technologies (hydro, wind and nuclear) and the natural gas power plant.

## 7. Conclusions

This study assessed the environmental potential of electricity and $CO_2$ production through oxyfuel combustion of lignite. The pure oxygen required for combustion is produced through chemical looping air separation from which power is recovered.

First, an attributional LCA of this technology has assessed the operations which mainly contribute to the total environmental impacts for two different scenarios at the limits of the operating window (100% and 25% active material). For both scenarios, the GWP is strongly affected by the avoided burdens associated to the avoided production of conventional $CO_2$ from fossil resources and both GWPs result negative (which means a net gain for the environment). A very similar trend is also shown for the ADP, AP, POCP, EP and TEPT. Conversely, the flue gas cleaning process after the lignite combustion strongly increases the toxicity impacts of the system.

Furthermore, this study has analysed the potential of electricity and pure $CO_2$ production through CLAS when compared with conventional power production technologies from renewable and fossil alternatives, including electricity from hydro power, electricity from wind power, electricity from nuclear, electricity from photovoltaic, electricity from biogas, electricity from biomass, electricity from waste, electricity from hard coal and electricity from natural gas. Electricity production through CLAS showed the lowest (and the only negative) GWP, again thanks to the associated capture and use of the pure $CO_2$ stream. Electricity production from fossil resources (hard coal and natural gas) as well as from waste showed the highest GWP impact due to fossil $CO_2$ emissions. The lowest GWP of hydro and wind were associated with the lack of direct $CO_2$ emissions during operation. The AP was dominated by the electricity production from biogas fired power plants because of their low energy efficiency and higher SOx emissions. Furthermore, it is worth pointing out that the electricity production from photovoltaic consistently performed worse than the other renewable power production technologies, especially for the ADP fossil, ADP element, HTP, ODP, and POCP because of the impacts due to the solar panels production process. As previously stated in the hot spot analysis, the environmental burden of electricity production from CLAS for the FAETP and ODP was limited in the comparative analysis, because of the contribution of the flue gas cleaning. This shows that the use of alternative fuel for oxyfuel combustion, which could decrease the burden of the flue gas cleaning, needs to be further investigated.

Finally, as shown in the discussion section, to evaluate the environmental burden of electricity production through the CLAS technology it is crucial to identify how the use of the $CO_2$ pure stream produced could be limited by the excess of other low cost $CO_2$ resources available on the market.

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