Life cycle assessment-based guidance for development of new energy technologies

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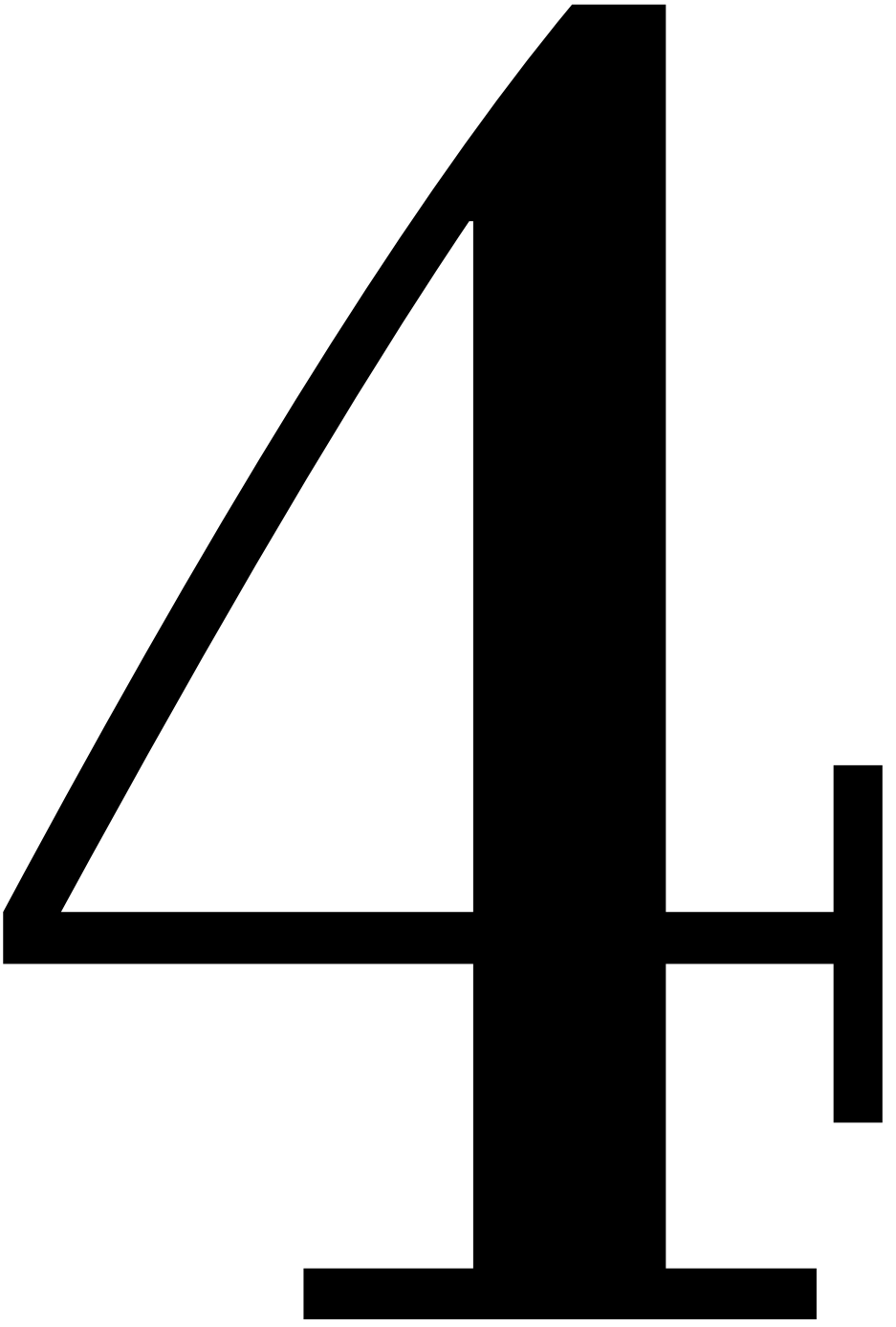

Chapter 4

A Life Cycle Assessment Case Study of Coal-fired Electricity Generation with Humidity Swing Direct Air Capture of CO_2 versus MEA-based Post-combustion Capture

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Abstract

Most carbon capture and storage (CCS) envisions capturing CO_2 from flue gas. Direct air capture (DAC) of CO_2 has hitherto been deemed unviable because of the higher energy associated with capture at low atmospheric concentrations. We present a Life Cycle Assessment of coal-fired electricity generation that compares mono-ethanolamine (MEA) based post-combustion-capture (PCC) of CO_2 with distributed, humidity-swing-based DAC (HS-DAC). Given suitable temperature, humidity, wind, and water availability, HS-DAC can be largely passive. Comparing energy requirements of HS-DAC and MEA-PCC, we find the parasitic load of HS-DAC is less than twice that of MEA-PCC (60-72 kJ/mol versus 33-46 kJ/mol, respectively). We also compare other environmental impacts as a function of net greenhouse gas (GHG) mitigation: To achieve the same 73% mitigation as MEA-PCC, HS-DAC would increase 9 other environmental impacts by on average 38%, whereas MEA-PCC would increase them by 31%. Powering distributed HS-DAC with photovoltaics (instead of coal) while including re-capture of all background GHG, reduces this increase to 18%, hypothetically enabling coal-based electricity with net-zero life-cycle GHG. We conclude that, in suitable geographies, HS-DAC can complement MEA-PCC to enable CO_2 capture independent of time and location of emissions and re-capture background GHG from fossil-based electricity beyond flue stack emissions.

4.1 Introduction

To keep global warming below 1.5-2°C targets, a balanced mix of greenhouse gas (GHG) mitigation measures is needed. Among these, carbon capture and storage (CCS) is projected to play an important role, as are increasing energy efficiency and deployment of renewable and nuclear power.(International Energy Agency 2012, 2008; Fuss et al. 2014) Besides traditional carbon capture from point sources, additional research into a wider portfolio of carbon dioxide removal (CDR) technologies, including distributed solutions, is seen as more and more important.(National Research Council 2015; Williamson 2016; Lackner et al. 2012) We henceforth refer to all such capture and storage options as CCS.

Relative (dis-) advantages of different CCS technologies

CCS is not tied to a specific technology. For coal-fired electricity generation, arguably the most mature technology is post combustion capture (PCC) using mono-ethanolamine (MEA), which scrubs CO_2 directly from the flue gas of fossil fuel power plants.(Rochelle et al. 2011; Abu Zahra 2009; Leung et al. 2014) It is often argued that there are no technological obstacles to large-scale implementation of PCC; rather that its main hurdle is a lack of policy or market mechanisms to incentivize adopting PCC amidst several challenges:(International Energy Agency 2012; Leung et al. 2014) The power plant must be located in sufficient vicinity of a suitable storage site, or else CO_2 transport from the plant to such a site will become prohibitively expensive. PCC results in reducing operating efficiency compared to a conventional power plant. It can best be implemented in modern coal-fired power plants, i.e. fewer than 29% of currently installed coal-fired power plants globally.(International Energy Agency 2012; Leung et al. 2014) Further, as an optimum design choice, MEA-PCC typically captures 85 - 90% of the flue stack CO_2 emissions (Reiter and Lindorfer 2015) but none of the so-called background emissions (e.g., from mining and transporting coal, or building and decommissioning the plant itself). MEA-PCC thus typically eliminates only ~70% of total life cycle greenhouse gas (GHG) emissions of coal-based electricity (Corsten et al. 2013; Sathre et al. 2012). Finally, many power plants are located near population centres, a fact that can be problematic for transport and storage of CO_2. The IEA notes that "examples in Germany and The Netherlands illustrate that under-appreciation of public concerns over CO_2 storage can easily be fatal for CCS".(International Energy Agency 2012)

CO_2 does not necessarily have to be captured at the point of combustion. A CCS alternative to PCC is Direct Air Capture (DAC), a form of CDR that captures CO_2 directly from the atmosphere.(Tavoni and Socolow 2013; Lackner et al. 2012; Keith 2009) Relative dis-advantages and advantages of DAC versus PCC have been previously discussed (American Physical Society 2011; Reiter and Lindorfer 2015). One possible

advantage of DAC is that its location and schedule of operation is not necessarily tied to the emitting power plant itself. This flexibility imbues the technology with benefits (Boot-Handford et al. 2014; American Physical Society 2011; Wang et al. 2011) that may partially alleviate above limitations of PCC-based CCS: With regard to location, DAC plants can be built at or near the final CO_2 storage site, so that CO_2 transport infrastructure can be largely avoided. This could alleviate social opposition arising from CO_2 transport through populated areas (The Royal Society 2009). And because its unit size and operating schedule is not tied to the power plant itself, DAC could be scaled to capture not 90% but 100% of CO_2 emissions emitted from a plant and in addition compensate for background GHG emissions, thus allowing for true, net-zero GHG electricity from coal. Similarly, DAC could compensate for emissions from non-stationary GHG sources in sectors where costs or technological feasibility may prohibit phasing out fossil fuels (e.g. aviation). The Royal Society notes that 'there is surprising value in the economic freedom to build a capture plant where it is cheapest to do so and near the best sequestration site' (The Royal Society 2009).

Above theoretical advantages notwithstanding, capturing low concentrations of CO_2 from air simply is thermodynamically more challenging than from concentrated point sources (American Physical Society 2011). In a recent, comprehensive review of DAC and other CDR, DAC is expected to consume up to 45GJ per ton carbon equivalent (540kJ/mol CO_2) – levels that would arguably render DAC economically unviable (Smith et al. 2016). Whether as part of a portfolio of climate responses (National Research Council 2015) or as a last-resort backstop technology to reduce atmospheric CO_2 levels via negative emissions (Pielke 2009), improved understanding of DAC's energy requirement and environmental impacts is therefore crucial.

Humidity-swing DAC (HS-DAC)

Previous research on DAC has focused on gas scrubbing technologies using sorbents such as $CaOH_2$, KOH and NaOH (American Physical Society 2011; Zenz House et al. 2011; Baciocchi et al. 2006). The binding potential of these is relatively high, and therefore large amounts of high-exergy energy (electricity and/or high grade heat) are required to release the CO_2 during sorbent regeneration (Zenz House et al. 2011; Goeppert et al. 2012). In contrast, HS-DAC utilizes ambient-temperature heat for the sorbent regeneration: It is realized through water drying off the sorbent, i.e. spontaneously evaporating at ambient conditions (driven by the surrounding air and, if available, sun shine).(Lackner 2013) This is akin to other DAC technologies, such as temperature-swing DAC, which can use low grade heat for cycling the sorbent material and whose energy balance analysis thus also shows limited requirement for mechanical (or electrical) energy (Wurzbacher et al. 2011, 2012). However, for HS-

DAC, the relatively low energy consumption of the overall capture process is in part achieved through reliance on ambient wind conditions to drive air through filters and later dry them for regeneration. This causes one of HS-DAC's main disadvantages: Its performance becomes weather/geography dependent (Goldberg et al. 2013). In this study, we quantify this dependence via sensitivity analyses.

Previously, relative (dis-)advantages of HS-DAC have been analysed, (Lackner et al. 2012) current understanding of its molecular process (Wang et al. 2013) and thermodynamics (Lackner 2013) described, and its energy consumption has been estimated at 50kJ/mol (Lackner 2009). Briefly, a non-toxic anionic exchange resin such as the commercially available MarathonA adsorbs CO_2 when dry and releases it when exposed to 100% humidity. Driven by ~2m/sec or higher ambient wind, air flows through filters in which the resin adsorbs some CO_2, reducing the air's CO_2 concentration from 400ppm to ~360 ppm (design-dependent). Using an electrically powered conveyor system, the filters enter a desorption chamber. After evacuating to 0,01 bar the sorbent is sprayed with water, causing the sorbent to release CO_2. The resulting low-pressure gas mix in the desorption chamber (CO_2 and H_2O, <=1% N2 and O2) is pumped through a cold trap (operating at variable temperature down to -10°C; see SI) where the H_2O in the gas mix condenses and is removed. The CO_2 is compressed to 100bar for underground injection. The filters are moved from the desorption chamber back outside where the remaining water dries off, thus regenerating filters for a new cycle.

Note that HS-DAC is unique primarily in the mechanism that cycles the sorbent's equilibrium binding characteristics back and forth between favouring adsorption and desorption. Following desorption into a vacuum, HS-DAC is similar to temperature-swing DAC. Both technologies have to include a step to remove the water vapour that comes off the sorbent along with the CO_2 from the desorption stream. Either, water vapour in the desorption stream is condensed out via cooling (Wurzbacher et al. 2011), followed by compression of CO_2 to the desired final pressure (typically 100 bar). Or the water vapour and the CO_2 are compressed concurrently such that the water gradually condenses as pressure increases, leaving mostly CO_2 (Lackner 2009). Different implementations of this CO_2 purification step are possible, including having a CO_2-enriched humid stream of air from a DAC machine purified via a conventional PCC-type, e.g. amine-based process. Different implementations will result in different water and energy consumptions. In this work, we assume a simple evacuation and drying via cooling implementation (Wurzbacher et al. 2011), which avoids the need for any specialized or costly hardware components. While not necessarily the most energy-efficient, the chosen implementation offers the advantage that its energy requirement can be robustly quantified as previously described (Lackner 2009). Here, we refine these

previous calculations based on more recent laboratory experiments on the sorbent's temperature and humidity dependence of the binding characteristics (Wang et al. 2011) as well its binding kinetics (Wang et al. 2016). This allows us to adjust the yield of the sorbent in response to varying weather conditions and thus quantify HS-DAC's sensitivity to suitable climate/geographies (see section 4.3).

Energy balance and life cycle environmental impacts

The goal of any CCS technology is to reduce anthropogenic CO_2 emissions. However, it is important to ensure that no prohibitive problem shifting occurs, because any form of CCS will cause increased energy and material use, with their own associated environmental impacts beyond GHG. A key aspect here is the previously identified inherent disadvantage of DAC over PCC which results from the more dilute CO_2 source and thus a priori higher parasitic energy losses: This underlines the importance of detailed energy balance studies and their system level effects on energy use per net GHG avoided (rather than per CO_2 captured) (American Physical Society 2011). While HS-DAC differs from PCC in other aspects as well, including hardware, consumable materials, and logistics of CO_2 transport, such background processes have much less impacts on the life cycle carbon balance and other environmental impacts (Results).

While detailed lifecycle assessments are available for PCC (Corsten et al. 2013) and environmental risks and constraints of various CDR technologies have been widely discussed (Williamson 2016; Smith et al. 2016; Reiter and Lindorfer 2015; von der Assen et al. 2016), a detailed life cycle assessment of HS-DAC has not been available to date. Thus, assessments of HS-DAC's relative performance vis-a-vis PCC are incomplete. In this work, we therefore perform a life cycle assessment (LCA) on HS-DAC to answer the following question: How do the lifecycle environmental impacts of HS-DAC compare with those of MEA-PCC when applied to CCS for coal-fired power generation?

4.2 Method and approach

LCA of emerging technologies

We used LCA to calculate environmental impacts, using CMLCA software (Institute of Environmental Sciences) with the ecoinvent 2.2 database (Swiss Centre for Life Cycle Inventories 2010) and the 10 CML-IA version 4.5 (baseline) impact characterization factors (CML-Department of Industrial Ecology 2015) recommended by the Handbook of LCA (Guinée et al. 2002) and commonly used in LCA of electricity generation (Koornneef et al. 2008; Corsten et al. 2013). The LCA thus covers climate change (measured as CO_2-eq on a 100a horizon, henceforth "GHG emissions"), eutrophication, resource depletion, acidification, summer smog, terrestrial ecotoxicity, marine

ecotoxicity, freshwater ecotoxicity, ozone depletion, and human toxicity. Additionally, "water depletion" was assessed via the RECIPE method (Goedkoop et al. 2009).

Performing an LCA on emerging technologies such as HS-DAC is challenging since empirical data other than laboratory implementations of the technology are lacking and therefore uncertainty is introduced where assumptions and predictions of real-world implementations need to be used (Frischknecht et al. 2009; Hetherington et al. 2014; Hospido et al. 2010; Miller and Keoleian 2015; Wender et al. 2014b). However, since energy demand is often responsible for the largest share of the environmental impacts in LCA studies (Huijbregts et al. 2010; Sugiyama et al. 2008; Patel et al. 2012), energy demand is considered an important focal point in LCAs of new technologies(Patel et al. 2012; von der Assen et al. 2016) and a key factor for the costs and environmental impacts of CO_2 capture (von der Assen et al. 2016).

In this study, HS-DAC's and MEA-PCC's energy demand dominate their respective LCAs as well (see section 4.3). We therefore place particular emphasis on the robustness of the energy demand in the inventory data: Energy demand per amount of CO_2 captured is based on technical engineering calculations parameterized by previously published laboratory experiments (HS-DAC) and by pilot capture installations (MEA-PCC). Other life cycle inventory (LCI) data such as sorbent and hardware consumption and estimated machine size are based on laboratory measurements such as sorbent yield per cycle and lifetime and calculations of required hardware components such as pumps and compressors. Sensitivity analyses of LCA results to these input data are provided.

In contrast to MEA-PCC, no large-scale pilot of HS-DAC is yet available. We therefore report its energy and hardware requirements to allow for critical review and insights into HS-DAC's technical feasibility. This also allows a methodological foundation for future studies that will have the benefit of more detailed engineering designs. Our results include sensitivity analyses to gauge the effect of uncertainties in all PCC and HS-DAC inventory data on their respective LCAs.

System boundaries and functional unit

The LCA covers impacts of all materials and processes in the power generation and CCS system up to providing 100 bar CO_2 at injection sites. This includes mining, transporting, and combusting coal, building the power plant, transmission, and the CO_2 capture and compression units, photovoltaic (PV) panels, as well as hardware decommissioning and material disposal. Only preparing the injection sites themselves is not included because the infrastructure requirements are the same for PCC and DAC and, in any case, have been shown to be of minor impact in CCS LCA studies (Koornneef et al. 2008; Modahl et al. 2012; Singh et al. 2011).

We devised two alternative CCS systems that both provide low-carbon electricity from coal to the grid (functional unit = 1 kWh for end-use (at desired amount of net GHG emissions), i.e. not counting electricity output used for capture). In the MEA-PCC scenarios (Figure 4.1) the coal-fired power plant provides the energy for capturing CO_2 using amines, compressing to 130 bar for pipeline transport over a typical 200 km and arrival at injection sites at ~100 bar (Torp and Brown 2005; Maldal and Tappel 2004; BAKLID et al. 1996; Wildbolz 2007). Larger transport distances might be considered but the LCA model shows very minor sensitivity to changes in this distance (see section 4.5), indicating this in not a crucial parameter. In addition, the larger the CO_2 transport distance, the less likely the implementation of CCS using PCC, because costs for pipeline construction will increase with distance.

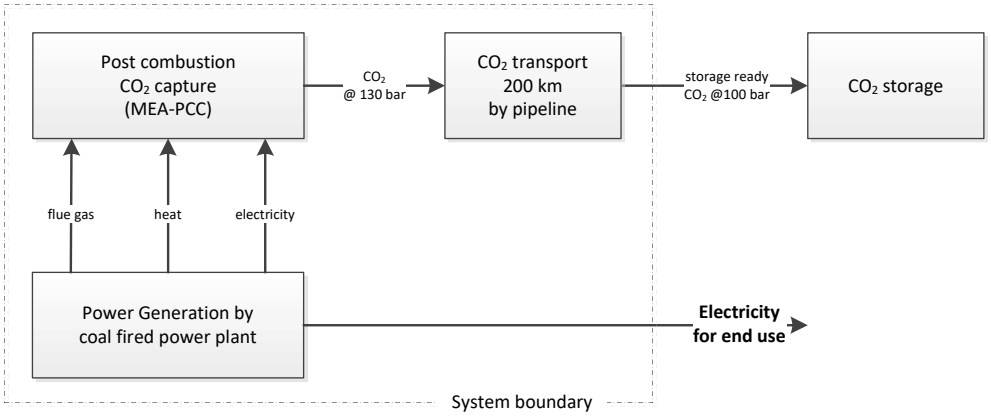

Figure 4.1: System boundaries of post combustion CCS (scenario "MEA-PCC$_{coal}$").

The HS-DAC system (Figure 4.2) is modelled as powered by either coal fired power plants or by a locally installed PV system; multicrystalline-Si, panel efficiency = 13.2%, average annual yield 820 kWh/kW$_{peak}$ (Jungbluth et al. 2007).

Our base case is electricity produced from coal with no carbon mitigation technologies yet in place. The respective inventory data are derived from the ecoinvent (v 2.2) database. The coal-fired power plant in this database (ID868) has an average efficiency of 35%. But newer power plants can have efficiencies up to 45% (World Coal Association 2014), thus reducing their CO_2 emissions in comparison with currently installed power plants. Since it can be expected that PCC will be installed on newly built plants, this study assumes a modern coal fired power plant with 45% efficiency (World Coal Association 2014). This is modelled by adjusting the primary energy demand required per kWh electricity generated (ID859) from 10.2 MJ to 8 MJ which also results in a corresponding reduction of upstream environmental emissions. The plant's CO_2 emissions, without capture, are 0.744 kg CO_2/kWh, or 78% of the emissions from the 35% efficient coal

fired power plant in said database. We base our LCA on an average 500 MW (peak) power plant operating for 4,000 hours per year at full capacity (Dones et al. 2007). Such plant thus generates 500 MW·4000 h = 2 TWh annually and emits $2 \cdot 10^9$ kWh·0,744 kg CO_2 / kWh = $1{,}49 \cdot 10^6$ tonnes CO_2 per year (1 tonne = 1,000 kg).

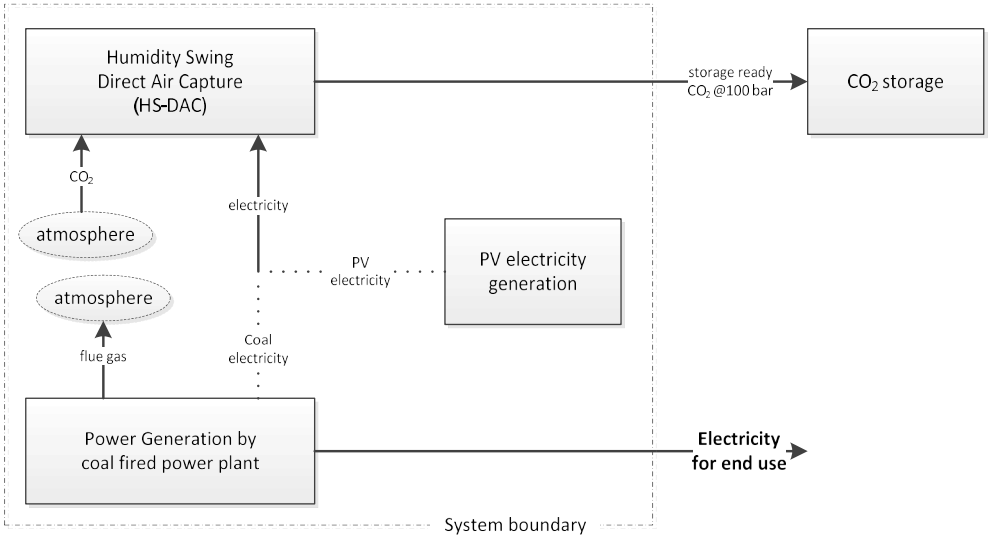

Figure 4.2: System boundaries of CCS using HS-DAC. HS-DAC is powered either by PV-based electricity (scenarios "HS-DAC$_{PV}$(Zero GHG)" and "MEA-PCC$_{coal}$+HS-DAC$_{PV}$(Zero GHG)") or by coal-based electricity (scenarios "HS-DAC$_{coal}$" and " HS-DAC$_{coal}$(Zero GHG)". HS-DAC re-captures the CO2 emissions in flue gas (scenario "HS-DAC"), and, in addition, life cycle GHG from mining and transporting coal, and manufacturing and operating infrastructure ("Zero GHG" scenarios).

We compare this base case to six scenarios, which add various CCS technologies to the base case in order to generate coal-fired electricity with reduced or zero net GHG per kWh for end-use.

- MEA-PCC$_{coal}$: Coal-fired electricity generation with typical MEA-based PCC that captures 90% of stack CO_2 emissions from combusting coal in the plant. The electricity output of the base case coal-plant (2 TWh) is reduced by the energy penalty (below). The LCA and our choice of functional unit quantifies life cycle impacts per kWh of the electricity remaining for end use. Considering the system beyond the power plant itself, this implicitly assumes that the plant's electricity shortfall for end users will be compensated by more MEA-PCC equipped coal plants of the same type (whose LCA per kWh would be the same).
- HS-DAC$_{coal}$: Coal-fired electricity generation combined with HS-DAC, set to capture an amount of CO_2 such that total net lifecycle GHG emissions per

kWh electricity available for end-use are the same as in the previous scenario. The electricity to power HS-DAC is assumed to come from the base case coal plant (or an equivalent plant in another geography).

- MEA-PCC$_{PV}$: Same as the MEA-PCCcoal scenario but the reduction of usable electricity output from the coal plant is compensated by PV instead of by coal plants. This scenario thus assumes a mixed coal-fired and PV generation per functional unit of electricity for end use. The LCA does not include possible need for energy storage to enable use of the intermittent PV electricity on the grid.
- HS-DAC$_{coal}$(zeroGHG): Same as HS-DAC$_{coal}$, but with increased HS-DAC capacity such that net lifecycle GHG emissions are further reduced to zero (i.e., compensating for 100% stack emissions as well as all background emissions).
- HS-DAC$_{PV}$(zeroGHG): Same as HS-DAC$_{coal}$(zeroGHG), but with electricity for HS-DAC provided by PV.
- MEA-PCC$_{coal}$+HS-DAC$_{PV}$(zeroGHG): Combination of previous two scenarios, such that MEA-PCCcoal captures 90% of flue gas CO_2 emissions and HS-DACPV all remaining life cycle GHG.

Life cycle inventory data

As motivated in the introduction, we focus on the energy requirements for both CO_2 capture technologies. Other inventory data (LCI) used in this study (base case and scenarios) are reported in detail in the supporting information (see section 4.5), including sensitivity analyses. Briefly, for MEA-PCC, the LCI data is derived from recent peer-reviewed publications. For HS-DAC, sorbent and hardware consumptions are based on extrapolations from laboratory prototypes and expected sorbent lifetimes. Per tonne CO_2 captured, neither energy nor other consumptions inherently depend on scaling laws such as surface-to-volume or structural performance and are thus largely independent of the HS-DAC's unit envisioned daily capture capacity (here 1 tonne CO_2/day; Discussion). Therefore, LCI data of a real life HS-DAC installation will likely not differ significantly from the hypothetical installation considered here.

Energy requirements for MEA-PCC

Energy requirements for PCC are typically expressed as an "energy penalty" (or equivalent work) that quantifies the drop in electricity output (per tonne CO_2 captured) of a PCC-enabled versus a non-capturing plant (Boot-Handford et al. 2014). Considerable uncertainty exists with regards to this performance metric (Zapp et al. 2012; Jenni et al. 2013): A recent review by Boot-Handford et al. (2014) cites a wide range for PCC in coal-fired power plants, 450kWh/ton to 200kWh/ton, for separation and compression to pipeline pressure to 100 bar or above (Boot-Handford et al. 2014). Specifically for MEA-based PCC (coal), van der Assen et al. (2016) reviewed 10 studies ranging from

473kWh/ton to 211kWh/ton (von der Assen et al. 2016). Importantly, both reviews show a trend towards lower energy penalties seen in more recent studies, indicating potential for lower energy penalties via current and future process improvements/ learning (Rochedo and Szklo 2013).

To account for both the wide range and the longitudinal trend, we therefore carried out LCAs for two energy penalties for MEA-PCC, (i) current/likely case; and (ii) future/ best case. These were determined as explained below and summarized in SI. For ease of comparison with related studies on PCC, we report energies in kWh/tonne captured (1 kJ/mol equals 6,31 kWh/tonne; 1 tonne = 1,000 kg). Further benchmarks and relevant equations to convert between other related performance metrics are given in section 4.5.

For the current/likely case, we determined the energy penalty based on specifications by engineering contractors and process licensors for the International Energy Agency (Davison 2007). For an implementation that reflects the standard studied here (MEA-based PCC for a new pulverized coal plant with 43% gross thermal efficiency), these find an energy penalty of (using parameters from "Fluor" scenario in Tables 1 and 2 from Davison (2007)):

$$\frac{(\eta_{\text{gross}} - \eta_{\text{net}})/\eta_{\text{gross}}}{90\% \cdot \text{PP}_{\text{CO2}}} = \frac{290\text{kWh}}{\text{tonne CO}_2}, \qquad \{\text{Eq1}\}$$

Where η_{gross} denotes the gross thermal power plant efficiency without capture, η_{net} the net thermal power plant efficiency with capture, and "PP_{CO2}" the amount of CO_2 emitted (tonne CO_2 per kWh generated) by the base case power plant.

We consider 290kWh/tonne (46 kJ/mol) to be at the low/optimistic end of the ranges cited in available studies and therefore representative of a current/likely case. For example, 290kWh/tonne closely matches the average energy penalty of the four most recent MEA-based studies reviewed in Boot-Handford et al. (2014); ~280kWh/tonne. Further, 290kWh/tonne is 15% lower than the average of 10 studies (coal, PCC with MEA) reported by van der Assen (2016) ; 1,22 GJ/tonne or 339kWh/tonne. 290kWh/ tonne is 21% lower than the energy penalty used in the landmark 2011 APS study (American Physical Society 2011) which was based on the DoE/NETL's engineering study of MEA-based PCC coal-fired plants (368kWh/tonne) (Netl- 2007).

For the future/best case, we selected one of the lowest energy penalties reported in recent literature (211kWh/tonne = 33kJ/mol), which was determined via simulation for

a MEA-based (9 molar) optimized PCC design (interheated columns).(Van Wagener and Rochelle 2011) 211kWh is in line with the 0.8GJ/tonne (222kWh) predicted by Rochedo et al. (2013, Figure 6) for future energy penalties expected by 2025 (Rochedo and Szklo 2013). 211kWh is also consistent with predictions by NEEDS for 2025 and 2050 (see details in section 4.5 and Bauer et al. (2008). Smaller simulated energy penalties have been reported for novel solvents such as aqueous piperazine (Boot-Handford et al. 2014), as well as solid or membrane sorbents (Goto et al. 2013). These sorbents are not the focus of this study.

Energy and water demand for HS-DAC

To calculate HS-DAC's energy and water consumption, we followed similar considerations as set forth in Lackner et al.(2009). However, we refined these based on recent laboratory studies and common adsorption-on-surface theory (Wang et al. 2011, 2013). We discern four steps of electricity consumption (Figure 4.3). Our model calculates the consumptions as a function of ambient temperature and relative humidity. Below we summarize the calculation steps for 20°C and 30% relative humidity. Further details for each calculation are provided in section 4.5. For ease of comparison with related studies on DAC, we report energies in kJ/mol (1 kJ/mol equals 6,31 kWh/tonne; 1 tonne = 1000 kg).

First, after adsorbing CO_2 from ambient air, sorbent filters are moved to a desorption chamber which is subsequently evacuated (0.01bar). Depending on the chosen filter geometry (i.e., sorbent versus air volume), the capture of 1 mol CO_2 requires evacuating 27 litres of air. Accounting for pump inefficiency (assume 70% electrical-to-mechanical conversion) (Wurzbacher et al. 2011), this evacuation requires 3.8 kJ/mol (24 kWh/tonne) electricity. 0,01 bar is chosen to remove enough of the residual air from the chamber such that the subsequently desorbing CO_2 is ~99% pure. This is followed by spraying the sorbent with water (see auxiliary electricity, below) which prompts the desorption process.

Second, CO_2 drying is achieved through condensation of water from the desorbing CO_2-H_2O gas mixture. Using previously measured sorbent characteristics (Wang et al. 2011, 2013) we calculate that on average 2.89 mol of H_2O have to be condensed per mol CO_2 captured. The condensation is triggered in a cold trap operated by an electrically powered heat pump with a coefficient of performance (COP_r) of 5 (section 4.5). Using 40,65 kJ/mol condensation heat for H_2O, this process requires 23 kJ/mol (145kWh/tonne) CO_2 electricity.

Third, compres

sing CO_2 from its varying desorption pressure (0,0044 bar – 0,0338 bar) to storage-ready 100 bar: We use Aspen HYSYS v8.6® to calculate the electricity input for a 15-stage, intercooled adiabatic compression train, followed for pressures above 57,3 bar (liquid) by a pump (compression ratio 1,88 to 1.64 per stage; Peng-Robertson properties; Schultz polytrophic method; adiabatic efficiency 75%(Mccollum and Ogden 2006)). To adjust for the gradually lower inlet pressure and mass flow rate as the sorbent depletes during each cycle, the train uses centrifugal compressors with controlled inlet guide vanes (or throttle valves) and variable speed (Boyce 2003; Biliyok et al. 2015). The electricity requirement for this step is 31 kJ/mol (196kWh/tonne).

Fourth, we estimate auxiliary electricity, for pumping water (40 molH_2O/mol CO_2), for powering the conveyor to move sorbent filters in and out of the chambers, and for overhead operations & controls (e.g., sensors, actuators, electronics) at ~3% of total electricity required for steps 1-3, or 2.0 kJ/mol (12.6kWh/tonne).

For HS-DAC, the regeneration of the sorbent occurs passively, via the sorbent drying at ambient conditions (see Introduction). Other than the electricity to propel the conveyor system (above), this drying does not require electricity or fuel input. Given a sorbent mass yield of 0,66%, the 37mol water per mol CO_2 captured correspond to ~100 gram of water evaporated per kg of sorbent regenerated. This water-to-material ratio is akin to drying damp sheets of paper on a clothesline. The air flowing through the sorbent filters will gain humidity and cool by 2.1 K (Section 4.5) without any other impact on the environment.

Therefore, for this step, only the water consumption itself, but no other energy or life cycle impact is included in the LCA.

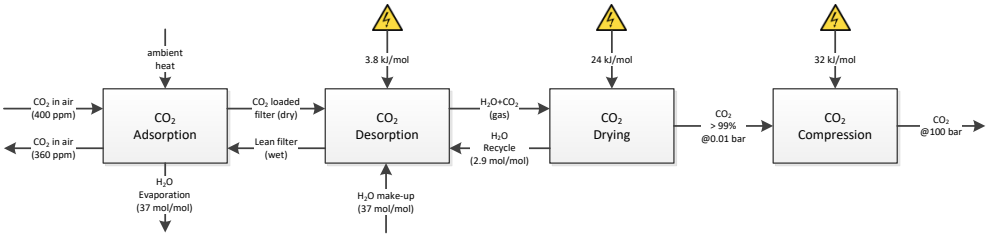

Figure 4.3: Schematic of HS-DAC process with electricity and water consumption as derived in Methods for ambient conditions of 20°C, 30% relative humidity, and 1 bar (details: SI). Not shown is the estimated 2.0 kJ/mol for auxiliary equipment and controls. Except for the temporary cooling during the drying and the multi-stage compression, all units and streams are at ambient temperature. Pressures for H_2O vapour and CO_2 in the desorption and drying steps vary as specified in Methods.

Comparison of energy balances and effect of CO_2 dilution

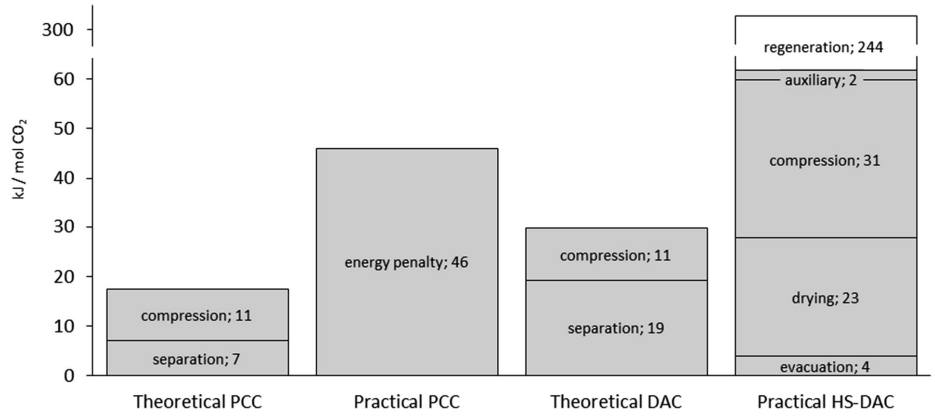

Figure 4.4: Theoretical (thermodynamic minimum) and practical energy breakdown for capturing CO_2 from a 12% concentration (PCC, 90% reduction, 20°C) versus 0.04% concentration (HS-DAC, 400ppm to 360ppm reduction, 20°C) and delivering this CO_2 at 100 bar (130bar for 'Practical PCC'). Required sorbent regeneration energy for HS-DAC (244 kJ/mol) was estimated based on experiments indicating that approximately 6 mol H_2O are swapped for any mol CO_2 on the anionic exchange membrane.(Wang et al. 2013) The required 40.65kJ/mol H_2O evaporation energy is drawn from ambient conditions and therefore not included in the LCI inventory. However, the actual water consumption of the process (37mol H_2O per mol CO_2) is higher, and this higher water consumption is reflected in the LCA.

Based on thermodynamic entropy considerations, separating CO_2 from air via DAC is expected to require ~3 times more energy than from flue-gas via PCC, because DAC captures CO_2 from a ~300 times more dilute source (Figure 4.4) (American Physical Society 2011; Goeppert et al. 2012; Lackner 2013). However, considering practical processes instead of thermodynamic minima, addressing the heat versus electricity portions of the total energy demand in HS-DAC versus PCC separately, and considering 100bar instead of 1bar, gives a more nuanced result: HS-DAC requires 60 kJ/mol CO_2 captured (20°C, 30% humidity case) versus 46 kJ/mol for MEA-PCC (current/likely case). This shows that while the higher dilution of CO_2 has a factor ~3 effect when analysing thermodynamic minimum conditions and 1 bar, this does not translate into equally different electricity requirements of the two processes for practical implementations. This feature of the energy balance is crucial for the comparative LCA of the two processes.

4.3 Results and impact assessment

Life cycle GHG balance

The life cycle GHG balance of the base case power generation and the six scenarios are presented in Figure 4.5. In line with previous LCA studies (Corsten et al. 2013), we find that PCC (at 90% capture rate) reduces the system-wide lifecycle GHG intensity of coal-based electricity by 73%, from 0,85 to 0,23 kg CO_2-eq/kWh.

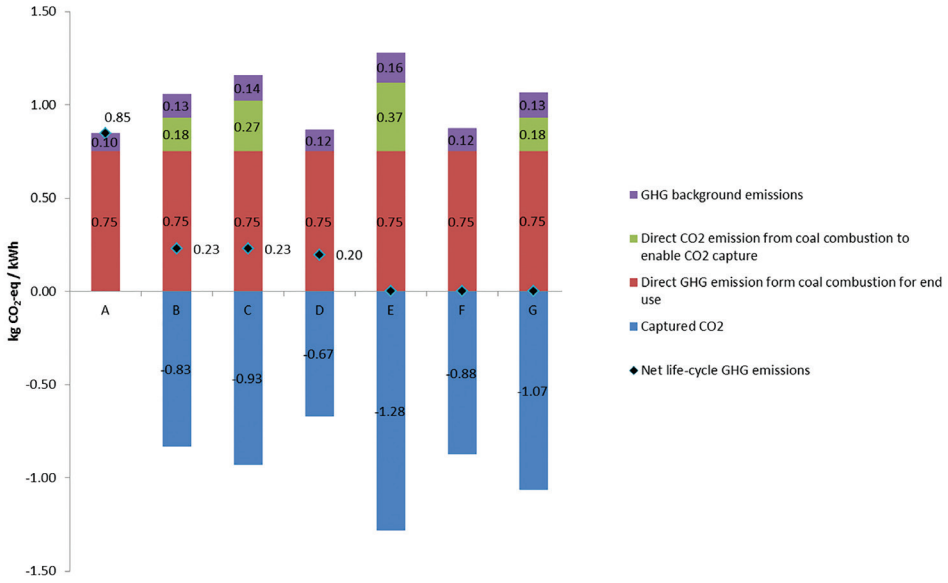

Figure 4.5: Net GHG emissions and breakdown per scenario. Background emissions refer to CO_2 and non-CO_2 GHG emissions that are not the result of coal combustion in the plant itself. Background emissions also include emissions related to the use of PV electricity in scenarios D, F and G. (A=Base case, B=MEA-PCC$_{coal}$, C=HS-DAC$_{coal}$, D=MEA-PCC$_{PV}$, E=HS-DAC$_{coal}$(ZeroGHG), F=HS-DAC$_{PV}$(ZeroGHG), G=MEA-PCC$_{coal}$+HS-DAC$_{PV}$).

When HS-DAC is sized to achieve the same net life-cycle GHG emissions as MEA-PCC (i.e, 0,23 kg CO_2-eq per functional unit), 0,93 kg CO_2/kWh need to be captured, compared to 0,83 kg CO_2/kWh for MEA-PCC. This is because of the higher (coal-based) electricity demand of HS-DAC per amount CO_2 captured. To show the potential of HS-DAC to be proportionally scaled to capture more CO_2 (simply by adding more of the same HS-DAC units), we also modelled two alternatives with zero net GHG emissions. When HS-DAC is powered by electricity from coal, the net zero scenario must capture 1,28 kg CO_2/kWh (0.88 kg CO_2/kWh when powered by PV). In addition, Figure 5 shows the GHG balance for supplementing coal-powered MEA-PCC with PV-powered HS-DAC and for employing PV to make up for the energy penalty of MEA-PCC (see Methods for details of all 6 scenarios).

Non-GHG environmental impacts, problem shift, and role of water

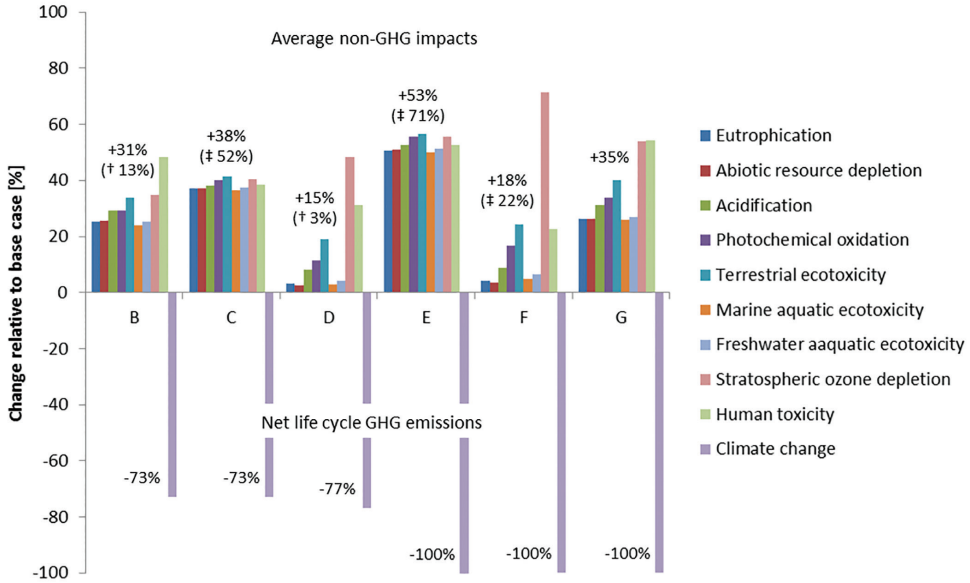

Figure 4.6: Relative change in environmental impacts relative to base case (500 MW coal fired power without CCS). The analysis assumes the current/likely energy penalty for PCC and 20°C/30% ambient conditions for HS-DAC. †Shows average non-GHG impacts (except water) for future/best case energy penalty for PCC. ‡Shows average non-GHG impacts for 10°C/60% ambient conditions for HS-DAC. (B=MEA-PCC$_{coal}$, C=HS-DAC$_{coal}$, D=MEA-PCC$_{PV}$, E=HS-DAC$_{coal}$(ZeroGHG), F=HS-DAC$_{PV}$(ZeroGHG), G=MEA-PCC$_{coal}$+HS-DAC$_{PV}$).

While MEA-PCC and HS-DAC scenarios reduce GHG emissions and thus climate change impacts of coal-based electricity, the increased background processes such as manufacturing additional hardware and mobilizing more materials such as coal (except in the PV scenarios), sorbent, and water increase other environmental impacts per functional unit (Figure 4.6). Consistent with previous LCA studies (Koornneef et al. 2008; Singh et al. 2011), we find that MEA-PCC increases the other 9 impacts tracked in our study (except water) by 31% on average versus the base case (see Figure 4.7 for water depletion). Employing HS-DAC to achieve the same GHG mitigation increases this "problem shift" from 31% to 38%, reflecting the higher electricity demand of HS-DAC per amount CO_2 captured. However, the LCA also shows that with HS-DAC powered by PV (intermittently), a complete removal of GHG per functional unit can be achieved with a smaller problem shift of 18% (Discussion). The increase in stratospheric ozone depletion in this scenario is due to upstream emissions in the production of the specific PV cells and is noticeable in all scenarios that use PV. For HS-DACcoal, the average penalty is 53% (all impacts except GHG and water). If CCS implementation does not aim for net zero GHG – but rather for the capture technology with the smallest

problem shift per GHG avoided – then the preferred scenario is employing MEA-PCC while compensating the energy penalty with PV (smallest problem shift 15%). Corresponding metrics of all impact categories expressed per kg net GHG avoided are provided in section 4.5.

HS-DAC requires fresh water in order to prompt the humidity swing (low quality, non-potable water is sufficient, however salt water would likely lead to prohibitive contamination of the sorbent) (Lackner et al. 2012). Direct water demand of HS-DAC is 15,2 m^3 per tonne CO_2 captured compared with 0,8 m^3 / tonne for MEA-PCC. Therefore, life cycle water depletion in scenarios involving HS-DAC is larger than in the two PCC scenarios (Figure 4.7). Note that the coal-burning power plant assumed in our base case does not feature coal transport via slurry pipeline or wet tower cooling, two technologies that would lead to higher water withdrawal even in the uncaptured base case (up to 4m3 and 6m3 water per tonne CO_2 emitted, respectively) (Fthenakis and Kim 2010).

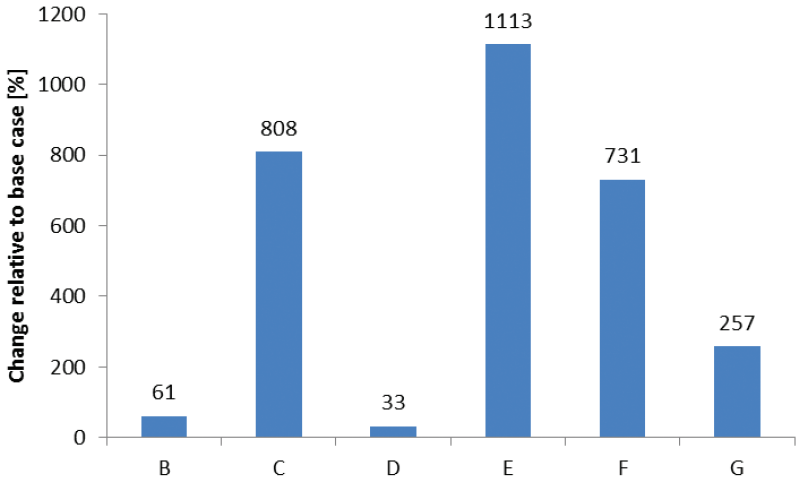

Figure 4.7: Water depletion for all scenarios (20°C, 30% relative humidity case for HS-DAC and current/likely case for MEA-PCC) in % relative to base case. (B=MEA-PCC_{coal}, C=HS-DAC_{coal}, D=MEA-PCC_{PV}, E=HS-DAC_{coal}(ZeroGHG), F=HS-DAC_{PV}(ZeroGHG), G=MEA-PCC_{coal}+HS-DAC_{PV}).

Uncertainty and sensitivity to ambient conditions

The dominant driver of the LCA results for both HS-DAC and MEA-PCC (except PV scenarios) is the additional combustion of coal (per functional unit) to enable the capture processes, rather than the additional materials and infrastructure (Figure 4.5). This is driven by the capture process' electricity requirement which could be robustly quantified based on previously published laboratory measurements (HS-DAC) and engineering studies (PCC) (Methods). In contrast, sorbent and hardware consumptions

are estimated, implying higher uncertainty. However, these parameters are less material to the LCA (sensitivity analyses on sorbent and hardware consumptions are included in section 4.5).

Crucially, the operation of a HS-DAC unit is sensitive to ambient conditions, particularly temperature and humidity, as these affect the sorbent's loading characteristics and thus HS-DAC's electricity and water consumption. We therefore ran sensitivity analyses to quantify this effect on electricity and water requirement and corresponding LCA results. For example, changing the 20°C and 30% humidity in our model to reflect a colder, more humid (10°C|60%) climate (or equivalent variations throughout the day) will lower the sorbent's initial CO_2 load when it enters the desorption chamber. The resulting detrimental effect on sorbent yield and thus long term sorbent consumption per tonne CO_2 captured can be partially counteracted by desorbing to lower pressures in a cycle. Such operation at 10°C|60% achieves 0.49% yield (versus 0.66% for the 20°C|30% operation; yield expressed as mass CO_2 captured per mass sorbent per cycle). But, this causes more direct electricity (+21%) and water consumption (+33%) relative to the base case. Therefore, under such cold/humid operation, the problem shift would increase from 38% to 52%, 53% to 71%, 18% to 22% and from 35% to 37% for the HS-DAC_{coal}, HS-DAC_{coal}(ZeroGHG), HS-DAC_{PV}(ZeroGHG) and MEA-PCC_{coal}+HS-DAC_{PV}(ZeroGHG) scenarios respectively (Figure 4.6). See section 4.5 for all impacts including water.

In contrast, due to the sorbent loading responses around these conditions, warmer and dryer conditions (30°C|10%) would only marginally increase the sorbent's initial CO_2 load. If desorption pressure is adjusted to maintain the same mass yield 0,66%, this would have only very small effects on electricity and water consumption (<1%) and likewise result in nearly the same average non-GHG LCA impacts and water demand as those for the 20°C|30% operation shown in Figure 4.6. Note however that such warmer and dryer conditions would likely accelerate sorbent drying (and vice versa). This would improve capacity utilization of a given machine and thus economics. Similarly, actual sorbent yields during continuous cycling will be somewhat lower than the equilibrium sorbent yields calculated here, again affecting economics but not energy balance (details on sorbent kinetics: section 4.5).

4.4 Discussion

In this paper we have analysed the comparative merit of flue gas capture and direct air capture, by comparing the environmental impacts of two CO_2 capture processes: the traditional one where CO_2 is captured from coal flue gas using MEA-based PCC; the other with CO_2 captured from air with HS-DAC. Previous studies stressed that HS-

DAC may be a promising technology but that more research on DAC is needed and that R&D and commercialization of DAC at larger scales will provide the necessary experience for further development (Williamson 2016; Smith et al. 2016). In particular, DAC has been viewed as being too energy-intensive (American Physical Society 2011), citing up to 45GJ per tonne carbon equivalent (540kJ/mol CO_2) (Smith et al. 2016). The research presented here contributes to the necessary broadened knowledge base for HS-DAC.

We conclude that - based on the currently available data - by virtue of its limited high-exergy energy requirement and corresponding environmental penalties, HS-DAC could expand the scope of CCS, including the realization of net-zero GHG electricity from coal. This could unlock potential advantages of DAC, including fewer social obstacles related to CO_2 transport and storage near population centres, the option to compensate for GHG emissions from sectors with diffuse emissions (e.g., aviation), the possibility of re-capturing 100% of the GHG emissions along the entire life cycle, and the possibility of CCS being used as a last resort back-stop technology to reduce GHG in the atmosphere (negative emissions) (National Academy of Sciences 2015). Still, PCC is clearly the preferred short-term option to capture CO_2 from flue gases, particularly when the goal is reducing GHG emissions (with minimal problem shift to other environmental impacts) rather than full net-zero-GHG electricity.

The higher electricity demand of HS-DAC versus MEA-PCC, other chemical and hardware requirements, and the separation of combustion and capture site could conceivably lead to a problem shift, i.e. prohibitive increases in environmental issues other than climate change. From our research we find that this fear is probably unfounded: Comparing MEA-PCC to HS-DAC (at equivalent GHG emissions), the shift to other environmental impacts does increase (from 31% to 38%, Figure 4.6) but is still lower than the factor 3 that might have been expected based on the increased minimum energy demand for separating CO_2 from a gas mixture. If HS-DAC were used to reduce lifecycle GHG emissions of coal-based power to net-zero (but is itself driven by PV), other impacts are on average lower than with MEA-PCC.

However, our analysis also shows HS-DAC's higher water consumption and sensitivity to climate (temperature, wind speed, humidity). Resulting crucial disadvantages of HS-DAC are therefore that it cannot be universally sited but rather requires access to water and minimal ambient wind speeds and temperature / humidity conditions outside which HS-DAC becomes less efficient. Quantifying sorbent drying times and how these can be optimized will be an important area of further study. As an extreme example, a complete standstill of the unit while in rainy, wind-still weather would raise capital

costs, albeit not energy requirement per tonne CO_2 captured. This makes HS-DAC somewhat akin to other distributed technologies in the sustainable energy realm such as wind farms or PV: its largely passive operation offers advantages for average energy balance; but exposure to the elements leads to intermittent operation and geographical limitations.

Decoupling emissions from capture

Zero-GHG DAC leverages two unique features of distributed CDR. First, to achieve net-zero GHG, HS-DAC is scaled up proportionally, without incurring the non-linear energy penalties caused by scrubbing flue gas to >90% captured from stacks. Also, PCC could not clean up any background emissions. Second, running HS-DAC intermittently on renewables (while still generating grid electricity via coal to serve society's continuous demand) leverages HS-DAC's ability to operate in remote geographies, possibly on continents other than the one on which the coal-fired power plant itself is located, where CO_2 may be easier to store and intermittent renewable electricity may be plentiful; meanwhile the atmosphere (and its coupling with ocean systems) acts as a temporal buffer for the emitted and re-captured GHG (Eisaman et al. 2012).

Implementation considerations

In addition to its reliance on suitable ambient temperature, wind, and relative humidity, HS-DAC's large water consumption means that its implementation will be limited to geographies where such amounts of water are available – even though this water is not polluted or contaminated but simply evaporates and thus returns to the natural water cycle.

While other practical considerations were not the immediate focus of this study, our process data on MEA-PCC and HS-DAC also allows us to evaluate the relative requirements of HS-DAC in terms of land use, installation size, and operating cost.

For MEA-PCC, the actual land requirement of the capture installation (excluding CO_2 transport infrastructure) can be estimated to be of roughly the same size as the power plant from which it captures (IEA Greenhouse Gas R&D programme 2005). For reference, the 500 MW power plant used in this study occupies an area of 125,000 m^2 (Dones et al. 2007). For HS-DAC, the land requirement will be dominated by and can thus be approximated as its required vertical wind facing cross section, using ambient wind speeds (2m/s, 20°C), and CO_2 uptake from air per pass through the DAC unit (40ppm or 10%). We further assume an idle time of 12 hours per day, for lack of sufficient wind or suitable weather. To capture 1 tonne CO_2 per day, an HS-DAC unit would thus require a wind facing cross section of 160m^2. This cross section must be

occupied by an array of sorbent filters that are in the adsorbing stage of the humidity swing cycle. Behind these filters, there are additional filters (in the drying phase) and a container-like structure on the ground where filters undergo desorption (as well as pumps, cold traps, compressors, and infrastructure for the captured CO_2). Capturing 90% of the 1.49×106 tonnes CO_2 / year emitted by the coal plant would thus require 3674 such HS-DAC units or 587.835 m^2 of land (4,7 times that of MEA-PCC). To allow CO_2 concentration and wind speed to recover in-between units, the units would be scattered over a larger land area. However, this area would be available for other use (akin to the space in-between individual windmills on large windfarms).

Regarding costs, consumption of electricity (20°C|30% case, 378 kWh/tonne CO_2 at \$0,07/kWh=\$26,5/tonne CO_2) (American Physical Society 2011), water (15,2m^3/tonne CO_2 at \$0,40 per m^3=\$6,1/tonne CO_2) and make-up sorbent filters using Marathon A (1,52kg/tonne CO_2 at \$6/kg=\$9,1/tonne CO_2) would yield direct operational costs of \$42 per tonne CO_2. To facilitate comparison to previous studies, we use the same electricity cost as in the APS study (American Physical Society 2011). Other costs are conservative estimates for non-potable water and customized, extruded resin filter elements. Benchmarking HS-DAC costs against a lower bound of \$60 per tonne CO_2 for MEA-PCC (American Physical Society 2011), this would leave \$18/tonne CO_2 for HS-DAC capital charge and O&M costs, or ~\$43.000 capital layout for a 1 tonne CO_2/ day HS-DAC unit (assuming capital charge of 10% of capital layout and annual O&M costs of 5% of capital layout). A more conservative CO_2 price of \$100/tonne would allow ~\$142.000 per unit. While it is difficult to predict the capital layout of HS-DAC units – as with any future technology –, we conclude that HS-DAC does not require difficult process conditions such as high temperatures, nor does it need rare or expensive materials or equipment. Furthermore, the economic benefit from the freedom to locate the HS-DAC units and injection at optimal sites independent of coal power plants and transport concerns is difficult to assess without specific case studies, but it may be significant (The Royal Society 2009). A detailed analysis of the total costs of HS-DAC should be the focus of future research.

Acknowledgements
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4.5 Appendix

This appendix contains the supporting information for this case study and includes details on the modelled processes, supporting calculations.

4.5.1. Energy analysis

HS-DAC energy consumption

The energy consumption for humidity swing based direct air capture (HS-DAC) is broken down into 5 steps, 4 for electricity, and 1 for ambient heat, addressed one by one below. The 5 steps deliver >99% pure CO_2 at 100 bar pressure. For ease of comparison with related studies, we report energies in kJ per mol CO_2 captured (1 kJ/mol equals 6,31 kWh / tonne; 1 tonne = 1000 kg).

Overview: Determining equilibrium loading of CO_2 for sorbent yield

To obtain the electricity consumption in the DAC process, we follow similar considerations and calculations as set forth in Lackner et al.(2009) However, we refined these calculations based on recent laboratory studies and common adsorption-on-surface theory.(Wang et al. 2011, 2013) Together, these let us quantify the equilibrium loading of the sorbent with CO_2 as a function of partial CO_2 pressure (P_{CO2}), relative humidity (H), and temperature (T) to which the sorbent is exposed. Following Langmuir adsorption theory, we write:

$$RL(P_{CO2}, H, T) = \frac{P_{CO2}}{K(H,T)\cdot\left(1+\frac{P_{CO2}}{K(H,T)}\right)} \quad (1)$$

where K (in atm) is the partial pressure at which the relative loading RL is 50%. RL is thus a dimensionless number between 0 and 1 that indicates the fraction of total available CO_2 adsorption capacity that is utilized (in equilibrium) at any specific PCO_2. The absolute loading AL (in mol CO_2 per kg of sorbent) can be found based on the carbon chemistry of the particular sorbent (Lackner 2009):

$$AL(P_{CO2}, H, T) = RL(P_{CO2}, H, T) \cdot \frac{1.78}{2} mol/kg \quad (2)$$

$K^*(T, H)$, in which * indicates dimensionless pressures expressed as fractions of the standard condition 1 atm, can be rewritten as a function of the standard molar Gibbs free energy of adsorption dG:

$$K^*(T, H) = e^{\frac{dG(T,H)}{RT}} \quad (3)$$

where R is the ideal gas constant. Laboratory measurements showed that *dG(T, H)* is well described by the following fit function (Table 2, 3rd of 4 parameter sets, in Wang et al. (2013).

$$dG(T,H) = -32.20\frac{kJ}{mol} + 15.45\frac{kJ}{mol}\cdot H + 0.02564\cdot(T-288.15)\frac{kJ}{mol\cdot K} \qquad (4)$$

Using above *AL*, we can now determine the sorbent yield Y_{CO2} (in mol CO_2 per kg of sorbent) per any one cycle, where a cycle is characterized by *H* and P_{CO2} during adsorption (here: 30% and 400 ppm of 1 bar, respectively) and *H* and P_{CO2} during desorption. At desorption, *H* is 100% per definition of the process and PCO_2 can be controlled by the flow-rate of the compression pump. At 30% and 400 ppm of 1 bar, the sorbent adsorbs to RL_{Start} = 0,97. Once in the desorption chamber, evacuated, and exposed to *H* = 100%, it desorbs gradually from *RL* = 0,97 (corresponding to P_{CO2} = 0,0340 bar) to RL_{End} = 0,80 (corresponding to P_{CO2} = 0,0044 bar, which is the lowest pressure from which CO_2 must be subsequently compressed). Based on above *AL* as function of *RL*, resulting YCO_2 is 0,15 mol CO_2 per kg of sorbent (0,66% mass yield).

Step 1: Evacuation and desorption (electricity)

The energy required for desorption of the sorbent consists of electricity to pre-evacuate ambient air from the desorption chambers. The DAC process studied herein uses sorbent elements constructed such that, in the desorption chamber, each kg sorbent is surrounded by 4 litres of air (i.e., ~20% volume ratio given that the sorbent's density is close to 1 kg/litre). Given above YCO_2, capture of every 1 mol CO_2 therefore requires evacuating 27 litre (design dependent). At 100 kJ per m^3 (1 bar, 20˚C) and further accounting for pump inefficiency (assume 70% electrical to mechanical conversion),(Wurzbacher et al. 2011) this gives 3,8 kJ/mol. Note this calculation uses a simple P·V view to estimate the mechanical work required to evacuate a given volume against a constant outside pressure. Overall, the contribution of the desorption step to the total electricity consumption (steps 1-4) is small (<10%). Therefore, we leave the calculation at this level of detail.

Step 2: CO_2 drying (electricity)

During desorption, the sorbent gives off a mixed stream of CO_2 and H_2O. While P_{CO2} in this stream drops gradually as loading decreases from RL_{Start} to RL_{End} (see explanation above pressure provided in main manuscript), the partial pressure of H_2O in the stream stays constant (given by the vapour pressure at 293 K: 0,024 bar). This means that the CO_2:H_2O mol ratio of the stream, which is proportional to the ratio of partial pressures in the stream, decreases during desorption of one cycle. The average mol-ratio for a full cycle can be found by numerically integrating equations (1) and (2) using standard

software. We find that on average 2,89 mol of H_2O have to be removed for every mol of CO_2 retrieved. The removal is achieved by condensation via a cold trap cooled by a heat pump with a coefficient of performance (COP_r) of 5. Using 40,65 kJ/mol condensation heat for H_2O, this yields 23 kJ per mol CO_2.

Efficiency of cold trap:

COP_r is derived as follows: Cooling the desorption stream by 30°C lowers the H_2O vapour pressure to 0,0032 bar (Thangavel et al. 2013). The theoretical maximum COP_r in an ideal Carnot process, assuming 20°C=293,15 Kelvin ambient, is thus (293,15K-30K)/30K = 8,77. With a further drop of 43% due to inefficiencies in a real system, we arrive at COP_r of 5,0 which is achievable with real refrigeration systems (Thangavel et al. 2013).

Phase changes of H_2O in cold trap:

In order to yield a sufficiently pure CO_2 stream (if near zero residual water is desired before entering the multi-stage compression), the partial pressure of water vapour in the desorption stream must be sufficiently reduced. At 20°C ambient, the cold trap therefore has to be cooled to temperatures below 0°C. In this regime, the water vapour in the desorption stream will undergo a phase change directly to a solid. After each cycle, the cold trap will refill with ambient air and warm up; the ice will melt and can thus be removed from the system by gravity. The HS-DAC installation envisioned in our study has a capacity of 1 metric tonne CO_2 per day and during its 12h/per day operation undergoes 30 cycles. About 39kg of ice have to be removed per cycle. However, this amount of ice is an upper bound. In a real implementation, the cold trap does not have to be at a single uniform temperature < 0°C. Rather, the water removal process will be spread out through a series of interconnected pipes: The desorption stream which exits the sorbent at 20°C with 0.024bar vapour pressure (above) will first be cooled to temperatures still above 0°C, such that a portion of the water already condenses out as a liquid. The lower temperature drop also increases COP_r in this portion of the piping system (because of the higher Carnot efficiency), thus reducing the electricity requirement for this portion of condensed water. The temperature of the desorption stream will then gradually be dropped further as the desorption stream is pumped through the piping system. Only the last portion, before entering the multi-stage compression train, has to be cooled to temperatures sufficiently low to reduce any remaining water vapour (by forming ice) in the desorption stream to less than 1/100 of the partial CO2 pressure (~99% purity).

Step 3: CO_2 compression (electricity)
Energy required to compress the produced CO_2 consists of electricity to drive a CO_2 compressor. As explained for the drying step above, different portions of CO_2 desorb at different pressures (to which they fill the desorption chamber). Therefore, each such portion requires different compression work to yield 100 bar CO_2. The calculation (Aspen) is explained in the main text.

Step 4: Auxiliary processes (electricity)
The steps above cover all energy-intensive processes of the proposed HS-DAC. Remaining are some electricity to move sorbent (in and out of the desorption chamber) and water, and furthermore some overhead for operations & controls (such as sensors, actuators, monitoring electronics).

This electricity was estimated bottom-up, using an envisioned machine size of 1 tonne CO_2 captured per day (which runs 24h-12h=12h each day). 2,0kJ/mol (or 13kWh per tonne) which amounts to 3,3% of the total electricity consumption (60kJ/mol) comprises the following elements (all other electricity requirements are accounted for in other steps (see Figure 4.3):

- 0,7kWh to move the filters on a horizontal conveyor-belt like structure (for drying and adsorption outside the regenerator as well as for shuttling them through the regenerator where CO_2 desorbs): Except for friction and efficiency losses of the electric motor powering the conveyor, this process does not consume energy. To estimate the electricity consumption of the real system (i.e., with friction), the conveyor motor (and appropriate gear system) is envisioned at a power to overcome 1% incline at 1m/s speed, i.e. 0,01W per kg mass conveyed. To move the total mass of filter elements in the envisioned machine (5,56 tonne) this accounts for 0,67kWh over 12h operation per day. This is only about 5% of the total auxiliary electricity requirement, and therefore a more accurate calculation is deemed unnecessary.
- 1,3kWh to spray water onto the sorbent filters in the evacuated regenerator: Per tonne CO_2, 16,4m^3 water needs to be sprayed (of which 1,2m^3 is later condensed out and thus recovered rather than consumed; see main manuscript). This is envisioned by compression of the water from 1 to 3 bar by a 70% efficient electric pump (Wurzbacher et al. 2011).
- 11kWh to power electronic controls, sensor, and actuators for water and air valves: We estimate this conservatively (i.e., high) to require 100W of continuous power per tonne of total machine weight. The total machine weight including filter elements is 8.9 tonnes. This accounts for 11kWh over 12h operation per day.

Step 5: Sorbent regeneration (ambient heat)

During regeneration the sorbent is returned to its thermodynamic base state (i.e., mostly dry, and low CO_2 loading). As shown previously and explained in the main manuscript (Wang et al. 2011, 2013), the required energy for this sorbent regeneration is driven predominantly by ambient heat (provided by the 30% humidity/20°C air) that causes the sorbent to dry. However, this heat does not have any associated life cycle impacts and is therefore not counted as part of the inventory in the environmental assessment.

MEA-PCC energy consumption

Energy consumption of MEA-PCC is treated via the concept of energy penalty (see the main text and Table 4.1 below).

Table 4.1 Energy penalties for PCC used in this study and literature benchmarks. All figures for 90% capture rate, MEA-based PCC in coal-fired plants, and inclusive of compression to ~130bar.

Table 4.1: Energy penalties for PCC used in this study and literature benchmarks. All figures for 90% capture rate, MEA-based PCC in coal-fired plants, and inclusive of compression to ~130bar.

Overall	Typical range reported in literature (Table S1 in Von der Assen et al. (2016), including older, not state-of-the-art implementations	211-473 kWh/tonne
Current/likely case used in this study: 290 kWh/tonne	Average of four most recent MEA-based studies reviewed in figure 3 in Boot-Handford et al. (2014)	280 kWh/tonne
	Specifications by engineering contractors and process licensors for the International Energy Agency (Davison 2007)	290 kWh/tonne
	Average of 10 studies reported in table 1 in Von der Assen (2016)	339 kWh/tonne
	American Physical Society (2011) and DoE/NETL's engineering study by Davison (2007)	368 kWh/tonne
Future/best case used in this study: 211 kWh/tonne	Future scenarios predicted for 2025 and 2050 horizons by NEEDS (2008) and Bauer et al. (2008) (Table 4.18, details see SI)	~200 kWh/tonne (6% efficiency penalty)
	Lowest recent simulation result reported in literature by Wagener and Rochelle (2011)	211 kWh/tonne
	Past PCC learning curve; figure 6 in Rochedo et al. (2013) extrapolated to 2025	222 kWh/tonne

Here we provide further benchmarks. In addition to energy penalty, which is specified in electric energy per mass CO2, three other often published performance metrics exist in literature (Vasudevan et al. 2016). As % figures, these are a function not only of the actual energy penalty (in kWh/tonne) but further of the plant's CO2 emissions and

thermal efficiency, and therefore a less direct metric when comparing different studies. However, we include these here as a further point of comparison for the energy penalties used in our calculations reported in the main article (current/likely and future/best case). We use the following definition for the three metrics (e.g., Vasudevan et al.(2016)

$$\textbf{\textit{Parasitic load}} = \frac{\eta_{gross} - \eta_{net}}{\eta_{gross}} \qquad \{\text{Eq S1}\}$$

Where η_{gross} denotes the gross thermal efficiency of the power plant without capture and η_{net} denotes the net thermal power plant efficiency with capture. Note that in some

$$\textbf{\textit{Increase in fuel use}} = \frac{1}{(1 - \%\ Parasitic\ load)} - 1 \qquad \{\text{Eq S2}\}$$

$$\textbf{\textit{Efficiency penalty}} = \eta_{gross} - \eta_{net} \qquad \{\text{Eq S3}\}$$

(Note this is sometimes referred to as energy penalty.)

Substituting EqS1 in Eq1 (main text) and using 744 gram CO_2/kWh from our base case, we find that the 290kWh/tonne for the current/likely case corresponds to a *Parasitic load* of 19,4% (or an *Increase in fuel use* of 24,1%). Further using the 45% thermal efficiency of the un-capturing plant in our base case, we infer from EqS1 and EqS3 that the *Efficiency penalty* corresponding to 290kWh/ton is 8,7%. 8,7% is at the low end of those reported in Zapp et al.(2012) and close to the 9% for PCC in pulverized coal power plants used by Reiter et al. (2015), table 2. The *Parasitic load* corresponding to our future/best case (211kWh/ton) is 14.1%, in line with the lower end (~15%) of the wide range predicted for 2025 by a group of 12 experts surveyed by Jenni et al.(2013), Figure 1. The *Efficiency penalty* corresponding to 211kWh/ton is 6.4%, consistent with the 6% reported for optimistic, future looking scenarios as those specified for 2025 and 2050 horizons by NEEDS (2008) Bauer et al. (2008), table 4.18.

Temperature drop in air as it aids water evaporation

The sorbent is regenerated and rendered back into its adsorbing state once all originally applied water is evaporated off the sorbent during the drying phase. We can therefore calculate the ambient heat drawn from the (30%humidity/20°C) ambient air per ton CO_2 captured and infer the resulting temperature drop in the airflow through the filters.

At a capture rate of 10% (400 ppm to 360 ppm, design-dependent), $13{,}9 \times 10^6$ m^3 (1bar, 20°C) need to pass through the array of sorbent filters for any metric ton of CO_2 captured. This amount of air will also pass through the second set of sorbent filters in the drying phase of the HS-DAC cycle (main manuscript). The heat to evaporate 37mol

H_2O per mol CO_2, at 40,7 kJ/mol (evaporation heat of water), will be drawn from this air. Given an isobaric volumetric heat capacity of air of 0,00121 J/(mol·K), this will cool the air by 2,1 Kelvin.

Note this calculation assumes that the airflow speed through the second set of filters (i.e., the ones in the drying state likely positioned at some distance behind the ones in the adsorbing state) is the same as that through the first set (and not slower). This assumption likely overestimates the air volume flowing through the second set and thus underestimates the temperature drop. At the same time, however, this calculation disregards the effect of evaporation heat supplied by sunlight, thus overestimating the temperature drop.

4.5.2. Environmental analysis

Defining the scenarios

A base case and six scenarios for comparison have been defined. Here we describe these scenarios as modelled in the LCA model, as well as one additional 7th scenario (scenario H=PV electricity) that is included here only for the purpose of reference and comparison.

Table 4.2: Scenarios for environmental assessment.

A. Base case	Providing electricity to the grid with a 45% efficient capture ready Europe based 500 MW coal fired power plant. Life cycle environmental impacts are based on this system delivering 1 kWh of electricity to the grid.
B. MEA-PCC_{coal}	Coal-fired electricity generation with typical MEA-based PCC that captures 90% of stack CO_2 emissions from combusting coal in the plant.
C. HS-DAC_{coal}	Coal-fired electricity generation combined with HS-DAC, set to capture an amount of CO_2 such that net lifecycle GHG emissions per kWh electricity available for end-use are the same as in the previous scenario (B). The electricity to power HS-DAC is assumed to come from the base case coal plant (or an equivalent plant in another geography). N.B. in the LCA model the amount of CO_2 captured is increased (to 0,935 kg) such that the end-to-end climate change impacts of both scenario B and C are the same (0,229 kg CO_2-eq).
D. MEA-PCC_{PV}	Same as the MEA-PCC scenario (B) but the reduction of usable electricity output from the coal plant, caused by post combustion capture, is compensated by PV. This scenario thus assumes a mixed coal-fired and PV generation per functional unit of electricity for end use. The LCA does not include possible need for energy storage to enable use of the intermittent PV electricity on the grid.
E. HS-DAC_{coal} (ZeroGHG)	Same as the HS-DAC_{coal} scenario (C), but with increased HS-DAC capacity such that net lifecycle GHG emissions are further reduced to zero (i.e., compensating for 100% stack emissions as well as all background emissions). N.B. to demonstrate the full potential of HS-DAC the amount of CO_2 captured is increased (to 1,27 kg) such that the end-to-end climate change impact is netted to zero (0 kg CO_2-eq).

F. HS-DAC$_{PV}$ (ZeroGHG)	Same as the previous scenario (E), but the electricity demand for HS-DAC is provided by PV. N.B. to demonstrate the full potential of HS-DAC the amount of CO_2 captured is increased (to 0,875 kg) such that the end-to-end climate change impact is netted to zero (0 kg CO_2-eq).
G. MEA-PCC$_{coal}$+ DAC $_{PV}$ (ZeroGHG)	Combination of previous two scenarios (E and F), such that MEA-PCC captures 90% of flue gas CO_2 emissions and HS-DAC all remaining GHG emissions such that the end-to-end climate change impact is netted to zero (0 kg CO_2-eq). N.B. capturing 0,235 kg CO_2 is used to bring the net-system GHG emission back to zero.
H. PV electricity	Delivering 1 kWh of electricity generated by PV (without any capture, see SI chapter 0).

Life cycle inventory (LCI) data

Capture ready power plant

It has been reported that an additional reduction of SO_2, NO_x and particulate matter (PM) emissions of respectively 95%, 2,5% and 50% can be expected when adding post combustion carbon capture to a power plant (without CO_2 capture). Installing additional flue gas cleaning will prevent the carbon capture equipment from being affected (Volkart et al. 2013; Giannoulakis et al. 2014) by SO_2, NO_x and particulate matter (PM). Therefore, in the LCA model the coal combustion process is adjusted to include the additional reduction of NO_x, SO_x and PM emissions. For NO_x and SO_x this results in larger quantities retained and less emitted, while for PM only a reduction in emissions is modelled. The PM that is retained is not accounted for and cut-off, due to lack of information for the electrostatic precipitator (ESP). The additional equipment need is not modelled since all scenarios use the same capture ready power plant and this will not influence the relative results. The changes in environmental emissions are displayed in Table 4.3.

Table 4.3: Difference in selected environmental emissions between basic and capture ready power plant.

	Non capturing	**Capture ready**
SO_x retained, in hard coal flue gas desulphurization	5.11 x 10^{-4} kg	5.69 x 10^{-4} kg
NO_x retained, in SCR	2.11 x 10^{-4} kg	2.13 x 10^{-4} kg
Nitrogen oxides	7.86 x 10^{-5} kg	7.66 x 10^{-5} kg
Sulfur dioxide	6.07 x 10^{-4} kg	3.04 x 10^{-6} kg
Particulates, < 2.5 um	1.82 x 10^{-6} kg	9.1 x 10^{-7} kg
Particulates, > 10 um	5.11 x 10^{-6} kg	2.56 x 10^{-6} kg
Particulates, > 2.5 um, and < 10um	2.14 x 10^{-7} kg	1.07 x 10^{-7} kg

Post combustion CO_2 capture (PCC)

Figure 4.8 shows the unit processes for the construction of the PCC plant and the capture process itself.

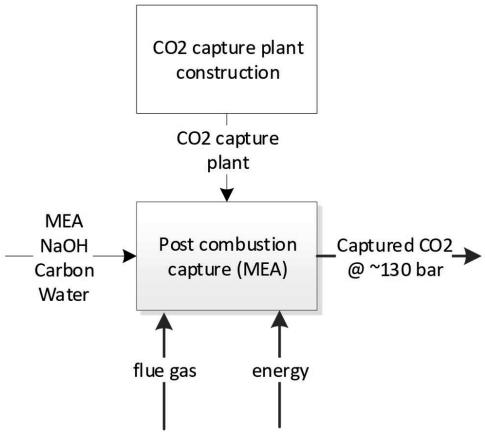

Figure 4.8: Post combustion CO_2 capture process (the 130 bar pressure of the captured CO_2 is based on the pressure needed for subsequent CO_2 transport over 200 km.

Construction of the PCC plant

The size of the capture installation and the amount of required resources depend on the amount and the concentration of CO_2 in the flue gas from which CO_2 is captured. Van der Giesen(Van der Giesen 2008) estimated the steel demand for a PCC installation that captures ~3MT CO_2/year from a PC power plant. The PCC installation is modelled on its expected steel demand, namely 5.560 tonne low –alloyed steel and 1.470 tonne stainless steel. For this study the steel demand is scaled down proportionally to represent a PCC plant that emits 1,5 MT a year. In this study a capture rate of 90% is assumed, hence an annual amount of 0,9 x 1,5 MT CO_2 = 1,35 MT CO_2 is captured. With a life time of 30 years it needs 1 / (1,35 x 10^6 x 30) = 2,46 x 10^{-8} units of PCC installation per tonne CO_2 captured.

The PCC installation does not solely consist of steel but also of other materials such as concrete and other building and installation equipment. In addition, energy has to be used to construct the PCC installation. It can be assumed that the PCC plant is rather similar in size and material mix to the power plant from which the CO_2 is captured. (Van der Giesen 2008) Therefore, in addition to the above derived steel demand, the appropriate LCI data from the coal fired power plant are used to represent the PCC installation. These numbers represent estimates. However, a sensitivity analysis, presented below, shows that these inputs only marginally contribute to the overall LCA impacts of the system.

Construction of the CO_2 compressor

Specific Life cycle inventory data for a CO_2 compressor capable of compressing 1,35 MTonnes per year to a pressure of 130 bar is not available. We roughly based the CO_2 compressor in our model on a MAN 25 MW RG80 compressor(MAN 2014) that is capable of compressing CO_2 from a pressure of 1 bar to 200 bar, with a displacement of ~50 kg /s and weight of 60 tonnes.(MAN Diesel & Turbo) To include the complete life cycle of the compressor we scale all inputs to an available 300 kW air compressor, weighing 4600 kg. A detailed LCA inventory is available for such a compressor in the ecoinvent database(Steiner and Frischknecht 2007). Using the weights of the equipment needed, we multiply the inputs and emissions of the air compressor with a factor 60,000/4,600 = 13.

As a lifetime for the compressor unit we assume 15 years. With a total displacement of 50 kg/s or 50x 60x60x4000=$7.2x10^5$ tonne CO_2/year or 1.1 x 10^7 tonne CO_2 over a life time, the PCC installation thus needs $1/1.1x10^7 = 9.26x10^{-8}$ compressor units / tonne CO_2 for compressing the CO_2. A sensitivity analysis (0) shows that these inputs only marginally contribute to the overall LCA impacts of the system.

Monoethanolamine (MEA) consumption and related emissions

To capture CO_2 from the flue gases, a 30% MEA solution is used (American Physical Society 2011; Volkart et al. 2013; Koiwanit et al. 2013; Singh et al. 2012; Fadeyi et al. 2013; Rochelle et al. 2011). Because of thermal degradation and oxidation of the MEA in the capture process, MEA is consumed and needs to be supplemented (Rochelle 2009; Rochelle et al. 2011). In literature MEA consumption is reported over a range from 0,5 – 3,0 kg MEA / tonne CO_2 (American Physical Society 2011; Volkart et al. 2013; Koiwanit et al. 2013; Singh et al. 2011; Fadeyi et al. 2013; Van der Giesen 2008; Koornneef et al. 2008). We therefore use 1,5 kg MEA / tonne CO_2 captured and carry out a sensitivity analysis presented below.

Due to the degradation of MEA, ammonia forms and is emitted to the atmosphere. Ammonia emissions of 0,136 kg NH_3 / tonne CO_2 (Koiwanit et al. 2013) and 0,035 kg NH_3 / tonne CO_2 have been reported (Fadeyi et al. 2013; Singh et al. 2011). Here we choose to use 0,035 kg NH_3 / tonne CO_2. In addition, MEA itself is emitted to the atmosphere for which values of 0,014 – 0,063 kg MEA / tonne CO_2 have been reported (Corsten et al. 2013). Here we estimate that 0,0385 kg MEA / tonne CO_2 is emitted.

Other PCC related requirements and emissions

Table 4.4 gives an overview of other reported material flows related to PCC that are also taken into account in this study.

Table 4.4: Additional, minor material flows of post-combustion capture (Koiwanit et al. 2013; Singh et al. 2011; Fadeyi et al. 2013).

IN: Sodium hydroxide	0.13 kg NaOH/tonne CO_2 captured
IN: Activated carbon	0.075 kg activated carbon / tonne CO_2 captured
IN: Water	0.8 m3 / tonne CO_2 captured
OUT: Formaldehyde	0.262 g / tonne CO_2 captured
OUT: Acetaldehyde	0.167 g / tonne CO_2 captured

CO_2 pipeline (PCC)

The LCA model is used to obtain the total CO_2 quantity that is captured from the flue gases per year. This number determines the size of the required pipeline. To dimension the pipeline we calculate transport of 1,35 MT CO_2/ year over 200 km. This is an estimated, typical distance for CCS, but a sensitivity analysis in which the distance is doubled shows a very limited change in outcomes of the model. Longer distances will increase the environmental impacts of the CO_2 transport because of larger material demands and energy use. Using McCoy's model (Mccoy 2008; Mccoy and Rubin 2008) the transport of this amount of CO_2 over a distance of 200 km requires a NPS 12 line pipe. His model shows an NPS line pipe with an outer diameter of 0,324 m and an inner diameter of 0,310 m, leaving a wall thickness of 0,007 m. According to the American Petroleum Institute (API) a pipeline with this specifications weighs 109,2 kg/m or 109,2 tonne / km (American Petroleum Institute (API) 2004). This number is used to adjust the ecoinvent process that "pipeline, natural gas, high pressure distribution network" [EI ID1339] that produces pipelines with an assumed lifetime of 40 years. Only the amount of reinforced steel per km pipeline is adjusted according to the numbers provided by APS. With an expected lifetime of 40 years and a transportation quantity of 1,35 MT / year, the transport of 1 tonne CO_2 over 200km needs 200 / ($1{,}35 \times 10^6 \times 40$) = $3{,}7 \times 10^{-6}$ km pipeline / tonne CO_2 transported.

Humidity swing-based direct air capture (HS-DAC)

Inventory data (consumption of water, hardware, and sorbent per CO_2 captured) in this study is derived from previously published laboratory experiments and respective publications (Lackner 2009; Wang et al. 2011, 2013; Lackner et al. 2012; Lackner 2013; Wang et al. 2016). An overview is presented in Table 4.5, the unit process is shown in Figure 4.9, and details are explained and derived in subsequent sections.

Table 4.5: LCI data for HS-DAC [resource demand for CO2 captured and delivered @ 100 bar].

	Cold	Standard	Warm
[RH = relative humidity]	10°C/60%RH	20°C/30%RH	30°C/10%RH
Electricity demand [kWh/tonne CO_2] (see section 1.1)	456	378	382
Sorbent consumption [kg/tonne CO_2]	2.02	1.52	1.52
Water consumption [m^3/tonne CO_2]	20.2	15.2	15.2
Hardware consumption [kg/tonne CO_2]	0.701	0.526	0.527
DAC equipment weight [tonne/1-tonne-a-day unit]	5.12	3.84	3.85
Land use [m^2/1-tonne-a-day unit]	155	160	166

N.B. All calculations, modelling results and graphs in the main document are based on "standard" conditions. Where appropriate, climate differences are taken into account, using the values for "cold" and "warm".

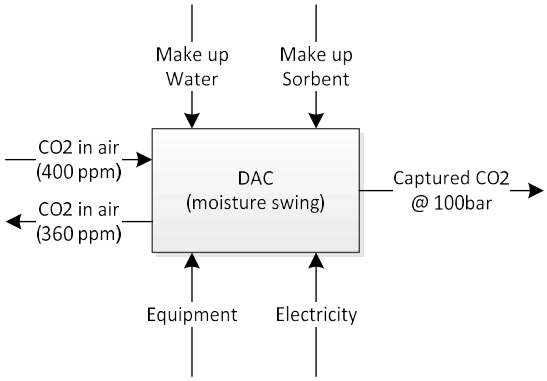

Figure 4.9: Unit process of DAC.

HS-DAC sorbent consumption

The sorbent used is a heterogeneous, quaternary amine, anionic exchange membrane. This sorbent is an approximate 50:50 blend (by mass) of polypropylene plastic and a quaternary anionic exchange resin like Marathon-A as produced by Dow Chemical. Marathon A is a non-hazardous chemical (Avantor Performance Materials 2011) that consists of 50% water and 50% Benzenemethanamium (CAS60177-39-1). Inventory data for Benzenemethanamium chemical is not available and since it can be considered to only have minor hazards (Avantor Performance Materials 2011) it is not taken into account in the LCA (cut-off). Thus, only the polypropylene portion of the HS-DAC sorbent is taken into account. However, a sensitivity analysis using MEA as a proxy for the Benzenemethanamium is available below. The sorbent is re-used and only a small fraction that reaches the end of its lifetime needs to be supplemented. Extrapolating from observations in the laboratory over 10+ years, we estimate a lifetime of the sorbent of 100.000 cycles. At a mass yield of 0.66% per cycle this is equivalent to 1,52 kg sorbent consumed per tonne CO_2 captured.

HS-DAC water consumption

At normal, not further optimized operation (Lackner 2009; Wang et al. 2011, 2013), 0,1kg water is applied per kg sorbent per cycle (to change sorbent kinetics to the 100% humidity regime). This water is subsequently lost to the environment via evaporation. At a mass yield of 0,659% per cycle this is equivalent to 15,2m^3 water per tonne CO_2 captured (= 15,2 tonne of water). Because of the nature of the sorbent, this water does not have to be high-purity. The only restriction is that the water must not have high concentrations of anions such as chloride, sulfates or nitrates because anions would contaminate the resin, reducing its yield over time (Lackner et al. 2012). To remain conservative, in our model we use tap water (higher LCA impacts).

Equipment and materials for HS-DAC

Based on laboratory scale prototypes and tests, a hypothetical industrial scale sized HS-DAC installation is defined here. This installation is capable of capturing 1 tonne of (> =99% pure) CO_2 per day @ 100 bar (see main text for sensitivity analysis to weather conditions). The installation has an assumed lifetime of 20 years (365 days/year). Over its lifetime a HS-DAC installation will capture 20 x 365 x 1 = 7300 tonne CO_2. For 1 tonne CO_2 in the DAC process we need 1 / 7300 = 1,37 x 10^{-4} installations. For capturing larger amounts of CO_2, the installation can be scaled up proportionally. Neither energy nor other consumptions (per tonne CO_2) inherently depend on scaling laws such as surface-to-volume or structural performance and are thus largely independent of a DAC's unit envisioned daily capacity.

The types of materials used and the complexity of manufacturing the HS-DAC installation are assumed similar to that of a passenger vehicle (i.e., steel, advanced plastics, pumps and compressors, electronics, actuators, electric motors, etc.). Here we use inventory data of a passenger vehicle as a proxy for the DAC installation. A 1 tonne-a-day installation weighs 3,84 tonnes (see Table 4.6) and assuming the material composition and distribution of a passenger car we can model the DAC installation (scaled proportionally by weight). In the ecoinvent 2.2 database a Golf A4 passenger car is modelled (EI-ID 1936), this car weighs around 1200 kg (1,2 tonne) (Volkswagen 2014). For modelling the DAC installation we need therefore 3,84/1,2=3,2 of these passenger cars. (5,12/1,2=4,3 for cold weather conditions, 3,85/1,2=3,2 for warm weather conditions).

Table 4.6: DAC installation weight (standard climate 20ºC, 30% relative humidity).

Total sorbent mass in a '1 tonne / day' HS-DAC unit	5.06	tonne
Sorbent enclosures (mass calculated as 10% of sorbent mass) to form filter elements	0.50	tonne
Filter elements (sorbent and enclosures)	**5.56**	**tonne**
Conveyor (calculated as 10% of filter elements)	0.56	tonne
Desorption chamber (calculated as 100% of the filter elements inside desorption chamber), but only 50% of all elements in desorption mode at any given time: 5.56 tonne·50%	2.78	tonne
Total hardware of '1 tonne/day' HS-DAC unit (excluding sorbent)	**3.84**	**tonne**

PV electricity

In this study PV electricity is used to power DAC (DAC PV scenario) and to make up for the electricity lost when applying PCC to a coal fired power plant (PCC PV scenario). This PV electricity is generated by multicrystaline-Si solar cells (panel efficiency = 13,2%, average annual yield = 820 kWh/kW_{peak}) for which the data is taken from the Ecoinvent 2.2. database. Details on this system can be found in ecoinvent report 6-XII (Jungbluth et al. 2007).

Environmental impact assessment

Baseline categories for the CML 2001 impact assessment method(CML-Department of Industrial Ecology 2015) are taken into account in this study. The category scores are displayed in Table 4.7. The impact category climate change is adjusted to include CO_2 captured from the atmosphere, which is usually excluded in the CML2001 impact assessment method. The impact category water depletion is taken from the RECIPE impact assessment method (Goedkoop et al. 2009).

Table 4.7: Impact assessment scores per kWh of electricity delivered to the grid.

Impact Category	Base Case	MEA-PCC$_{coal}$	HS-DAC$_{coal}$	MEA-PCC$_{PV}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DACPV (ZeroGHG)	MEA-PCC$_{coal}$ +DAC$_{PV}$ (ZeroGHG)	PV electricity	Unit
Eutrophication	1.97E-03	2.47E-03	2.70E-03	2.03E-03	2.97E-03	2.05E-03	2.49E-03	1.85E-04	kg PO4-Eq
Abiotic resource depletion	6.48E-03	8.13E-03	8.88E-03	6.64E-03	9.78E-03	6.70E-03	8.19E-03	4.53E-04	kg antimony - Eq
Acidification	1.60E-03	2.07E-03	2.21E-03	1.73E-03	2.44E-03	1.74E-03	2.10E-03	2.98E-04	kg SO_2-Eq
Photochemical oxidation	4.89E-05	6.32E-05	6.85E-05	5.45E-05	7.60E-05	5.71E-05	6.54E-05	1.86E-05	kg ethylene-Eq
Terrestrial ecotoxicity	7.85E-04	1.05E-03	1.11E-03	9.34E-04	1.23E-03	9.76E-04	1.10E-03	4.48E-04	kg 1.4-DCB-Eq
Marine aquatic ecotoxicity	1.04E+03	1.29E+03	1.42E+03	1.07E+03	1.56E+03	1.09E+03	1.31E+03	1.46E+02	kg 1.4-DCB-Eq
Freshwater aaquatic ecotoxicity	2.97E-01	3.72E-01	4.08E-01	3.09E-01	4.49E-01	3.16E-01	3.77E-01	4.56E-02	kg 1.4-DCB-Eq
Stratospheric ozone depletion	6.88E-09	9.28E-09	9.67E-09	1.02E-08	1.07E-08	1.18E-08	1.06E-08	1.38E-08	kg CFC-11-Eq
Human toxicity	2.40E-01	3.56E-01	3.32E-01	3.15E-01	3.66E-01	2.94E-01	3.70E-01	1.46E-01	kg 1.4-DCB-Eq
Climate change	8.48E-01	2.29E-01	2.29E-01	1.97E-01	~0	~0	~0	6.20E-02	kg CO2-Eq
Water depletion	2.07E-03	3.34E-03	1.88E-02	2.75E-03	2.51E-02	1.72E-02	7.40E-03	3.03E-04	m^3

Baseline categories for the CML 2001 impact assessment method(CML-Department of Industrial Ecology 2015) are taken into account in this study. The category scores are displayed in Table 4.7. The impact category climate change is adjusted to include CO_2 captured from the atmosphere, which is usually excluded in the CML2001 impact assessment method. The impact category water depletion is taken from the RECIPE impact assessment method (Goedkoop et al. 2009).

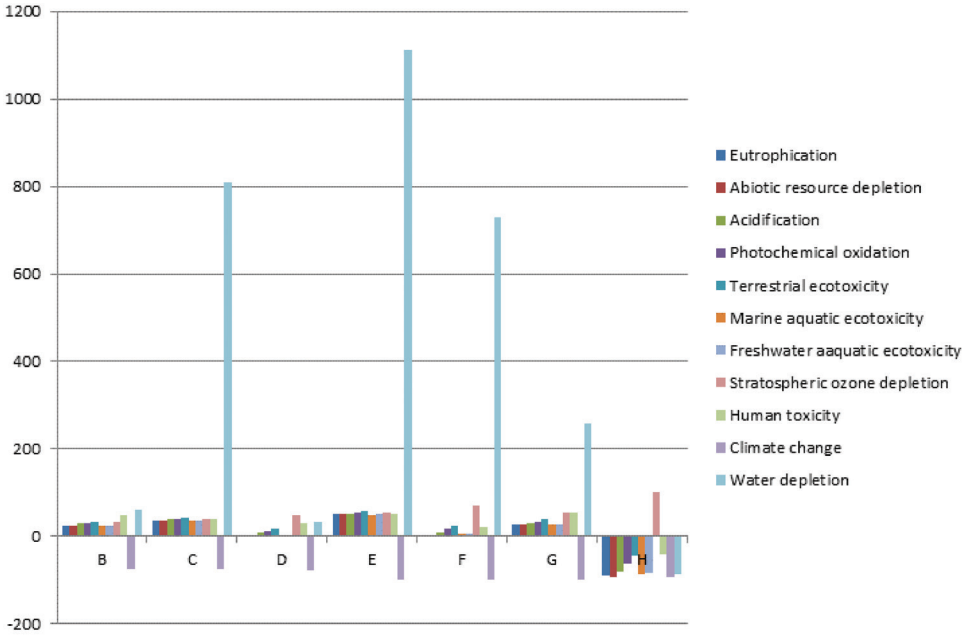

Figure 4.10: LCA results: %Change of impact categories across all scenarios relative to the base case. (B=MEA-PCC$_{coal}$, C=HS-DAC$_{coal}$, D=MEA-PCC$_{PV}$, E=HS-DAC$_{coal}$(ZeroGHG), F=HS-DAC$_{PV}$ (ZeroGHG), G=MEA-PCC$_{c\ al}$+HS-DAC$_{PV}$, H= PV electricity).

Figure 4.10 shows an overview of all impacts in relation to the base case and allows for direct comparison of the average increase in non-GHG impacts with the net life cycle GHG emissions. The numbers for Figure 4.10 LCA results: %Change of impact categories across all scenarios relative to the base case.

Table 4.8: LCA results: %Change of impact categories across all scenarios relative to the base case.

Impact Category	Base Case	MEA-PCC$_{coal}$	HS-DAC$_{coal}$	MEA-PCC$_{PV}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DAC$_{PV}$ (ZeroGHG)	MEA-PCC$_{coal}$ +DAC$_{PV}$ (ZeroGHG)	PV electricity
Eutrophication	0	25	37	3	51	4	26	-91
Abiotic resource depletion	0	25	37	2	51	3	26	-93
Acidification	0	29	38	8	53	9	31	-81
Photochemical oxidation	0	29	40	11	55	17	34	-62
Terrestrial ecotoxicity	0	34	41	19	57	24	40	-43
Marine aquatic ecotoxicity	0	24	37	3	50	5	26	-86
Freshwater aquatic ecotoxicity	0	25	37	4	51	6	27	-85
Stratospheric ozone depletion	0	35	41	48	56	72	54	101
Human toxicity	0	48	38	31	53	23	54	-39
Climate change	0	-73	-73	-77	-100	-100	-100	-93
Water depletion	0	61	808	33	1113	731	257	-85

Table 4.9: Change in Impact assessment scores per net kg CO_2-eq avoided (base case equals zero)
Impact score per kg CO_2 avoided = ($Impact_{scenario}$ – $Impact_{basecase}$)/($GHG_{basecase}$-$GHG_{scenario}$).

Impact Category	MEA-PCC$_{coal}$	HS-DAC$_{coal}$	MEA-PCC$_{PV}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DAC$_{PV}$ (ZeroGHG)	MEA-PCC$_{coal}$ +DAC$_{PV}$ (ZeroGHG)	PV electricity	
Eutrophication	8.08E-04	1.18E-03	9.22E-05	1.18E-03	9.43E-05	6.13E-04	-2.27E-03	kg PO4-Eq / kg CO_2-eq avoided
Abiotic resource	2.67E-03	3.88E-03	2.46E-04	3.89E-03	2.59E-04	2.02E-03	-7.67E-03	kg antimony-Eq/kg CO_2-eq avoided
Acidification	7.59E-04	9.85E-04	2.00E-04	9.91E-04	1.65E-04	5.90E-04	-1.66E-03	kg SO2-Eq/kg CO_2-eq avoided
Photochemical oxidation	2.31E-05	3.17E-05	8.60E-06	3.20E-05	9.67E-06	1.95E-05	-3.85E-05	kg ethylene-Eq/kg CO_2-eq avoided
Terrestrial ecotoxicity	4.28E-04	5.25E-04	2.29E-04	5.25E-04	2.25E-04	3.71E-04	-4.29E-04	kg 1,4-DCB-Eq/kg CO_2-eq avoided
Marine aquatic ecotoxicity	4.04E+02	6.14E+02	4.61E+01	6.13E+02	5.90E+01	3.18E+02	-1.14E+03	kg 1,4-DCB-Eq/kg CO_2-eq avoided
Freshwater aquatic ecotoxicity	1.21E-01	1.79E-01	1.84E-02	1.79E-01	2.24E-02	9.43E-02	-3.20E-01	kg 1,4-DCB-Eq/kg CO_2-eq avoided
Stratospheric ozone	3.88E-09	4.51E-09	5.10E-09	4.50E-09	5.80E-09	4.39E-09	8.80E-09	kg CFC-11-Eq/kg CO_2-eq avoided
Human toxicity	1.87E-01	1.49E-01	1.15E-01	1.49E-01	6.37E-02	1.53E-01	-1.20E-01	kg 1,4-DCB-Eq/kg CO_2-eq avoided
Climate change	-1.00E+00	-1.00E+00	-1.00E+00	-1.00E+00	-1.00E+00	-1.00E+00	-1.00E+00	kg CO_2-Eq/kg CO_2-eq avoided
Water depletion	2.05E-03	2.70E-02	1.04E-03	2.72E-02	1.78E-02	6.29E-03	-2.25E-03	m3/kg CO_2-eq avoided

Sensitivity analyses

In addition to the sensitivity analyses reported above (MEA-PCC: current/likely versus future/best case; HS-DAC: effect of temperature and relative humidity) a series of further sensitivity analyses were performed to assess the effect of any uncertainties in our inventory data on the LCA results and the robustness of our conclusions. We found that our conclusions remain valid even when varying inventory data within their respective uncertainties based on ranges reported in the literature. Details are given below.

CO_2 compressor use in post combustion capture

To examine the sensitivity of the model to the CO_2 compressor hardware used in the PCC installation we doubled the inputs of this compressor to the PCC process from $9{,}26 \times 10^{-8}$ compressor / ton CO_2 captured and compressed to $1{,}85 \times 10^{-7}$ compressor / ton CO_2 captured and compressed. This change in inputs shows no noteworthy change (<0,1%) in the outcome of our model (Table 4.10).

Table 4.10: Change in impact scores for sensitivity to CO_2 compressor hardware.

	Original scores		New scores		New/Old	
	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}
Eutrophication	0.00247	0.00203	0.00247	0.00203	100.0%	100.0%
Abiotic resource depletion	0.00813	0.00664	0.00813	0.00664	100.0%	100.0%
Acidification	0.00207	0.00173	0.00207	0.00173	100.0%	100.0%
Photochemical oxidation	6.32E-05	5.45E-05	6.32E-05	5.45E-05	100.0%	100.0%
Terrestrial ecotoxicity	0.00105	0.000934	0.00105	0.000935	100.0%	100.1%
Marine aquatic ecotoxicity	1.29E+03	1.07E+03	1.29E+03	1.07E+03	100.0%	100.0%
Freshwater aquatic ecotoxicity	0.372	0.309	0.372	0.309	100.0%	100.0%
Stratospheric ozone depletion	9.28E-09	1.02E-08	9.28E-09	1.02E-08	100.0%	100.0%
Human toxicity	0.356	0.315	0.356	0.315	100.0%	100.0%
Climate change	0.229	0.197	0.229	0.197	100.0%	100.0%
Water depletion	0.00334	0.00275	0.00334	0.00275	100.0%	100.0%

Physical size of the PCC plant

The capture of 1 tonne of CO_2 with PCC requires $2{,}46 \times 10^{-8}$ PCC plants. This number is based on estimations of the materials required. Here we check the sensitivity of our LCA model with regard to this parameter by increasing its value by 10% to $1{,}1 \times 2{,}46 \times 10^{-8} = 2{,}71 \times 10^{-8}$ and checking how much the outcomes of the model will change. The model does not show sensitivity to changes in this parameter.

To examine the sensitivity of the model to the PCC plant CO_2 we doubled the inputs of the PCC plant to the PCC process from $2{,}46 \times 10^{-8}$ PCC installation/ton CO_2 captured to $4{,}92 \times 10^{-8}$. This change in inputs results in an only very minor change in the

outcome of our model (Table 4.11).

Table 4.11: Change in impact scores for sensitivity to PCC installation.

	Original scores		New scores		New/Old	
	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}
Eutrophication	0.00247	0.00203	0.00248	0.00203	100.4%	100.0%
Abiotic resource depletion	0.00813	0.00664	0.00815	0.00665	100.2%	100.2%
Acidification	0.00207	0.00173	0.00208	0.00173	100.5%	100.0%
Photochemical oxidation	6.32E-05	5.45E-05	6.42E-05	5.53E-05	101.6%	101.5%
Terrestrial ecotoxicity	0.00105	0.000934	0.00108	0.000961	102.9%	102.9%
Marine aquatic ecotoxicity	1.29E+03	1.07E+03	1.30E+03	1.07E+03	100.8%	100.0%
Freshwater aquatic ecotoxicity	0.372	0.309	0.374	0.31	100.5%	100.3%
Stratospheric ozone depletion	9.28E-09	1.02E-08	9.49E-09	1.03E-08	102.3%	101.0%
Human toxicity	0.356	0.315	0.36	0.318	101.1%	101.0%
Climate change	0.229	0.197	0.232	0.199	101.3%	101.0%
Water depletion	0.00334	0.00275	0.00334	0.00275	100.0%	100.0%

CO_2 pipeline transport distance

With an expected lifetime of 40 years and a transportation quantity of 1,35 MT / year. The transport of 1 tonne CO_2 over 200km needs 200 / (1,35 x10^6 x 40) = 3,7 x 10^{-6} km pipeline / tonne CO_2 transported. To examine the sensitivity of our model we doubled the input of this parameter to 7,4 x 10^{-6} km pipeline / tonne CO_2 transported. This change in inputs shows no noteworthy change in the outcome of our model (Table 4.12).

Table 4.12: Change in impact scores for sensitivity to CO_2 pipeline transportation distance.

	Original scores		New scores		New/Old	
	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}
Eutrophication	0.00247	0.00203	0.00247	0.00203	100.0%	100.0%
Abiotic resource depletion	0.00813	0.00664	0.00813	0.00664	100.0%	100.0%
Acidification	0.00207	0.00173	0.00207	0.00173	100.0%	100.0%
Photochemical oxidation	6.32E-05	5.45E-05	6.35E-05	5.48E-05	100.5%	100.6%
Terrestrial ecotoxicity	0.00105	0.000934	0.00106	0.000942	101.0%	100.9%
Marine aquatic ecotoxicity	1.29E+03	1.07E+03	1.29E+03	1.07E+03	100.0%	100.0%
Freshwater aquatic ecotoxicity	0.372	0.309	0.372	0.309	100.0%	100.0%
Stratospheric ozone depletion	9.28E-09	1.02E-08	9.32E-09	1.02E-08	100.4%	100.0%
Human toxicity	0.356	0.315	0.356	0.315	100.0%	100.0%
Climate change	0.229	0.197	0.23	0.197	100.4%	100.0%
Water depletion	0.00334	0.00275	0.00334	0.00275	100.0%	100.0%

MEA consumption

In literature MEA consumptions in the range of 0,5-3 kg / tonne CO_2 have been reported (see above). The MEA consumption in the PCC scenarios is set to 1,5 kg MEA / tonne CO_2. To examine the sensitivity of our model we doubled the input of this parameter to 3 kg MEA / tonne CO_2. This change in inputs results in only a negligible change in the outcome of our model (Table 4.13).

Table 4.13: Change in impact scores for sensitivity to MEA consumption.

	Original scores		New scores		New/Old	
	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}
Eutrophication	0.00247	0.00203	0.00248	0.00203	100.4%	100.0%
Abiotic resource depletion	0.00813	0.00664	0.00818	0.00668	100.6%	100.6%
Acidification	0.00207	0.00173	0.00209	0.00174	101.0%	100.6%
Photochemical oxidation	6.32E-05	5.45E-05	6.41E-05	5.53E-05	101.4%	101.5%
Terrestrial ecotoxicity	0.00105	0.000934	0.00108	0.000958	102.9%	102.6%
Marine aquatic ecotoxicity	1.29E+03	1.07E+03	1.30E+03	1.07E+03	100.8%	100.0%
Freshwater aquatic ecotoxicity	0.372	0.309	0.373	0.309	100.3%	100.0%
Stratospheric ozone depletion	9.28E-09	1.02E-08	9.69E-09	1.05E-08	104.4%	102.9%
Human toxicity	0.356	0.315	0.408	0.357	114.6%	113.3%
Climate change	0.229	0.197	0.234	0.2	102.2%	101.5%
Water depletion	0.00334	0.00275	0.00334	0.00275	100.0%	100.0%

Hardware requirements of a HS-DAC unit

The actual equipment weight of DAC units has considerable uncertainty because it has been estimated based on laboratory prototypes. The inventory of a passenger car is used as a proxy to model the DAC installation and based on the weight of equipment needed assumptions are made. For the capture of 1 tonne CO_2 from the atmosphere we need 1,37 x 10^{-4} DAC installation. To examine the sensitivity of our model we doubled the input of this parameter to 2,74 x 10^{-4}.

This change in inputs results in no noteworthy change in the outcome of our model (Table 4.14).

HS-DAC sorbent composition

To examine the sensitivity of our model to the cut-off assumption for Benzenemethanamium (see above) we used MEA as a proxy for Benzenemethanamium (1:1 mass ratio).

This change in inputs results in no noteworthy change in the outcome of our model (Table 4.15).

HS-DAC sorbent amount consumed

As average consumption 1,5 kg HS-DAC sorbent per tonne CO_2 captured is modelled. To examine the sensitivity of our model we doubled the input of this parameter to 3 kg sorbent per tonne CO_2 captured.

This change in inputs results in no noteworthy change in the outcome of our model (Table 4.16)

Water use of HS-DAC

The HS-DAC processes consumes 15,2 m^3 water / tonne CO_2 captured. As shown in the impact assessment the water depletion of DAC is a factor 9 higher than for PCC. Water demand therefore seems an important parameter in our model. To examine the sensitivity of our model we doubled the input of this parameter to 30,4 m^3 water per tonne CO_2 captured.

As expected, this change in inputs results in a large change in the outcome of our model for water depletion (but only minimal change for all other environmental impacts). When the input is doubled, the life cycle water depletion changes by 50 – 90% (Table 4.17).

Table 4.14: Change in impact scores for sensitivity to DAC installation size.

Impact Category	Original scores				New scores				New / Old			
	$HS\text{-}DAC_{coal}$	$HS\text{-}DAC_{coal}$ (ZeroGHG)	$HS\text{-}DAC_{PV}$ (ZeroGHG)	$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)	$HS\text{-}DAC_{coal}$	$HS\text{-}DAC_{coal}$ (ZeroGHG)	$HS\text{-}DAC_{PV}$ (ZeroGHG)	$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)	$HS\text{-}DAC_{coal}$	$HS\text{-}DAC_{coal}$ (ZeroGHG)	$HS\text{-}DAC_{PV}$ (ZeroGHG)	$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)
Eutrophication	2.70E-03	2.97E-03	2.05E-03	2.49E-03	2.70E-03	2.97E-03	2.05E-03	2.49E-03	100.0%	100.0%	100.0%	100.0%
Abiotic resource depletion	8.88E-03	9.78E-03	6.70E-03	8.19E-03	8.89E-03	9.80E-03	6.71E-03	8.19E-03	100.1%	100.2%	100.1%	100.0%
Acidification	2.21E-03	2.44E-03	1.74E-03	2.10E-03	2.23E-03	2.46E-03	1.75E-03	2.11E-03	100.9%	100.8%	100.6%	100.5%
Photochemical oxidation	6.85E-05	7.60E-05	5.71E-05	6.54E-05	6.93E-05	7.70E-05	5.78E-05	6.56E-05	101.2%	101.3%	101.2%	100.3%
Terrestrial ecotoxicity	1.11E-03	1.23E-03	9.76E-04	1.10E-03	1.13E-03	1.26E-03	9.94E-04	1.11E-03	101.8%	102.4%	101.8%	100.9%
Marine aquatic ecotoxicity	1.42E+03	1.56E+03	1.09E+03	1.31E+03	1.42E+03	1.57E+03	1.10E+03	1.31E+03	100.0%	100.6%	100.9%	100.0%
Freshwater aquatic	4.08E-01	4.49E-01	3.16E-01	3.77E-01	4.09E-01	4.51E-01	3.17E-01	3.77E-01	100.2%	100.4%	100.3%	100.0%
Stratospheric ozone depletion	9.67E-09	1.07E-08	1.18E-08	1.06E-08	9.77E-09	1.09E-08	1.18E-08	1.06E-08	101.0%	101.9%	100.0%	100.0%
Human toxicity	3.32E-01	3.66E-01	2.94E-01	3.70E-01	3.35E-01	3.70E-01	2.96E-01	3.71E-01	100.9%	101.1%	100.7%	100.3%
Climate change	2.29E-01	~0	~0	~0	2.30E-01	~0	~0	~0	100.4%	x	x	x
Water depletion	1.88E-02	2.51E-02	1.72E-02	7.40E-03	1.88E-02	2.51E-02	1.72E-02	7.41E-03	100.0%	100.0%	100.0%	100.1%

Table 4.15: Change in impact scores for sensitivity to DAC sorbent composition.

Impact Category	Original scores				New scores				New / Old			
	$HS\text{-}DAC_{coal}$	$HS\text{-}DAC_{coal}$ (ZeroGHG)	$HS\text{-}DAC_{PV}$ (ZeroGHG)	$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)	$HS\text{-}DAC_{coal}$	$HS\text{-}DAC_{coal}$ (ZeroGHG)	$HS\text{-}DAC_{PV}$ (ZeroGHG)	$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)	$HS\text{-}DAC_{coal}$	$HS\text{-}DAC_{coal}$ (ZeroGHG)	$HS\text{-}DAC_{PV}$ (ZeroGHG)	$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)
Eutrophication	0.0027	0.00297	0.00205	0.00249	0.0027	0.00297	0.00205	0.00249	100.0%	100.0%	100.0%	100.0%
Abiotic resource depletion	0.00888	0.00978	0.0067	0.00819	0.00889	0.0098	0.00671	0.00819	100.1%	100.2%	100.1%	100.0%
Acidification	0.00221	0.00244	0.00174	0.0021	0.00222	0.00245	0.00174	0.00211	100.5%	100.4%	100.0%	100.5%
Photochemical oxidation	6.85E-05	7.60E-05	5.71E-05	6.54E-05	6.88E-05	7.63E-05	5.73E-05	6.54E-05	100.4%	100.4%	100.4%	100.0%
Terrestrial ecotoxicity	0.00111	0.00123	0.000976	0.0011	0.00112	0.00125	0.000983	0.00111	100.9%	101.6%	100.7%	100.9%
Marine aquatic ecotoxicity	1.42E+03	1.56E+03	1.09E+03	1.31E+03	1.42E+03	1.56E+03	1.10E+03	1.31E+03	100.0%	100.0%	100.9%	100.0%
Freshwater aquatic	0.408	0.449	0.316	0.377	0.408	0.45	0.316	0.377	100.0%	100.2%	100.0%	100.0%
Stratospheric ozone depletion	9.67E-09	1.07E-08	1.18E-08	1.06E-08	9.79E-09	1.09E-08	1.19E-08	1.06E-08	101.2%	101.9%	100.8%	100.0%
Human toxicity	0.332	0.366	0.294	0.37	0.347	0.387	0.308	0.374	104.5%	105.7%	104.8%	101.1%
Climate change	0.229	~0	~0	~0	0.23	~0	~0	~0	100.4%	x	x	x
Water depletion	0.0188	0.0251	0.0172	0.0074	0.0188	0.0251	0.0172	0.0074	100.0%	100.0%	100.0%	100.0%

Table 4.16: Change in impact scores for sensitivity to DAC sorbent demand.

	Original scores				New scores				New / Old			
Impact Category	**$HS\text{-}DAC_{coal}$**	**$HS\text{-}DAC_{coal}$ (ZeroGHG)**	**$HS\text{-}DAC_{PV}$ (ZeroGHG)**	**$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)**	**$HS\text{-}DAC_{coal}$**	**$HS\text{-}DAC_{coal}$ (ZeroGHG)**	**$HS\text{-}DAC_{PV}$ (ZeroGHG)**	**$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)**	**$HS\text{-}DAC_{coal}$**	**$HS\text{-}DAC_{coal}$ (ZeroGHG)**	**$HS\text{-}DAC_{PV}$ (ZeroGHG)**	**$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)**
Eutrophication	0.0027	0.00297	0.00205	0.00249	0.0027	0.00297	0.00205	0.00249	100.0%	100.0%	100.0%	100.0%
Abiotic resource depletion	0.00888	0.00978	0.0067	0.00819	0.0089	0.00981	0.00672	0.00819	100.2%	100.3%	100.3%	100.0%
Acidification	0.00221	0.00244	0.00174	0.0021	0.00222	0.00245	0.00174	0.00211	100.5%	100.4%	100.0%	100.5%
Photochemical oxidation	6.85E-05	7.60E-05	5.71E-05	6.54E-05	6.88E-05	7.64E-05	5.73E-05	6.55E-05	100.4%	100.5%	100.4%	100.2%
Terrestrial ecotoxicity	0.00111	0.00123	0.000976	0.0011	0.00111	0.00123	0.000976	0.0011	100.0%	100.0%	100.0%	100.0%
Marine aquatic ecotoxicity	1.42E+03	1.56E+03	1.09E+03	1.31E+03	1.42E+03	1.56E+03	1.09E+03	1.31E+03	100.0%	100.0%	100.0%	100.0%
Freshwater aquatic	0.408	0.449	0.316	0.377	0.408	0.449	0.316	0.377	100.0%	100.0%	100.0%	100.0%
Stratospheric ozone depletion	9.67E-09	1.07E-08	1.18E-08	1.06E-08	9.67E-09	1.07E-08	1.18E-08	1.06E-08	100.0%	100.0%	100.0%	100.0%
Human toxicity	0.332	0.366	0.294	0.37	0.332	0.366	0.294	0.37	100.0%	100.0%	100.0%	100.0%
Climate change	0.229	~0	~0	~0	0.23	~0	~0	~0	100.4%	x	x	x
Water depletion	0.0188	0.0251	0.0172	0.0074	0.0188	0.0251	0.0172	0.0074	100.0%	100.0%	100.0%	100.0%

Table 4.17: Change in impact scores for sensitivity to DAC water demand.

Impact Category	Original scores				New scores				New / Old			
	$HS\text{-}DAC_{coal}$	$HS\text{-}DAC_{coal}$ (ZeroGHG)	$HS\text{-}DAC_{PV}$ (ZeroGHG)	$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)	$HS\text{-}DAC_{coal}$	$HS\text{-}DAC_{coal}$ (ZeroGHG)	$HS\text{-}DAC_{PV}$ (ZeroGHG)	$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)	$HS\text{-}DAC_{coal}$	$HS\text{-}DAC_{coal}$ (ZeroGHG)	$HS\text{-}DAC_{PV}$ (ZeroGHG)	$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)
Eutrophication	0.0027	0.00297	0.00205	0.00249	0.00271	0.00299	0.00206	0.0025	100.4%	100.7%	100.5%	100.4%
Abiotic resource depletion	0.00888	0.00978	0.0067	0.00819	0.00891	0.00982	0.00673	0.00819	100.3%	100.4%	100.4%	100.0%
Acidification	0.00221	0.00244	0.00174	0.0021	0.00223	0.00247	0.00175	0.00211	100.9%	101.2%	100.6%	100.5%
Photochemical oxidation	6.85E-05	7.60E-05	5.71E-05	6.54E-05	6.96E-05	7.74E-05	5.81E-05	6.57E-05	101.6%	101.8%	101.8%	100.5%
Terrestrial ecotoxicity	0.00111	0.00123	0.000976	0.0011	0.00114	0.00127	0.001	0.00111	102.7%	103.3%	102.5%	100.9%
Marine aquatic ecotoxicity	1.42E+03	1.56E+03	1.09E+03	1.31E+03	1.42E+03	1.57E+03	1.10E+03	1.31E+03	100.0%	100.6%	100.9%	100.0%
Freshwater aquatic	0.408	0.449	0.316	0.377	0.41	0.453	0.318	0.378	100.5%	100.9%	100.6%	100.3%
Stratospheric ozone depletion	9.67E-09	1.07E-08	1.18E-08	1.06E-08	9.90E-09	1.10E-08	1.20E-08	1.06E-08	102.4%	102.8%	101.7%	100.0%
Human toxicity	0.332	0.366	0.294	0.37	0.334	0.37	0.296	0.371	100.6%	101.1%	100.7%	100.3%
Climate change	0.229	~0	~0	~0	0.233	~0	~0	~0	101.7%	x	x	x
Water depletion	0.0188	0.0251	0.0172	0.0074	0.0348	0.0471	0.0323	0.0114	185.1%	187.6%	187.8%	154.1%

Impact of weather conditions on HS-DAC performance

Table 4.18 presents operational parameters for HS-DAC in different weather conditions. Running the LCA model with these parameters shows that under cold and humid (CH) conditions HS-DAC is likely to perform worse than under standard (S) or warm and dry (WD) conditions. There difference between standard and warm dry conditions is minimal (see Table 4.19).

Table 4.18: Operational parameters of HS-DAC under different weather conditions (RH=relative humidity).

Operation mode	**CH**	**S**	**WD**
	Cold / Humid (10°C/60% RH)	**Standard (20°C/30% RH)**	**Warm / Dry (30°C/10% RH)**
Max. sorbent loading (determined by ambient conditions. see section 0)	0.876	0.968	0.985
Min. sorbent loading (tunable)	0.750	0.800	0.817
--> Sorbent mass yield [CO_2 per sorbent]	0.49%	0.66%	0.66%
Sorbent consumption [per tonne CO_2]	2.02 kg	1.52 kg	1.52 kg
Electricity consumption [per tonne CO_2]	456 kWh	378 kWh	382 kWh
Water consumption [per tonne CO_2]	20.2 m^3	15.2 m^3	15.2 m^3
Hardware excl. sorbent [per "1tonne a day" machine] [8]	5.12 tonnes	3.84 tonnes	3.85 tonnes
Hardware consumption [per tonne CO_2]	0.701 kg	0.526 kg	0.527 kg
Land-use [per "1tonne a day" machine]	155 m^2	160 m^2	166 m^2

Table 4.19: Impacts under different weather conditions (Cold. Standard and Warm) for different alternatives.

	MEA-PCC$_{coal}$	**HS-DAC$_{coal}$**	**MEA-PCC$_{PV}$**	**HS-DAC$_{coal}$ (ZeroGHG)**	**HS-DAC$_{PV}$ (ZeroGHG)**	**MEA-PCC$_{coal}$ +DAC$_{PV}$ (ZeroGHG)**	**PV electricity**
COLD							
Eutrophication	25	50	3	68	5	27	-91
Abiotic resource depletion	25	50	2	68	4	27	-93
Acidification	29	52	8	71	11	32	-81
Photochemical oxidation	29	55	11	75	21	35	-62
Terrestrial ecotoxicity	34	57	19	78	30	43	-43
Marine aquatic ecotoxicity	24	49	3	67	7	26	-86
Freshwater aquatic ecotoxicity	25	51	4	69	8	27	-85
Stratospheric ozone depletion	35	56	48	76	86	58	101
Human toxicity	48	52	31	71	28	55	-39
Climate change	-73	-73	-77	-100	-100	-100	-93
Water depletion	61	1195	33	1634	977	322	-85

STANDARD							
Eutrophication	25	37	3	51	4	26	-91
Abiotic resource depletion	25	37	2	51	3	26	-93
Acidification	29	38	8	53	9	31	-81
Photochemical oxidation	29	40	11	55	17	34	-62
Terrestrial ecotoxicity	34	41	19	57	24	40	-43
Marine aquatic ecotoxicity	24	37	3	50	5	26	-86
Freshwater aquatic ecotoxicity	25	37	4	51	6	27	-85
Stratospheric ozone depletion	35	41	48	56	72	54	101
Human toxicity	48	38	31	53	23	54	-39
Climate change	-73	-73	-77	-100	-100	-100	-93
Water depletion	61	808	33	1113	731	257	-85
WARM							
Eutrophication	25	38	3	51	4	26	-91
Abiotic resource depletion	25	38	2	51	3	26	-93
Acidification	29	39	8	53	9	32	-81
Photochemical oxidation	29	41	11	55	17	34	-62
Terrestrial ecotoxicity	34	43	19	58	25	40	-43
Marine aquatic ecotoxicity	24	37	3	50	6	26	-86
Freshwater aquatic ecotoxicity	25	38	4	52	6	27	-85
Stratospheric ozone depletion	35	41	48	56	72	54	101
Human toxicity	48	39	31	53	23	54	-39
Climate change	-73	-73	-77	-99	-101	-100	-93
Water depletion	61	813	33	1103	736	257	-85

In the different alternatives the following values change due to differing weather conditions (see also Table 4.2).

Table 4.20: LCA model lay-out for different scenarios.

	Cold weather scenario	**Warm weather scenario**
HS-DAC_{coal}	captures 1.039 kg CO_2 to have same climate change (CC) impacts as PCC	captures 0.935 kg CO_2 to have same CC impacts as PCC
HS-DACcoal (ZeroGHG)	captures 1.42 kg CO_2 to have no net CC impacts	captures 1.27 kg CO_2 to have no net CC impacts
HS-DAC_{PV} (ZeroGHG)	captures 0.88 kg CO_2 to have no net CC impacts	captures 0.875 kg CO_2 to have no net CC impacts
MEA-PCC_{coal}+DAC_{PV} (ZeroGHG)	captures 0.235 kg CO_2 to have no net CC impacts	captures 0.235 kg CO_2 to have no net CC impacts

4.5.3. Alternative scenario: Changing coal based to PV electricity

While outside the scope of our work, a logical question to ask with respect to the study is what the environmental impact would be if coal-based electricity generation were replaced 1:1 by PV electricity (without any capture). Based on ecoinvent data(Jungbluth et al. 2007) representing multi-crystalline photovoltaic electricity generation we find that replacing coal-based electricity with PV electricity on average reduces non-GHG emissions by over 50% while the GHG-emissions are reduced by over 90%.

It needs to be taken into account here that this calculation assumes only the PV installations. Any required electricity storage to ensure non-intermittent supply is not taken into account.