Environomic optimal design of power plants with CO_2 capture.

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Abstract

Life cycle impact assessment indicators are integrated for studying the process integration of renewable resources and CO_2 capture and storage (CCS) in power plants. Besides the expected reduction in global warming potential (GWP), CCS induces energy and cost penalties. This paper presents a systematic multi-objective optimisation framework for the optimal design of power plants with CO_2 capture considering environomic criteria to systematically assess the trade-off between environmental impacts, efficiency and costs. Life cycle assessment is combined with flowsheeting, energy integration, economic evaluation and multi-objective optimisation techniques. Post- and pre-combustion CO_2 capture options for electricity generation processes, using fossil and renewable resources, are assessed. Multi-objective optimisations are performed for various objectives to reveal the influence on the decision making. The calculated CO_2 emissions, allow to assess the impact of the CO_2 tax, considering not only the on-site emissions but taking into account the overall life cycle of the fuel supply, electricity generation and CO_2 capture. The results show that the environomic optimal process design and competitiveness of CO_2 capture highly depends on the considered environmental impact and on the introduction of a carbon tax.

Keywords: CO_2 capture, Life cycle assessment, Multi-objective optimisation, Process design, Power plants

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Nomenclature

Abbreviations

ATR Autothermal Reforming
BM Biomass
BM pre Biomass fed power plant with pre-combustion CO_2 capture
CC Carbon Capture
CCI Climate Change Impact
CCS Carbon Capture and Storage
EI99 Ecoindicator 99 impact method
EQ Ecosystem Quality
FGR Flue Gas Recirculation
FICFB Fast Internally Circulating Fluidised Bed
FU Functional Unit
GHG Greenhouse Gas
GWP Global Warming Potential
HH Human Health
HP High Pressure
Imp Impact 2002+ method
IPCC International Panel on Climate Change
LCA Life Cycle Assessment
LCI Life Cycle Inventory
LCIA Life Cycle Impact Assessment
LP Low Pressure
MEA Monoethanolamine
MILP Mixed Integer Linear Programming
MOO (moo) Multi-Objectif Optimisation
Nb Number
NG Natural Gas
NGCC Natural Gas Combined Cycle
NG pre Natural gas fed power plant with pre-combustion CO_2 capture
NG post Natural gas fed power plant with post-combustion CO_2 capture
PSA Pressure Swing Adsorption
Res Resources
RME Rape Methyl Ester
WGS Water Gas Shift

Greek letters

Δh^o Lower heating value, kJ/kg

ϵ_{tot}	Energy efficiency, %
η_{CO2}	CO_2 capture rate, % or -
Roman letters	
COE	Electricity production cost, \$/GJ$_e$
$\dot{E}$	Mechanical/electrical power, kW$_e$
$\dot{m}$	Mass flowrate, kg/s
$\dot{n}$	Molar flowrate, kmol/s
$\dot{Q}$	Heat, kW
Subscripts	
cc	Plant with carbon capture
ref	Reference plant without carbon capture
Superscripts	
$^+$	Material/energy stream entering the system
$^-$	Material/energy stream leaving the system

1. Introduction

Carbon capture and storage (CCS) is regarded as a promising measure to reduce the greenhouse gas emissions. For CO_2 capture in power plants three different concepts can be distinguished: post-, pre- and oxy-combustion. Post-combustion CO_2 capture consists in the end-of-pipe separation of the CO_2 from the flue gas of fuel combustion, while in oxy-fuel combustion pure oxygen is used for the combustion resulting in a flue gas containing mainly CO_2 and water which is removed by condensation. In pre-combustion CO_2 capture the CO_2 is separated after the gasification and reforming of fuel and the remaining H_2 is used in a gas turbine to generate electricity. Different technologies can be applied for separating the CO_2 the most common ones being based on absorption principles (Olajire (2010)).

The competitiveness of these options depends on the power cycle, the resources, the capture technology and the economic scenario. Previous studies made for European ZEP (2011) and OECD countries Finkenrath (2011) have mainly focused on technology and economy issues, which is a crucial part but not sufficient for decision making with regard to sustainable development. Several studies have investigated the environmental impacts of CCS. The review of existing LCA literature made by Corsten et al. (2013) gives a good insight into environmental impacts of CCS chains and highlights the large variation in reported data. For CCS, the different literature data indicate reductions of the greenhouse gas (GHG) emissions of 65-84%

for pulverized hard coal-fired power plants and of 47-80% for natural gas fired plants corresponding to an absolute GWP of 22-76$kg_{CO2,eq}/GJ_e$ and of 21-68$kg_{CO2,eq}/GJ_e$ respectively. Zapp et al. (2012) identified that the energy penalty, the capture efficiency and the fuel type have a significant impact on the environmental effects of CCS. A change in the capture rate of ± 5% results in a variation of the GWP of ± 20%. The trade-off between the global warming potential (GWP) and other environmental impacts is revealed by Pehnt and Henkel (2009) for coal power plants and by Singh et al. (2011) for different CCS options in natural gas and coal power plants. Singh et al. (2011) state for NGCC plants with pre-combustion CO_2 capture a reduction in GWP of 64%, an increase in terrestrial acidification of 20% and in human toxicity of 62%. The LCA analysis of Volkart et al. (2013) included biomass based CCS options and Viebahn et al. (2007) compared the impacts of CCS with the one of renewables. For biomass based power plants the GWP can become negative with CCS which means that more GHG emissions are removed from than emitted to the atmosphere assuming sustainable usage of biomass (ranging from -40 to -320 $kg_{CO2,eq}/GJ_e$ Volkart et al. (2013)).

So far, only reduced multi-criteria assessments were applied to power plants with CCS. When comparisons are made, they are mostly made for a given process design. Multi-objective optimisation of the process design with regard to objectives resulting from a rigorous life cycle assessment (LCA) such as presented by Bernier et al. (2010) is rarely performed.

Therefore, the objective of this paper is to systematically compare and optimise different CO_2 capture options taking into account thermodynamic, economic and environmental considerations simultaneously. The process design is optimised in terms of operating conditions and energy integration. In Tock and Maréchal (2013) the systematic methodology for thermo-environomic modelling and optimisation presented by Gerber et al. (2011) has already been applied to assess the competitiveness of CO_2 capture options for natural gas (NG) and biomass (BM) fed power plants. This optimisation focused on the minimisation of the energy penalty and of the local CO_2 emissions (i.e. maximisation of the captured CO_2). However, since there is a trade-off between GWP and other environmental impacts (i.e. resources depletion), different life cycle impact objectives will be considered here in order to reveal the influence on the environomic optimal process design and on the decision making.

2. Methodology

To make a consistent evaluation of different post- and pre-combustion CO_2 capture process options for electricity generation with regard to environmental, economic and energetic criteria a systematic methodology is applied. For each process option the same design and performance evaluation principles are applied and the same assumptions are made, which allows to make a thorough competitiveness assessment on a common basis. The applied thermo-environomic optimisation methodology follows the one previously presented (Gassner and Maréchal, 2009a; Gerber et al., 2011; Tock and Maréchal, 2012a) combining flowsheeting and energy integration techniques with economic evaluation and life cycle assessment in a multi-objective optimisation framework (Figure 1).

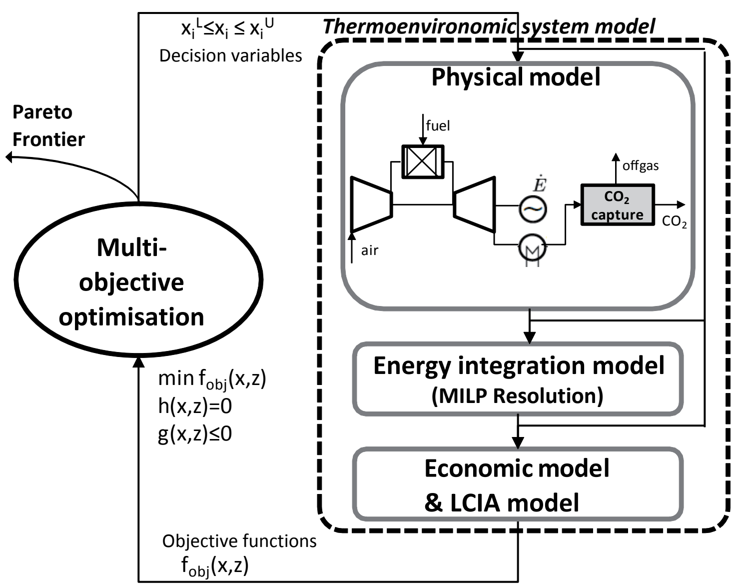

Figure 1: Multi-objective optimisation methodology for environomic optimal process design.

After having summarised potential candidate technologies in a superstructure, flowsheeting models are developed for each unit operation option based on literature data in order to compute the chemical and physical transformations and to identify the process heat transfer requirements. The processes are modelled with the conventional flowsheeting software Belsim Vali and Aspen Plus. The maximal heat recovery and the optimal utility integration are then computed in the energy-integration model by applying energy integration techniques (i.e. pinch analysis) solving the heat cascade problem to close the thermal energy balance as explained in Maréchal and Kalitventzeff (1998). Using the data from the flowsheet and process integration models, the costs are estimated based on equipment sizing and cost correlations from literature (Turton, 2009; Ulrich and Vasudevan, 2003) and the environmental impacts are evaluated by applying the LCA technique. Finally, the trade-off between the competing objectives, like investment and life cycle emissions or energy efficiency, is assessed by multi-objective optimisation simultaneously optimising several objectives with regard to the decision variables (i.e. technology selection and operating conditions). Applying an evolutionary algorithm (Molyneaux et al. (2010)) implemented in Matlab® the Pareto frontiers are generated and the values of the decision variables defined. Evolutionary algorithms working with populations instead of a single data point, do not generate one single optimal solution but multiple promising solutions in the form of a Pareto-optimal frontier. The Pareto-optimal solutions correspond to the configurations for which it is not possible to improve one objective without simultaneously downgrading one of the other objectives. In contrast to multi-criteria evaluations which compare given solutions (i.e. process designs), the gain of multi-objective optimisation is to generate the best solutions. The advantage of including the process integration model and the life cycle assessment model in the design process is that the influence of the design and operation is reflected on the thermo-environomic performance of the energy balanced system. This allows to make a systematic comparison of CO_2 capture options in power plants applications.

2.1. Process description and modeling

Different pre- and post-combustion CO_2 capture options for electricity generation using natural gas (NG) or woody biomass (BM) as a resource are investigated and illustrated in Figure 2. Oxy-fuel combustion processes and coal fed power plants are not considered in this study. However, based on the energy integration analysis made in Urech et al. (2014) these options could be included following the same approach.

The three representative CO_2 capture options that are studied are:

- Post-combustion CO_2 capture by chemical absorption with monoethanolamine (MEA) applied to a natural gas combined cycle (NGCC) plant. The plant size is defined by the thermal input of natural gas being in the order of 582 $MW_{th,NG}$. This option is abbreviated hereafter as *NG post-*. The post-combustion CO_2 capture process is the same as the one described in Tock and Maréchal (2014).

- Pre-combustion CO_2 capture by physical absorption with Selexol in a natural gas fueled power plant based on autothermal reforming. The plant size is defined by the thermal input of natural gas of 725 $MW_{th,NG}$. This option is hereafter referred to as *NG pre-*. The natural gas based pre-combustion CO_2 capture process models have been described and analysed previously in Tock and Maréchal (2012b) and Tock and Maréchal (2012d) for H_2 production applications.

- Pre-combustion CO_2 capture by physical absorption with Selexol in a biomass fired power plant based on fast internally circulating fluidised bed gasification. The plant size is defined as 380 $MW_{th,BM}$. The biomass plant's scale is limited by the biomass availability and the logistics of wood transport, as explained in Gerber et al. (2011). The biomass resource is wood characterised by a weight composition of 51.09%C, 5.75%H, 42.97%O and 0.19% N, and a humidity of 50%wt. This option is hereafter labeled as *BM pre-*. The biomass based pre-combustion CO_2 capture process models have been described and analysed previously in Tock and Maréchal (2012c).

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For all the cases CO_2 compression to 110 bar for subsequent transport and storage is included (Belsim Vali model) to evaluate the thermo-environomic performance. However, the storage itself being beyond the scope of this study is not accounted for. The decision variables for the investigated pre- and post-combustion processes are reported in Tables 2 and 3.

2.2. Process modeling

The process models are developed with the conventional flowsheeting software (Aspen Plus for the CO_2 capture unit and Belsim Vali for the other process units) based on common operating conditions reported in literature.

2.2.1. Natural gas reforming

The autothermal reforming reactor is modeled as an isothermal reactor assuming that the reforming with water, the partial oxidation with air and

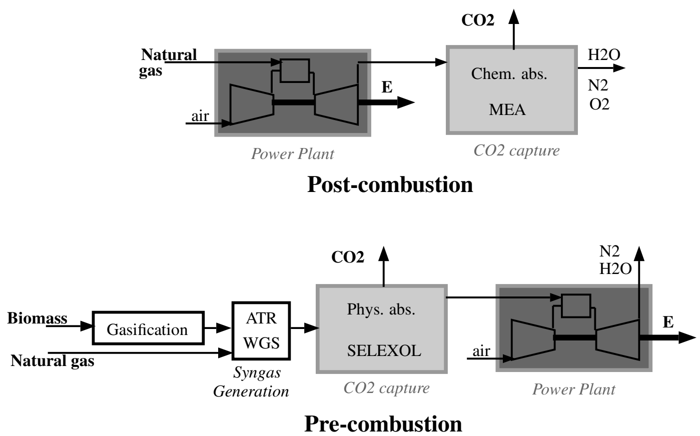

Figure 2: Investigated CO_2 capture process options.

the water gas shift reactions reach thermodynamical equilibrium defined by the reaction temperature. The H_2 and CO_2 content is increased after the reformer by a dual shift reactor modeled as isothermal reactor following the approach outlined in Marechal et al. (2005) applying the minimum exergy losses representation. This modeling approach allows to decouple the heat transfer from the chemical reaction heat and consequently to maximise the energy recovery for power generation. After CO_2 removal (section 2.2.3), the H_2 is purified by pressure swing adsorption modeled based on the approach of Gassner and Maréchal (2009b) with data for H_2/CO_2 separation from Jee et al. (2001). The purity and the amount of H_2 and CO_2 recovered in the respective outlet streams is essentially defined by the PSA cycle design, namely the durations of the adsorption, recycling and purging periods. The H_2-rich fuel is fed to a gas turbine for heat and power generation. The main operating conditions are reported in Table 3.

2.2.2. Biomass gasification

The major process steps are wood drying, indirectly heated fast internal fluidised bed gasification (FICFB) with steam oxidant, gas cleaning, gas treatment by reforming and water gas shift followed by H_2 separation and purification as described in Tock and Maréchal (2012c). The chemical conversion in the gasifier operating at around 0.1 MPa and 1000 K is modeled by equilibrium relationships with an artificial temperature difference as explained in Gassner and Maréchal (2009b). After the gasification the syngas is treated in two sequential water gas shift (WGS) reactors to increase the H_2 and CO_2 concentrations before CO_2 removal. The gas treatment and purification technologies are the same as in the natural gas fed process. The high temperature heat required for the gasification and the reforming is satisfied by the combustion of process off gas and if necessary by burning part of the process syngas. The main operating conditions are reported in Table 3.

2.2.3. CO_2 capture model

The chemical absorption with monoethanolamine model is based on the one presented in Bernier et al. (2010) and Tock and Maréchal (2014). In the thermodynamic model, the electrolyte NRTL method is used for the liquid phase and the Redlich-Kwong method for the vapour phase. The absorber and desorber are modelled in Aspen Plus as rate based RadFrac columns including reaction kinetics. The CO_2 capture rate is defined by the columns design (i.e. number of stages, diameter, etc.) and the operating conditions summarised in Table 2. The major drawback of chemical absorption is large energy requirement for the solvent regeneration which is in the range of 1.5-3.4 GJ/t_{CO2} (Metz et al. (2005)).

Compared to chemical absorption the thermodynamic modelling of the physical absorption with Selexol is less complex since no ions are involved and no chemical reactions take place in the absorber/desorber. The model is adapted from the default models for physical solvents available from *AspenTech*. To model the thermo-physical properties the PC-SAFT equation of state model for vapour pressure, liquid density, heat capacity and phase equilibrium is used. The absorber is modelled as a RadFrac column and the desorber as a single stage flash unit. The CO_2 capture rate is defined by the flowrate of the lean solvent and the columns design. The main decision variables of the physical absorption process are reported in Table 3.

2.3. Thermo-economic performance

The thermo-economic performance is evaluated based on the following indicators. The energy efficiency ϵ_{tot} is defined by the ratio between the net electricity output ($\Delta\dot{E}^- = \dot{E}^- - \dot{E}^+$) and the resources energy input (Eq. 1). The reported efficiencies are expressed on the basis of the lower heating value.

$$\epsilon_{tot} = \frac{\Delta\dot{E}^-}{\Delta h^o_{feed,in} \cdot \dot{m}_{feed,in}} \quad (1)$$

The CO_2 capture rate η_{CO2} is expressed by the molar ratio between the captured CO_2 and the carbon entering the system (Eq. 2). The CO_2 capture rate depends on the process design, especially on the operating conditions and on the design of the absorber and desorber units. The CO_2 capture rate is based on the local CO_2 emissions and does not account for all the CO_2 emissions from resource extraction and transportation which are evaluated in the LCA (section 2.4).

$$\eta_{CO2} = \frac{\dot{n}_{CO2,captured}}{\dot{n}_{C,in}} \cdot 100 \qquad (2)$$

The electricity production costs (COE) include the annual capital investment and the operation and maintenance costs. The capital investment of each equipment is update to year 2013 with the Marshall and Swift cost index accounting for inflation. The total capital investment is annualised taking into account the interest rate and the plant lifetime. The maintenance costs are assumed to be 5% of the initial annual investment. The operating costs mainly consist of the purchase of the resources, which are here the natural gas and biomass feedstock. The resource price is based on the price of natural gas reported by ZEP (2011). A sensitivity analysis is made in section 3.2.2 to reveal the influence of the resource price. If indicated a carbon tax on local or life cycle CO_2 emissions (i.e. tax CO_2 local / LCA) is accounted in the COE. The influence of the carbon tax is studied in section 3.2.1. The economic assumptions are summarised in Table 1.

Table 1: Definition of the economic the economic assumptions.

Parameter	Value
Yearly operation [h/y]	7500
Economic lifetime [y]	25
Interest rate [%]	6
Marshall & Swift Index^{2003} [-]	1473.3
Resource price [$/GJ$_{res}$]	9.7

The CO_2 capture cost is evaluated by the CO_2 avoidance costs, which are expressed in Eq.3 by the difference of the local CO_2 emissions and the difference of the total production cost with regard to a reference plant without CO_2 capture. The competitiveness is compared with a conventional NGCC plant (559 $MW_{th,NG}$) without CO_2 capture yielding an efficiency of 58.8 % (Table 4). All the reported cost data refer to year 2013.

$$\$/t_{CO2,avoided} = \frac{COE_{CC} - COE_{ref}}{\dot{m}_{CO_{2,emitted_{ref}}} - \dot{m}_{CO_{2,emitted_{CC}}}} \frac{[\$/GJ]}{[t_{CO2}/GJ]} \quad (3)$$

2.4. Life cycle assessment model

With regard to CO_2 emissions mitigation, an assessment of the overall life cycle environmental impacts from the resource extraction along the production chain to the final product, including off-site emissions and construction emissions, is essential. Life cycle assessment (LCA), standardised in ISO 14040 & 14044, has been proven to be suitable for this scope. In this study the objective of the LCA is to get life cycle impact assessment (LCIA) indicators which reflect the influence of the system design on the performance and allow to identify the environomic optimal process design. Therefore, the life cycle inventory (LCI) is expressed as a function of the characteristics of the thermo-economic model (i.e. design variables, mass and energy balances, equipment size) following the adapted LCA methodology for the conceptual design presented by Gerber et al. (2011). The four main stages of LCA are the mandatory ones of the ISO-norm: the goal and scope definition, the life cycle inventory, the impact assessment and the interpretation.

2.4.1. Goal and scope definition

The scope of this study is to evaluate and compare the environmental performance accounting for the whole life cycle from cradle-to-grave of different configurations of power plants with CO_2 capture for a wide range of environmental impacts not only limited to the GWP but as well accounting the impacts on human health, ecosystem quality and resources depletion. Therefore, the functional unit (FU) is defined as 1 GJ_e of net electricity produced by the plant. The expected lifetime of the plant is assumed to be 25 years.

2.4.2. Life cycle inventory

In the LCI phase every flow, crossing the system boundaries as an extraction or an emission, which is necessary to one of the unit processes, is identified and quantified based on the thermo-economic model. For the process equipments of the thermo-economic model, the methodology presented in

Gerber et al. (2011) is used for a non-linear impact scaling. The LCI model is illustrated in Figure 3 for typical pre-combustion CO_2 capture processes. For each LCI element, the data available from the ecoinvent® database (Ecoinvent, 2013) are used to compute the different contributions of the process modelled in this study. The major process steps are resource extraction and transport, heat and power generation and CO_2 removal. The inventory is made for the European /Swiss context. The main inputs are the feedstocks (natural gas and biomass), the MEA for CO_2 capture, the auxiliary materials for the gasification and gas cleaning (olivine, sorbalit, rape methyl ester, calcium carbonate, limestone) and the catalysts for the reforming and water gas shift (zinc, nickel and aluminum oxide catalysts). For natural gas, the standard natural gas mix for Switzerland transported by long distance pipeline mainly from Germany, Russia, Norway and the Netherlands is considered. The biomass is assumed to be a mix of soft and hardwood residues from European forests transported to the plant by diesel trucks of 28 t having a capacity of 40 m^3. The average distance for the wood transport is linked to the plant size and corresponds for a plant of 350 $MWth_{BM}$ to 88 km. The main emissions and wastes are the combustion products CO_2, NOx and particle matter, and the MEA degradation losses which are assumed to be 1.6 kg_{MEA}/t_{CO2}.

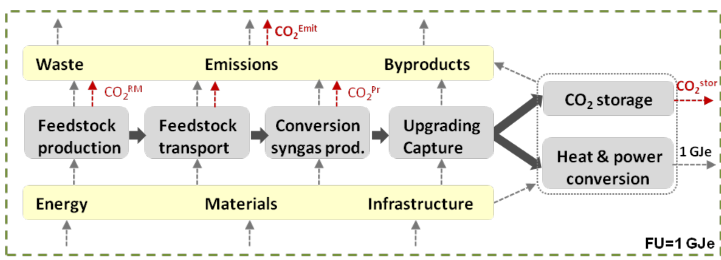

Figure 3: System's boundary for life cycle inventory of pre-combustion CO_2 capture processes.

The impacts of the CO_2 transport and storage are not included in the inventory because it is not known exactly where and how this will be done, as the technology is still in development and there are a lot of uncertainties, especially with regard to the environmental consequences. This simplification is justifiable for this comparative study, as the specific impact per kg of CO_2 captured will be equal in all the cases.

2.4.3. Life cycle impact assessment

In the LCIA step the environmental impact is computed by aggregating the vector of the different elementary flows of emissions and extractions obtained for each element of the LCI in indicators of environmental significance, termed as impact categories. The aggregation is performed with an impact assessment method, which is a matrix containing the weightings for the elementary flows. In this study, different impact methods are compared to address the influence on greenhouse gas emissions, ecosystem, human health and resources. The method of the International Panel on Climate Change (IPCC) 2007 (IPCC (2007)) is used to calculate the global warming potential in terms of equivalent CO_2 emissions on a 100 years time-horizon. It has to be noted that the GWP of fossil CO_2 emissions is standardised to 1, while for biogenic CO_2 emissions the GWP is considered as 0. Storage of fossil CO_2 accounts as zero to GWP, while storage of biogenic CO_2 leads to a GWP of -1. The negative balance is due to the fact that the released CO_2 was previously fixed in the plant as hydrocarbon by photosynthesis. In addition to the climate change impact (CCI), the impacts on resources (Res), human health (HH) and ecosystem quality (EQ) are evaluated by the Impact 2002+ method (endpoint categories) and the damage-oriented Ecoindicator-99-(h,a) method (hierarchist perspective, single score) . In the Ecoindicator-99 method (Goedkoop and Spriensma (2000)) climate change is accounted in the human health impact aggregating also carcinogenic, ozone layer depletion and respiratory effects. The respective weighting factors are for the Ecoindicator-99 method 40 % HH, 40 % EQ and 20 % Res.

2.5. Multi-objective optimisation

The decision variables for the optimisation are the process operating conditions (i.e. T and P of the process units, design of ab- and desorption columns). The details are reported in Tables 2 and 3. Four different multi-objective optimisation problems are considered to study the influence of the environmental objective on the environomic optimal process design. In each multi-objective optimisation problem, the efficiency is maximised, the electricity production costs are minimised and the environmental impact (assessed by different LCA indicators) are minimized. The three objectives are simultaneously optimised without applying any weighting or normalisation.

- MOO CO_2 capt.: max ϵ_{tot}, max η_{CO2}
- MOO GWP: max ϵ_{tot}, min GWP $kg_{CO2,eq}/GJ_e$, min COE

- MOO EI99: max ϵ_{tot}, min total impact Ecoindicator-99, min COE
- MOO Imp.: max ϵ_{tot}, min total impact Impact 2002+, min COE

Table 2: Decision variables for the post-combustion CO_2 capture process using chemical absorption process with monoethanolamine.

Operating parameter	Range
FGR [-]	[0-0.56]
Lean solvent CO_2 loading [kmol/kmol]	[0.18-0.25]
Rich solvent CO_2 loading [kmol/kmol]	[0.4-0.5]
Rich solvent pre-heat T [^oC]	[95-105]
Rich solvent re-heat T [^oC]	[115-125]
LP stripper pressure [bar]	[1.7-2.1]
HP / LP pressure ratio [-]	[1-1.5]
MEA % in solvent [-]	[0.3-0.35]
Absorber steam out [kg_{H2O}/t_{FG}]	[306-309.5]
Split fraction [-]	[0-0.7]
Nb stages absorber	[10-17]
Nb stages HP stripper	[8-15]
Nb stages LP stripper	[6-10]
Absorber diameter [m]	[6-12]
HP stripper diameter [m]	[3-6]
LP stripper diameter [m]	[2-5]

3. Results

3.1. Base case configurations

Three base case configurations are first analysed and compared to a state of the art natural gas combined cycle. For the post- and pre-combustion capture in natural gas combined cycles a capture rate of 90 % CO_2 is considered, while a CO_2 capture rate of 60 % is considered for the biomass based processes. The thermo-environomic performance are evaluated considering an average Swiss - European context. For the biomass process a lower capture rate is chosen to get a good compromise between efficiency and capture rate, higher capture rates being difficult to reach due to the intrinsic inefficiency of the biomass conversion process. It has however to be highlighted that the carbon captured in the case of the biomass fed processes is biogenic carbon which leads de facto to a reduction of the CO_2 concentration in the atmosphere. Table 4 summarises the thermo-environomic performance. The results show that for natural gas fed power plants, CO_2 capture induces an

Table 3: Decision variables for the pre-combustion CO_2 capture (physical absorption with Selexol solvent) processes using natural gas or biomass as a feedstock.

Section	Specification	Range
Biomass drying	T [K]	473
Biomass pyrolysis	T [K]	533
Biomass gasification	$\theta_{wood,gasif\_in}$ [%wt]	[5-35]
	T [K]	[1000-1200]
	P [bar]	[1-15]
SMR after gasification	T [K]	[950-1200]
ATR	T [K]	[780-1400]
	P [bar]	[1-30]
	S/C [-]	[0.5- 6]
WGS	T_{HTS} (NG/BM) [K]	[523-683]/[573-683]
	T_{LTS} (NG/BM) [K]	[423-523]/[423-573]
	P (BM) [bar]	[1-25]
	S/C (BM) [-]	[0.2-4]
CO_2 capture	DEPG/CO_2 ratio [kg/kg]	[8-14]
	Absorber T [^oC]	[-18-173]
	Absorber P [bar]	[10-60]
	Nb stages absorber	10
	Absorber packing	Pall ring
	Regeneration P [bar]	[1-10]
	Regeneration T [^oC]	[25-100]

energy penalty of 6-9 percentage points and a cost penalty of about 5-6 $\$/GJ_e$ yielding CO_2 avoidance cost around 60-66 $\$/t_{CO2,avoided}$. The penalty of CO_2 capture is explained by the additional cost and energy consumption for CO_2 capture (4-7 %) and compression (2 %).

3.1.1. Environmental performance

The life cycle environmental performance assessed with different impact methods is illustrated in Figures 4 and 5. The detailed contributions buildup of the impact categories (HH, Res, EQ) assessed with the Ecoindicator-99 method are reported in Appendix Figures 12-14.

With regard to the climate change impact assessed with the IPCC 2007 method (Figure 4) the benefit of capturing CO_2 is clearly seen compared to a plant without CO_2 capture. With a capture rate of 90 %, the GWP is reduced to 34 $kg_{CO2,eq}/GJ_e$ with post-combustion CO_2 capture compared to a

Table 4: Thermo-economic performance of the base case configurations.

Process	NGCC	NG post-	NG pre-	BM pre-
Feed [$MW_{th,NG/BM}$]	559	587	725	380
CO_2 capture [%]	0	89.5	89.1	59
ϵ_{tot} [%]	58.75	49.6	52.6	34.8
	Power Balance			
Net electricity [MW_e]	328	291	375	132
$\dot{E}^{+}_{Consumption}$ [$MJ_e/GJ_{e,net}$]	-	108.3	146.6	342.4
$\dot{E}^{-}_{SteamNetwork}$ [$MJ_e/GJ_{e,net}$]	340.7	341.3	177.6	346.2
$\dot{E}^{-}_{GasTurbine}$ [$MJ_e/GJ_{e,net}$]	659.3	767	969	996.2
	Economic Performance			
Invest. [$\$/kW_e$]	555	909	813	3880
COE no CO_2 tax [$\$/GJ_e$]	18.3	23.7	24.5	49.5
Avoidance costs [$\$/t_{CO2,avoided}$]	-	60	66	113
	Environmental Performance			
Local CO_2 emissions [kg_{CO2}/GJ_e]	105	14.9	11.5	0
IPCC GWP [$kg_{CO2,eq}/GJ_e$]	120	34	31.9	-134.2
EI99 [pts/GJ_e]	7.48	7.7	8.1	6.1
Impact 2002 [$10^{-3}pts/GJ_e$]	28.9	20.8	22.4	3.2

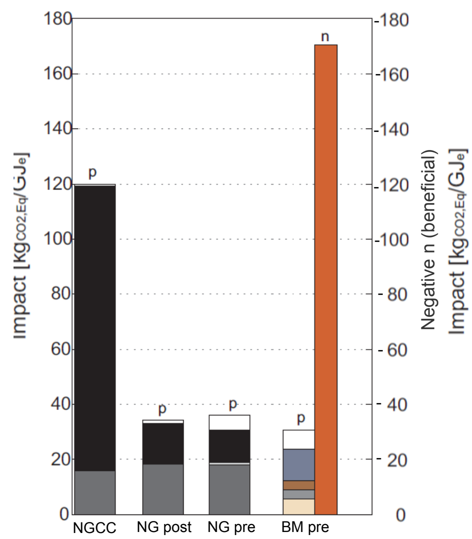

Figure 4: Comparison of the climate change impact based on the impact method IPCC 07. Positive (harmful) contributions are label with a p, and negative (beneficial) contributions are labeled with an n.

conventional NGCC plant (120 $kg_{CO2,eq}/GJ_e$). Pre-combustion CO_2 capture (60 %) in biomass fed power plants even leads to a negative balance of -135 $kg_{CO2,eq}/GJ_e$ due to the advantage of capturing biogenic CO_2. In Figure 4 the positive emissions of the plant are distinguished from the negative contributions related to the CO_2 captured from the atmospheric CO_2 by the photosynthesis during the biomass production. For the natural gas fed processes the major contributions to the greenhouse gas emissions are coming from the natural gas extraction and transport, and from the uncaptured CO_2. With CO_2 capture, the contribution from the natural gas is sligthly larger because of the lower power plant efficiency. Due to the energy demand for CO_2 capture and compression, the natural gas consumption is increased to produce 1 GJ of electricity compared to a conventional NGCC having a higher productivity.

With the Impact 2002+ method, the benefit of capturing CO_2 is also revealed (Figure 5). The overall environmental impact of the power plants with CO_2 capture is lower than for the plants without capture due to the reduced climate change impact, even if the resources impact is increased. However, with the Ecoindicator-99 method, the overall impact of CO_2 capture in a NGCC plant is 3 % higher than without capture because of the impact on the depletion of fossil resources. In this method the resources impact overweights the climate change benefit (included in the human health impact). For natural gas fed processes, the largest impact is coming from the resources depletion followed by the human health and the ecosystem. For CO_2 capture in a biomass fed power plant the overall impact is however lower than for the reference plant without CO_2 capture, even if the impact on the ecosystem is much more important. The impact on the ecosystem is large, due to the extraction of a renewable resource and due to the contribution of the rape methyl ether (RME) used in the syngas cleaning step. When using palm biodiesel instead of RME, the ecosystem impact could be reduced by 35%. This results from the Ecoinvent data reporting 2.08 $kg_{CO2,eq}/kg_{RME}$ and 1.71 $kg_{CO2,eq}/kg_{PalmOil}$ respectively for the GWP assessed with the IPCC method.

It is interesting to note that with respect to the selected environmental indicator, the CO_2 capture options on fossil fuel have a higher impact then the configurations without CO_2 capture. This is explained by the decrease of the process efficiency that translates into a higher consumption of resources to produce the same amount of electricity. This highlights the difficulty of the single score life cycle assessment methods where the weighting factors may create biases in the analysis. This also stresses on the need of conduct-

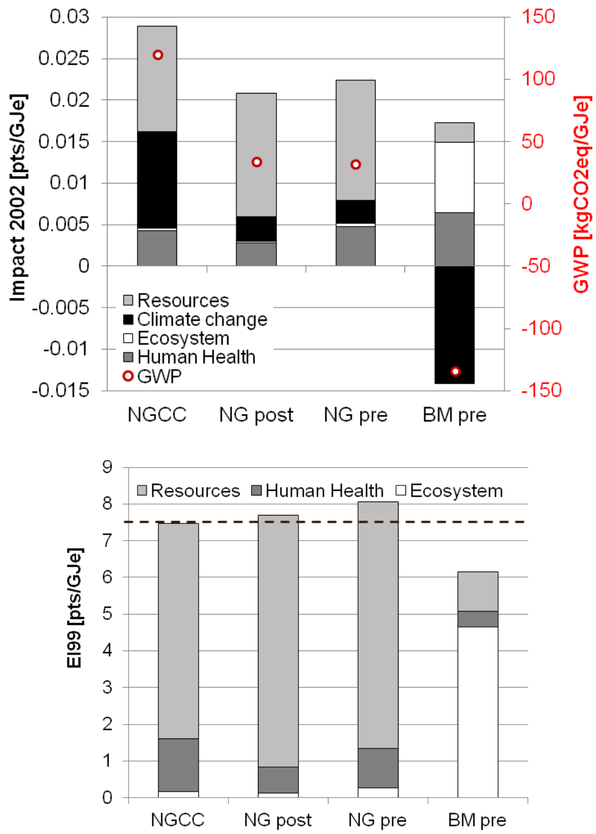

Figure 5: Environmental impacts comparison for base case power plant designs without and with CO_2 capture. Top: Impact method Impact 2002+, Bottom: Impact method Ecoindicator-99 (h,a).

ing multi-objective optimisation strategies, if such indicators are used for optimising the system design as the choice of the impact method influences the CO_2 capture options performance evaluation and thus on the selection of the optimal process design.

3.2. Thermo-environomic optimisation

In order to see the influence of the choice of the environmental objective on the environomic optimal process design, different multi-objective optimisations, defined in Section 2.5, are performed. The trade-off between the competing objectives is illustrated by the Pareto frontiers in Figure 6.

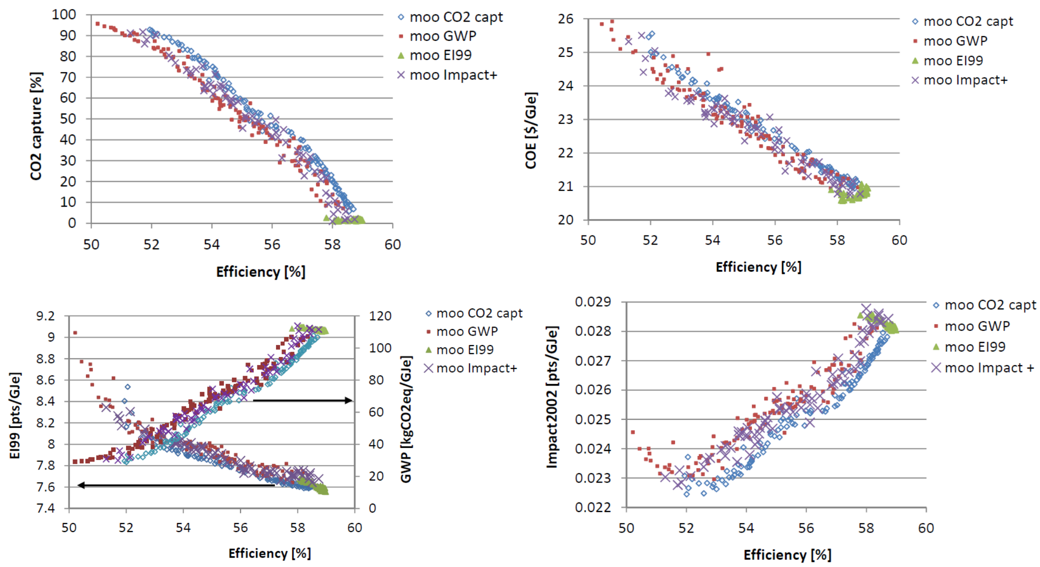

Figure 6: Influence of the objective function on the Pareto optimal solutions for the natural gas fed power plant with pre-combustion CO_2 capture: Thermo-economic and environmental trade-off.

The efficiency and cost penalty of CO_2 capture can clearly be seen (Figure 6 top). At high capture rates the efficiency is decreased by around 8 percentage points and the COE is increased by $5\$/GJ_e$ due to the additional energy consumption and equipments for CO_2 capture and compression. In terms of environmental performance, the opposite behaviour between the Ecoindicator-99 and GWP impact is clearly revealed for the option of a natural gas fed plant with pre-combustion CO_2 capture. Optimising local CO_2 emissions or the GWP or the total impact assessed with the Impact 2002+ method leads to the same process designs. That is to say that the assessed process operating conditions (i.e. the decision variables values) are the same for the same CO_2 capture rate, which leads consequently also to the same performance. However, when minimising the Ecoindicator-99 total impact, the optimisation leads to solutions which yield high efficiencies and low CO_2 capture rates (i.e. high emissions). This trend can be explained by the increased impact on the resources which overweights the decreased impact on the human health (incl. climate change) at high capture rates. The same conclusions can be drawn from the optimisation results of the post-combustion CO_2 capture process. While for CO_2 capture in biomass fed power plants, the optimisation with the objective function Ecoindicator-99 impact leads to the same optimal solutions as with the other impact methods, because the capture of biogenic CO_2 results in a decrease of the environmental burdens assessed with any of the three impact methods.

3.2.1. Carbon tax influence

To evaluate the economic competitiveness of each process design generated by the optimisation and to support decision making, the impact of the introduction of a carbon tax on the local CO_2 emissions and on the whole life cycle CO_2 emissions is assessed. Figure 7 reveals that for low CO_2 taxes process designs with high GWP (i.e. low η_{CO2}, high ϵ_{tot}) lead to the lowest COE, while for taxes higher than 50 $/t$_{CO2}$ process designs with low GWP become profitable.

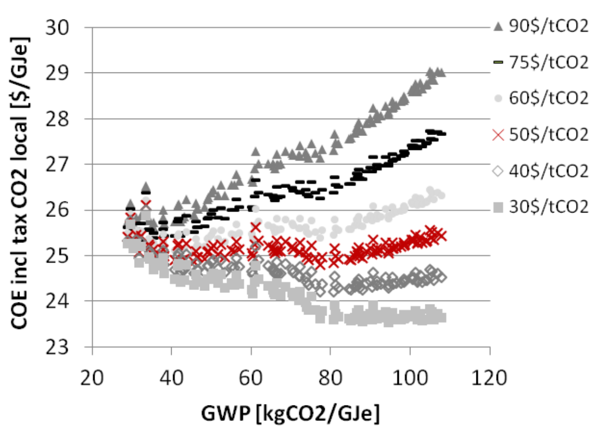

Figure 7: Influence of carbon tax on the COE of the natural gas fed power plant with pre-combustion capture.

For a given carbon tax, the process design yielding the lowest COE (incl. tax) has been identified from all the Pareto optimal solutions generated for the the post- and pre-combustion options fed with biomass or natural gas. The results are illustrated in Figure 8 which highlights also the break-even carbon tax for which the CO_2 capture becomes competitive compared to a conventional NGCC plant. The slope change is related to a switch of the optimal process design with CO_2 capture. The decrease in COE (incl. tax CO_2 LCA) after the maximum is due to a transition of the resource from natural gas to biomass. The performance results of the most competitive processes designs for different carbon tax values are reported in Table 5 and Figures 9 and 10.

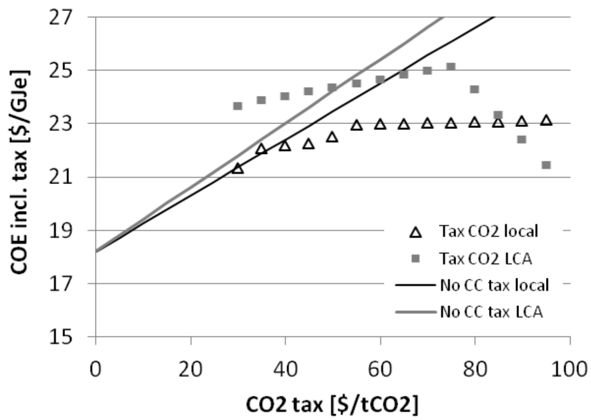

Figure 8: Influence of carbon tax on the COE of the most economically competitive process identified among all the investigated options (NG-post, NG-pre, BM-pre).

Table 5: Performance of the optimal process designs yielding the lowest COE (Figures 9 and 10).

	tax local CO_2				tax LCA CO_2		
Carbon tax [$/t$_{CO2}$]	30	35	50	55	30	35	80
Process	NG pre-	NG pre-	NG pre-	NG post-	NG post-	NG post-	BM pre-
CO_2 capture rate [%]	1.2	33.6	38.8	83.9	76.6	83.9	71.9
Efficiency [%]	58.2	56.8	56.5	50.6	51.6	50.5	39.3
COE incl. tax [$/GJ$_e$]	21.3	22.1	22.5	23.0	23.7	23.9	24.9

With a carbon tax up to 50 $\$/t_{CO2}$ on the local CO_2 emissions, pre-combustion designs with capture rates up to 38 % are competitive, while post-combustion capture with high capture rates becomes interesting for taxes above 55 $\$/t_{CO2}$. Figure 9 shows the reduction of the climate change impact with the increasing tax, leading to a lower overall environmental impact evaluated with the Impact 2002+ and IPCC method and a slightly higher one with the Ecoindicator-99 method due to the resources impact. If a tax is introduced on the life cycle CO_2 emissions, then high capture rates (80 % post-combustion) reducing the climate change impact (Figure 10) already become competitive for low taxes 30-75 $\$/t_{CO2}$, while for higher taxes biomass processes emerge due to the environmental benefit of capturing biogenic CO_2.

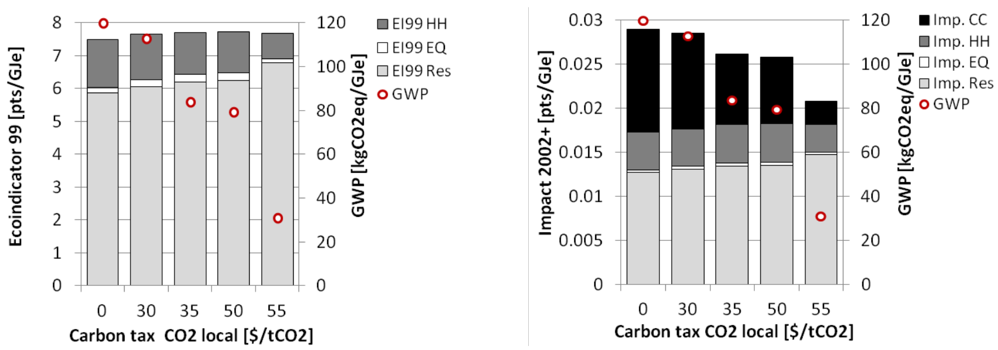

Figure 9: Environmental impact of the process designs with the lowest COE including a tax on the local CO_2 emissions (Table 5).

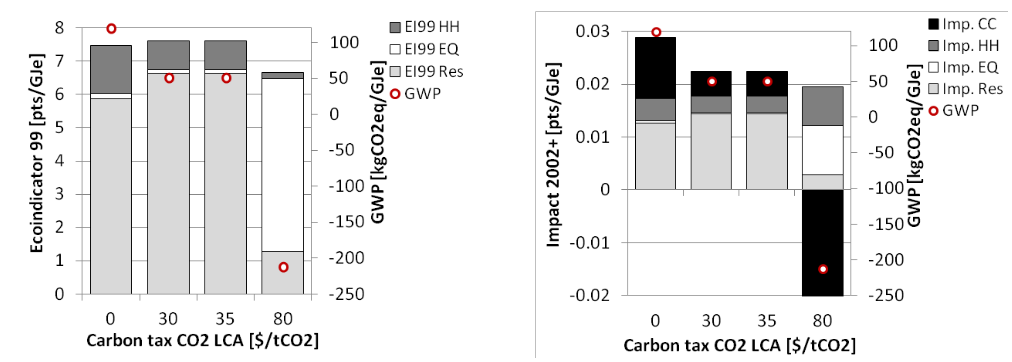

Figure 10: Environmental impact of the process designs with the lowest COE including a tax on the life cyle CO_2 emissions (Table 5).

These results reveal that the environomically optimal process design is highly influenced by the introduction of a carbon tax.

3.2.2. Resource price influence

The environomic optimal process design is not only influenced by the introduction of a carbon tax but also by the resource price. In the previous analysis it was assumed that the biomass and the natural gas price are the same (9.7 $\$/GJ_{res}$). However, if in the future biomass becomes available at a lower price (6.5 $\$/GJ_{BM}$) and the natural gas price increases (10 $\$/GJ_{NG}$), the competitiveness of CO_2 capture in power plants will be influenced. Using these resource prices, the COE of the pareto solutions obtained for the different objective functions have been recalculated and the optimal environomic process design has been identified for different carbon taxes. Figure 11 shows the influence of the economic conditions on the optimal process design. With a natural gas price of 10 $\$/GJ_{NG}$ the break-even carbon tax (on local CO_2 emissions) for which carbon capture becomes competitive with conventional NGCC plants is 35 $\$/t_{CO2}$. Under these conditions biomass fed processes emerge as being the best environomic solution for a carbon tax above 80$\$/t_{CO2}$. These results highlight the influence of the resource price and of the introduction of a carbon tax on the competitiveness of carbon capture.

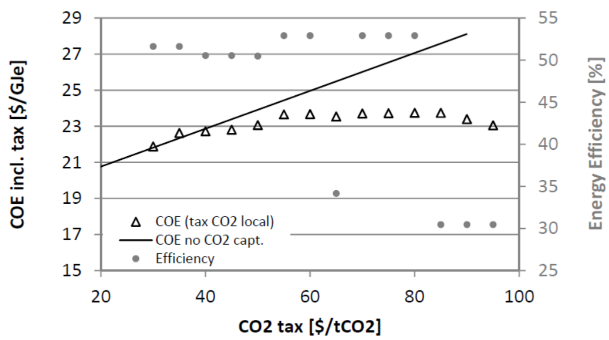

Figure 11: Influence of carbon tax on the COE (incl. tax local CO2) and efficiency of the most economically competitive process (resource price 6.5 $\$/GJ_{BM}$ and 10 $\$/GJ_{NG}$) identified among all the investigated options (NG-post, NG-pre, BM-pre).

3.3. Comment on the use of the LCIA for the process design and process comparison

The use of life cycle assessment indicators for the design of processes has to be considered with care, since it relies on the quality of the inventory data and on the acceptance of the weighting factors used in the calculation of the indicators. The use of LCIA indicators in the optimisation is a new application of such indicators, which are most of the time used for comparing different scenarios. Here the design decisions are taken as a function of the selected objectives and therefore depend on sensitivity of such parameters. As a consequence, the evaluation methodology should be revisited to consider the impact of these assumptions not only on the performance indicator value of the process configurations but also on their sensitivity on the decision variables. The uncertainty of the inventory data needs to be *considered in the optimisation strategies. Provided that the uncertainty distributions are available in the inventory data bases (as it is the case in the ECOINVENT data base), optimisation under uncertainty like the stochastic programming approaches (Dubuis and Maréchal (2012)) or uncertainty analysis method (Tock and Maréchal (2015)) should therefore be considered to select the most probable best options in the Pareto front generated by the multi-objective optimisation. In such context, the comparison of process options should then be based on probability tests and each solution should be reported with an error bar. Adopting a life cycle impact assessment approach introduces the question of the substitution options and allocation assumptions in the process design boundary conditions. Supply chains of feedstocks and equipments become therefore part of the optimisation problem and should be considered as decision variables. This requires extending the system boundaries up to the decision scope of the engineers in charge of the design. As a consequence, not only mean values from the observed market have to be used in the system design, but more precise values allowing to distinguish the suppliers should be used. This would therefore require a comprehensive approach where each supplier will be advertising its own LCIA indicators using certified and validated methodologies that make the data comparable. Finally, in the proposed approach, Life Cycle Inventory data are used to estimate the impact of a carbon tax on the resource supply chain. This assumes that the supply chain will not be affected by the carbon tax and that business as usual operation will be continued. This to some extend contradicts the principle of the approach, since engineers responsible for the processes in the supply chain could apply the same methodology to optimise the processing steps to reduce the* CO_2 *emissions and so limit the economic impact of the carbon tax. This would mean that the system boundaries should be extended and that mitigation options should be included into the supply chain model as decision options (Bernier et al. (2013)).*

4. Conclusion

Different CO_2 capture options using natural gas and biomass resources are systematically compared and optimised in terms of energetic, economic and environmental considerations. By including LCA impacts as an objective function in the multi-objective optimisation it is highlighted how the environmental target influences the environomic optimal process design and consequently the decision making. Different impact methods are compared to address the influence on greenhouse gas emissions, ecosystem, human health and resources depletion. It is interesting to note that different endpoint indicators lead to different conclusions. With the Ecoindicator-99-(h,a) method the environmental impact of natural gas fed power plants with CO_2 capture appears to be worse than without capture because of the larger resources depletion impact, related to the energy penalty, over-weighting the climate change benefit aggregated in the human health impact. When the climate change impact is accounted in a separate impact category as in the Impact 2002+ and IPCC impact methods, CO_2 capture shows a clear environmental benefit. The introduction of a carbon tax favours power plants with CO_2 capture. For a tax on the local CO_2 above 50 $\$/t_{CO2}$, natural gas power plants with 80 % post-combustion capture are the most competitive and allow to reduce the GWP by around 75 % to 31 $kg_{CO2,eq}/GJ_e$. Biomass plants become competitive with a tax on the life cycle CO_2 emissions around 80 $\$/t_{CO2}$ and lead to a negative GWP of -187 $kg_{CO2,eq}/GJ_e$. Consequently, the optimal CO_2 capture process design highly depends on the chosen impact method to evaluate the environmental impact and on the introduction of a carbon tax. To complete the evaluation of CCS options, the impact of the CO_2 transport and storage has to be included in a future study and the sensitivity analysis of the economic parameters has to be extended.

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Appendix

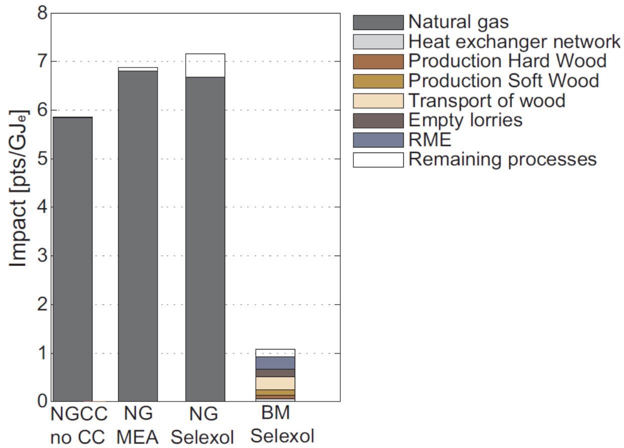

Figure 12: Contributions to the resources impact based on the impact method Ecoindicator-99 (h,a).

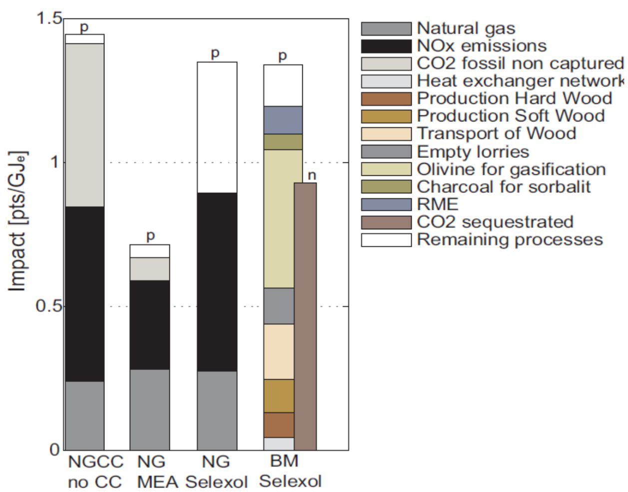

Figure 13: Contributions to the human health impact based on the impact method Ecoindicator-99 (h,a).Positive (harmful) contributions are label with a p, and negative (beneficial) contributions are labeled with an n.

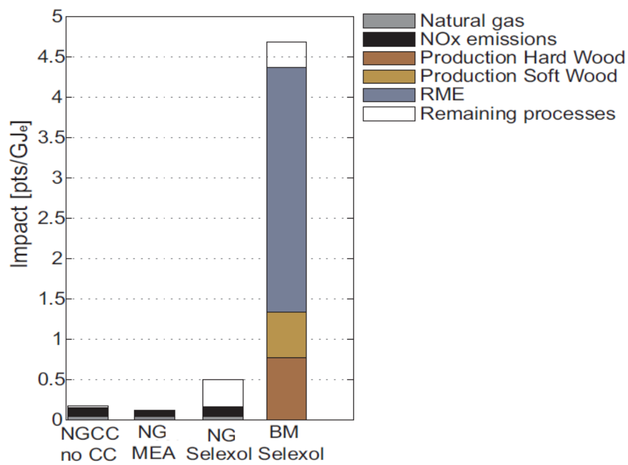

Figure 14: Contributions to the ecosystem impact based on the impact method Ecoindicator-99 (h,a).