THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

System studies of different CO_2 mitigation options in the oil refining industry: Post-combustion CO_2 capture and biomass gasification

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Göteborg, Sweden 2013

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This thesis is based on work conducted within the interdisciplinary graduate school Energy Systems. The national Energy Systems Programme aims at creating competence in solving complex energy problems by combining technical and social sciences. The research programme analyses processes for the conversion, transmission and utilisation of energy, combined together in order to fulfil specific needs.

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The research groups that participate in the Energy Systems Programme are the Department of Engineering Sciences at Uppsala University, the Division of Energy Systems at Linköping Institute of Technology, the Department of Technology and Social Change at Linköping University, the Division of Heat and Power Technology at Chalmers University of Technology in Göteborg as well as the Division of Energy Processes at the Royal Institute of Technology in Stockholm.

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ABSTRACT

To reach the objective endorsed by the EU Council for reducing EU greenhouse gas (GHG) emissions by 80-90% compared to 1990 levels by 2050, extensive cuts are necessary in all sectors. The oil refining sector is a major energy user and thus a major GHG emitter. The overall aim of the work presented in this thesis is to analyse the potential for reductions in GHG emissions in the oil refining industry. The focus is on the implementation of three development routes at case refineries: *Large-scale biomass gasification, to hydrogen or Fischer-Tropsch fuel, as well as post-combustion CO_2 capture and storage (CCS)*. The analysis has been conducted both at the aggregated level; investigating the potential for on-site CO_2 mitigation for the EU refining sector, and at the case study level; focusing on three development routes and including global reduction in GHG emission as well as economic performance.

The findings indicate that the potential for reduction in CO_2 by the available short-term mitigation options in the oil refining industry are relatively limited. The potential for CO_2 capture varies depending on the targeted CO_2 emission point source, the CO_2 capture technology considered and whether CCS is assumed to be limited to areas with large point sources of CO_2 emissions or not. Further, implementing a post-combustion CO_2 capture process at a refinery could be profitable for future conditions characterized by a high charge for CO_2 emissions. The cost for post-combustion CO_2 capture at a refinery is significantly reduced if excess heat from the refining process is used with or without the aid of a heat pump. From a global perspective heat supply by a natural gas combined cycle can also be an interesting option. The potential for global GHG mitigation for the implementation of large-scale biomass gasification for production of hydrogen or Fischer-Tropsch fuel at a refinery is significantly increased with the possibility to capture the clean CO_2 stream generated in the biomass gasification process. Fischer-Tropsch fuel production could be an economically interesting option for a refinery, presupposing economic support for renewable fuel production. Finally, it is important to consider uncertainties in the future energy market. In a comparison between Fischer-Tropsch and post-combustion CO_2 capture, the most profitable option depends on the assumptions on the future energy market.

All the studied development routes lead to reductions in GHG emissions. However, as biomass should be considered a limited resource it is reasonable to assume that the biomass will be used in applications with highest efficiency. In this thesis it is shown that, in most cases, large-scale biomass gasification at a refinery results in lower potential for GHG emission reduction compared to using biomass in coal condensing power plants.

Keywords: CO_2 emissions, GHG emissions, process integration, post-combustion CO_2 capture. CO_2 capture and storage (CCS), oil refining industry, biomass gasification, Fischer-Tropsch (FT), energy market scenarios

To my Family

"You'll never walk alone"

- Rodgers and Hammerstein musical Carousel, 1945

"non scholae sed vitae discimus"

—Lucius Annaeus Seneca 4fkr-65

Appended papers

This thesis is based on the work described in the following papers. The papers are not listed in chronological order based on date of publishing, but rather in the order that makes it easier for the reader to follow the structure of this thesis.

I. Assessment of strategies for CO_2 abatement in the European petroleum refining industry
Johansson D, Rootzén J, Berntsson T, Johnsson F (2012)
Energy, 42 (1): 375–386

II. Hydrogen production from biomass gasification in the oil refining industry – A system analysis
Johansson D, Franck P-Å, Berntsson T (2012)
Energy, 38 (1): 212-227

III. CO_2 capture in oil refineries: Assessment of the capture avoidance costs associated with different heat supply options in a future energy market
Johansson D, Franck P-Å, Berntsson T (2013)
Energy Conversion and Management, 66 (0),127-142

IV. Integration of Fischer-Tropsch Fuel Production with a Complex Oil Refinery
Johansson D, Franck P-Å, Berntsson T
Submitted to Journal of Environment and Sustainable Development (2012)
(A similar version of this paper is available in Proceedings of SDEWES - the 7th Conference on Sustainable Development of Energy, Water and Environment Systems, Ohrid, Republic of Macedonia, 1-7 July 2012, paper 082)

V. Comparative study of Fischer-Tropsch production and post-combustion CO_2 capture at an oil refinery: Economic evaluation and GHG balances
Johansson D, Franck P-Å, Pettersson K, Berntsson T
Submitted to Energy-the International Journal (2013)

Co-authorship statement

Daniella Johansson has been the main author of **Papers II-V**. **Paper I** was co-authored with Johan Rootzén. Daniella Johansson was responsible for the assessment of and the data collection for the analysis with the adjacent infrastructures, the review of energy efficiency measures, fuel substitution and utilisation of excess heat, while Johan Rootzén was responsible for the aggregated assessment of the CO_2 mitigation for the entire oil refining sector, the review of CO_2 capture technologies and key market trends. Professor Thore Berntsson supervised the work in all papers. Dr. Per-Åke Franck co-supervised the work in **Papers II-V**. Professor Filip Johnsson together with Professor Thore Berntsson supervised the work in **Paper I**. Dr. Karin Pettersson has contributed to the system analysis and discussions in **Paper V**.

Related work not included in this thesis

- Heat supply alternatives for CO_2 capture in the process industry
  Johansson D, Sjöblom J, Berntsson T (2012)
  Int. Journal of Greenhouse Gas Control, 8, 217-232

- Options for Increased Use and Refining of Biomass – the Case of Energy – intensive Industry in Sweden
  Ljungstedt H, Johansson D, Karltorp M, Johansson M (2011)
  In Proceedings of WREC – World Renewable Energy Congress, Linköping, Sweden, 8-13 May, Linköping Electronic Conference Proceedings, ISSN 1650-3686, 57, 17-24

Other work by the author not included in this thesis

- An Analysis for Identifying Energy Saving Opportunities for a Petrochemical Cluster in Times of Climate Change
  Johansson D and Harvey S (2009)
  Chemical Engineering Transactions, 18 (Vol 1), 469-474

Commentary on related work

The reason for not including the article with the title "*Heat supply alternatives for* CO_2 *capture in the process industry*" is that this paper primarily focuses on the petrochemical industry and the main focus in this thesis is limited to the oil refining industry. The two other papers are excluded with the same argument: To maintain the focus of this thesis on the oil refining industry.

Table of contents

1 Introduction 1

1.1 Objectives 3
1.2 Papers 4

2 Scope, Delimitations an Key assumptions 7

2.1 Scope 7
2.2 Delimitations 7
2.3 Key assumptions throughout this thesis 8

3 The European oil refining sector 11

3.1 Future predictions of the petroleum market and associated CO_2 emissions 12
3.2 Adjacent infrastructures that could facilitate CO_2 mitigation 13

4 CO_2 mitigation measures for the oil refining industry – related work 15

4.1 Energy efficiency measures and fuel shift 15
  4.1.1 Energy efficiency opportunities 15
  4.1.2 Fuel shift 17
4.2 CO_2 capture and storage 17
4.3 Biomass gasification (BG) 20
  4.3.1 Biomass gasification to hydrogen 22
  4.3.2 Biomass gasification to Fischer-Tropsch fuels 22
4.4 System studies of post-combustion CO_2 capture and biomass gasification to hydrogen and Fischer-Tropsch fuels 24
4.5 Related work at industry level and interactions with adjacent infrastructures 25

5 Studied systems and input data 27

5.1 The EU oil refining sector 27
5.2 The studied case refineries 28
  5.2.1 A future hydroskimming case refinery in Paper II 28
  5.2.2 A current hydroskimming case refinery in Paper III 29
  5.2.3 Current complex case refinery in Paper III and Paper V 29
  5.2.4 A future complex refinery in Paper IV 29
  5.2.5 CO_2 emissions considered for capture in Papers III and V as well as excess heat levels in Papers II, III and V 30
5.3 General aspects of the studied biomass gasification systems 31
  5.3.1 Pre-treatment and gasification 32
  5.3.2 Gas cleaning 33
  5.3.3 Gas upgrading 33
  5.3.4 Hydrogen separation 34
  5.3.5 Fischer-Tropsch synthesis 34

5.3.6 Specific aspects of the hydrogen production in Paper II 34
5.3.7 Specific aspects of Fischer-Tropsch fuel production in Paper IV and Paper V 35
5.4 Post-combustion CO_2 capture and heat supply options 37

6 Methodologies 39
6.1 Methodology approach for analysing the oil refining industry at sector level .40
6.1.1 General methodology approach for Paper I..40
6.1.2 Chalmers Industrial Energy Database 41
6.2 Methodology approach for analysing the oil refining industry at case refinery level...41
6.2.1 General methodology approach in Papers II-V 41
6.2.2 System analysis of GHG emission balances and economic performance. 42
6.3 Energy market scenarios 44
6.3.1 Additional sensitivity analysis..45
6.4 Techno-economic evaluation..45
6.5 Process integration...46

7 Economic input data 49
7.1 Investment cost data 49
7.2 Data for the energy market scenarios 51

8 Results 55
8.1 CO_2 mitigation potential for the European oil refining industry 56
8.2 Effect of large-scale biomass gasification at a refinery on global GHG emissions..60
8.3 Heat supply alternatives for post-combustion CO_2 capture at a refinery 66
8.4 The global CO_2 effect of post-combustion CO_2 capture at a refinery 70
8.5 Comparison of biomass gasification for Fischer-Tropsch fuel production and post-combustion CO_2 capture at a refinery:Economic evaluation and GHG emissions...72

9 Discussion 77
9.1 The need for a functioning infrastructure for CO_2 transportation and storage .77
9.2 Generality of the results...77
9.3 Assumptions regarding CO_2 capture sources and utilization of excess heat 78
9.4 System analysis and availability and reliability of input data 78
9.5 Applicability of the results and relevant comparisons.....................................79
9.6 Using the scenario tool 80

10 Conclusions 81

11 Future work 83

Abbrevations 85

References 87

Acknowledgements 99

1

Introduction

This chapter begins with a short introduction to the thesis. Then the chapter continues with the aim of the thesis, a description of the research questions posted and ends with an overview of the appended papers.

The European Union (EU) has set targets for reducing CO_2 emissions, starting with a 20% reduction in CO_2 emissions by 2020 and with an objective of reducing GHG emissions by 80-95% by 2050 (EC, 2011a), compared to 1990 levels. The latter goal is in line with the recommendation from the Fourth Assessment Report by IPCC (2007), which concludes that in order to limit the global average temperature increase to 2°C or less, the developed countries need to reduce their emissions by 80-95% by 2050. Other targets that have been set by the EU are a 20% share of renewables in the energy mix and a target of 20% energy efficiency improvements by 2020. To specifically stimulate the transition into renewables in the transportation sector, the EU has set an additional mandatory target of 10% renewables in this sector by 2020 (EC, 2009a).

There are three main categories that are estimated to be key mitigation options (IPCC 2007):

- Energy efficiency
- Using low-carbon energy sources (such as renewable energy)
- Using CO_2 capture and storage (CCS)

In this thesis the contribution of oil refineries to a reduction in GHG emissions based on the previously mentioned mitigation strategies is studied. CO_2 emission is the most important GHG emission caused by human activity. The oil refining industry is by its very nature part of the fossil fuel chain and emits significant amounts of CO_2. Most of the emissions from petroleum products are related to the use of fuels and are included in the statistics for CO_2 emissions from the transportation sector or residential sector. However, approximately 8-10% of the cradle to grave CO_2 emissions for petroleum fuel are from the refining process itself (EUROPIA, 2011). The total GHG emissions from EU refineries account for approx. 15% of GHG emissions from the EU industry sector[1], and a total of around 3% of the total GHG emissions in EU27 (EEA, 2012). If adding the GHG emissions from the transportation sector the oil refining process and its petroleum products are responsible for approx. 23% of the total GHG emissions in EU27. In contrast to the total CO_2 emissions in the EU which have declined since 1990, the emissions from the European oil refineries have increased by approx. 18% over the past decades. The

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[1] Based on year 2010 and including: EU manufacturing industries and construction plus petroleum refineries.

trend is primarily due to greater demand for transportation fuels, especially due to the greater demand for lighter distillates, cleaner fuels and heavier crude oils.

Recently, several studies have been published on the opportunities for energy efficiency and CO_2 mitigation in the oil refining industry (Alfke et al., 1999; IPPC, 2003; Szklo and Schaeffer, 2006; Worrell and Galitsky, 2005). These aggregate studies have primarily focused on the possibility to apply already available technologies and measures. However, to realise the goal of further extensive emission cuts, more ambitious mitigation strategies for CO_2 abatement need to be introduced, such as improving overall energy efficiency by means of utilising nearby energy infrastructure, for example through district heating systems, by introducing the production of renewable fuels and renewable feedstocks as well as carbon capture and storage (CCS).

There is increasing pressure on the EU refining sector to increase its production of renewable fuels. During recent years there has been a trend towards quota requirements on renewable production for EU refineries rather than tax relief. Introducing more biofuels has been recognized as one of the key solutions for reducing greenhouse gas emissions from the transportation sector. From January 2017, the actual savings of greenhouse gas emissions from biofuels compared to conventional fossil alternatives must be at least 50%[2] in order to fulfill the sustainability criteria set by the EU commission in 2009 (EC, 2009b). Additional reductions are necessary for new installations after 2017 (60% reduction). Another concern for biofuel is the competition with raw material for the production of food. In the last decade, a lot of hope has been placed on the 2nd generation[3] biofuels that are based on waste material and non-food crops and, thus, fulfill the future sustainability criteria set by the European Commission and do not compete with food production. Lignocellulosic ethanol, Fischer-Tropsch diesel, DME and Methanol are examples of 2nd generation biofuels with high GHG emission saving values[4]. Producing biofuels from the gasification of residues from forestry via Fischer-Tropsch (FT) synthesis has received growing attention as an option for producing transportation fuels. The primary argument is that the fuel can be used directly in today´s car engines. However, FT syncrude needs further processing and using existing refineries for this has several advantages. The refinery structure offers a utility system already in place – process units in which FT syncrude can be co-processed along with crude oil, as well as a long tradition and knowledge of producing transportation fuels.

However, biomass is a limited resource which makes efficient resource utilisation essential. Biomass gasification processes, hereinafter referred to as BG, generate significant amounts of surplus heat, which can be integrated with different heat sinks for higher total conversion efficiency. Examples of different heat sinks are district heating systems, industrial processes or post-combustion CO_2 capture plants.

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[2] According to a standardized accounting method (EC, 2009a).

[3] 2nd generation biofuels are derived from lignocellulosic biomass, such as corn stover, corncobs, straw, wood, and wood byproducts.

[4] Based on the standardized method in (EC, 2009a) in which the whole list of biofuels and their GHG emission reduction values compared to fossil fuels are listed.

A refinery emit large amounts of CO_2 emissions even with a highly energy efficient refinery process and CO_2 capture is one option for cutting the CO_2 emissions further. The post-combustion process is described as being the most promising CO_2 capture technology in a short-to-medium perspective (CONCAWE, 2011; Kuramochi et al., 2012). This technology is very energy intensive and the choice of heat supply is essential. Heat integrating the capture plant with the refinery process would be an opportunity for primary energy savings.

When evaluating the CO_2 mitigating potential for the entire oil refining industry, most studies focus on the best available technology, such as energy efficiency and fuel switching. However, there are also studies that include emerging technologies. Bernstein et al. (2007) is one example of an aggregated study that includes CCS as one of several CO_2 abatement options. Articles published by Holmgren and Sternhufvud (2008) and Elkamel et al. (2008) are examples of case-level studies that includes CO_2 separation as one of several abatement options. However, those authors do not consider the impact of the geographical location. This thesis narrows the knowledge gap by including CCS and the roles of adjacent infrastructures in an aggregated analysis of the potential for CO_2 mitigation in the oil refining industry. This thesis also contributes to new knowledge by studying the consequences of the integration of post-combustion CO_2 capture , BG-to-H_2 and BG-to-FT fuels at case refineries, from a systems perspective, a techno-economic perspective as well as from the perspective of global GHG emissions.

1.1 Objectives

The overall aim of this thesis is to analyse the potential of CO_2 mitigation options in the European petroleum refining industry as well as, from a case refinery perspective, analyse the consequences for the global energy system of integrating selected development routes at a refinery. Further, the thesis aims at investigating key parameters affecting the GHG emission balance, and economic performance in terms of future energy market conditions, such as policy instruments, marginal technologies for electricity generation and energy prices. The CO_2 mitigation options selected are of a strategic character and are significant contributors to global CO_2 and GHG mitigation.

Based on this aim, the objectives of this thesis can be divided into the following research questions:

1. *What is the potential for on-site CO_2 mitigation for the European oil refining industry and where are refineries located that have good access to adjacent infrastructures?*

2. *How would the implementation of large-scale biomass gasification (BG) technology with hydrogen or Fischer-Tropsch fuel production at an oil refinery affect global GHG emissions?*

3. *What is the most cost-effective and robust heat supply alternative for a post-combustion CO_2 capture plant?*

4. *What is the effect of post-combustion CO_2 capture on CO_2 emissions when the perspective is changed from a refinery perspective to a global perspective?*

5. *Under what conditions is it more advantages to invest in a CO_2 capture system (post-combustion technology) than in a large-scale biomass gasification for Fischer-Tropsch fuel production at a complex oil refinery?*

1.2 Papers

The thesis is based on five papers. Brief descriptions of the papers are presented below.

The thesis starts from a high systems perspective in which the first paper presents an overview of the European oil refining industry and its on-site CO_2 mitigation potential, which is based on data from existing European statistics and databases. In **Paper I** (*Assessment of strategies for CO_2 abatement in the European petroleum refining industry*) an assessment of the key characteristics and future trends for the European (EU-27 + Norway) oil refining industry is presented as well as the identification of suitable mitigation options and mapping of adjacent infrastructure. By combining inputs from the assessment of the current status of the refining industry and a review of potentials for various abatement strategies with results from analysis of adjacent infrastructures, an assessment of the CO_2 mitigation potential is made.

Paper II (*Hydrogen production from biomass gasification in the oil refining industry – a system analysis*), and the following papers analyse the potential for the implementation of different CO_2 mitigation options in case refineries. In **Paper II**, the production of hydrogen via BG from a variety of technologies is compared with a conventional way of producing hydrogen and analysed for global CO_2 emissions. The case refinery in this paper is a hydroskimming refinery, which is a small refinery with diesel and gasoline as two of the main products. The findings are shown in relation to the conventional way of producing hydrogen and the emission reduction is compared to alternative usage of biomass.

In **Paper III (***CO_2 capture in oil refineries: Assessment of the capture avoidance costs associated with different heat supply options in a future energy market*), post-combustion CO_2 capture is evaluated in two different case refineries; a hydroskimming refinery (the same as in **Paper II**) and a complex refinery, which is a large refinery with low production of heavy hydrocarbons and high production of high value added fuels. The post-combustion process is a very energy demanding process and therefore the analysis is based on a cost estimate of the CO_2 capture avoidance cost for four different heat supply options; excess heat from the refinery process (with or without the aid of a heat pump), natural gas CHP[5], biomass CHP and a natural gas combined cycle. The analysis is made for four different scenarios that outline possible cornerstones of the future energy market.

In **Paper IV (***Integration of Fischer-Tropsch Fuel Production with a Complex Oil Refinery*) the same approach as in **Paper II** is used, with the exception that the final products from the BG process are FT fuels and the case refinery is a complex refinery (the same as in **Paper III**). In this paper the potential for GHG emission reduction through the integration of BG-to-FT fuel production is compared to stand-alone FT fuel production as well as the alternative use of biomass.

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[5] Combined heat and power

The last paper, **Paper V (***Comparative study of Fischer-Tropsch production and post-combustion CO_2 capture at an oil refinery –Economic evaluation and GHG balances*), unifies this thesis with a comparative analysis of two of the studied development routes, post-combustion CO_2 capture and BG-to-FT fuel production. **Paper V** includes an analysis of global GHG emissions, and of profitability and systems aspects. The analysis in this paper is, as in **Paper III**, based on four different scenarios that outline possible cornerstones of the future energy market.

A summary of the research questions addressed in each of the papers is given in Table 1.

Table 1. Research question addressed in the appended papers.

Research question	**Papers**				
	I	**II**	**III**	**IV**	**V**
1	**Q**				
2		**Q**		**Q**	**Q**
3			**Q**		
4			**Q**		
5					**Q**

The same research question (*Research question 2)* is addressed in **Papers II, IV** and **V** but the product from the gasification process differs. In **Paper II** the large-scale BG technology produced hydrogen for internal use in the refinery processes. While in **Papers IV** and **V** the purpose of the gasification process was to produce Fischer-Tropsch fuels. The CO_2 emissions in **Paper II** were evaluated in relation to a reference refinery consisting of a future hydroskimming case refinery and a new steam methane reformer (SMR) for hydrogen production. In **Paper IV** the reference refinery was an optimally heat integrated refinery, while the reference refinery in **Paper V** was a complex case refinery under current operation.

2

Scope, Delimitations and Key assumptions

This chapter presents the scope of the thesis and the delimitations of the appended papers on which this thesis is based.

2.1 Scope

The thesis is based on a bottom-up approach and examines the European oil refining industry at two different systems levels: the whole European market, mainly based on information available in official and research databases, and from the case perspective in which data is taken from specific case refineries. The focus is mainly on three development routes: BG-to-H_2, BG-to-Fischer-Tropsch (FT) fuels and post-combustion CO_2 capture and storage (CCS). **Paper I** includes a review of several mitigation options, other than CO_2 capture and BG. Two case refineries were studied: one simple refinery (a hydroskimming refinery) and one large and complex refinery.

2.2 Delimitations

Even though the case refineries were local and specific, the system study was conducted from a European energy systems perspective, using different energy prices on the European energy market. This thesis is limited to development routes that can be used in combination with the existing oil refining structure. There are several other emerging technologies that could be interesting options for reducing CO_2 emissions related to the oil refining industry that are beyond the scope of this thesis, such as BG to SNG or methanol or electricity and the production of biofuels via pyrolysis or via hydro treating of bio-oils as well as other CCS technologies (e.g. oxyfuel combustion).

This thesis is limited to BG for hydrogen and FT fuels production, for two main reasons. First, there is an increasing demand for hydrogen in the oil refining industry and most refineries will most likely suffer from a deficit of hydrogen in the near future. This is further described in Chapter 3. The reason for studying BG-to-H_2 in a simple refinery, and not in a complex refinery, is because this type of refinery has a more urgent need for hydrogen, especially if gasoline production must be reduced. Second, the renewable energy directive endorses a mandatory renewable content of 10% in the transportation sector for all member states by 2020 (EC, 2009a), which creates an incentive for the oil refining industry to increase its renewable fuel production and search for new alternatives.

FT fuel production is considered since part of the process is often considered to be utilised at a refinery, using existing refining units (e.g. processing the FT wax in the current hydro-cracking units) and/or utilising the refining utility system. In addition, the final FT fuels have properties similar to the current gasoline and diesel fuels and, therefore, are suitable in today's car engines. However, the FT wax has different properties compared to the vacuum gas oil (VGO) which is currently processed in the refinery's hydro-cracking units. It is reasonable to assume that the FT wax can be co-processed with VGO in the refinery up to a level of 10% co-processing of the FT wax without any production changes. Furthermore, with FT fuel production a refinery will continue to produce products for the same market as today.

This thesis focuses on the post-combustion CO_2 capture process. The main reason is because the post-combustion process is the technology closest to commercialisation and no major reconstruction of the existing refinery is necessary. By using the post-combustion process it is also possible to capture CO_2 from several point sources, e.g. from the several stacks. Other capture technologies are considered in **Paper I**, but not considered in detail in this thesis.

The focus of biomass is on forest-residue biomass (wood fuel). Other resources, such as agricultural and waste resources, are not considered here. There are discussions regarding the environmental effects of utilizing different types of biomass. The discussion includes forest residues, but to a lesser extent than other types of biomass. This discussion, however, is beyond the scope of this thesis. The timeframe is mainly a medium-term future (2030).

2.3 Key assumptions throughout this thesis

Throughout this thesis it is assumed that all available excess heat can be utilised. In all papers, except **Paper IV**, the excess heat is based on the current operation of the case refineries and is presented as process streams cooled with air and water as well as an additional cooling of flue gases.The existing process-to-process heat exchange network was assumed to be well constructed and accepted without further analysis. In **Paper IV** an energy-optimised refinery was assumed. In this paper it was assumed that all energy measures that are theoretically possible are executed before a BG-to- FT fuel production is installed. Furthermore, the FT syncrude process is also assumed to be maximally heat integrated and all excess heat is assumed to be available for usage.

In this thesis biomass is regarded as a limited resource, i.e. there is not enough biomass to substitute for all fossil-fuel based applications. In addition, the future available bioenergy potential is uncertain. Various estimates show a wide range of potentials. For example, within the Biomass Energy Europe project (Torén et al., 2011) the bioenergy potential for Europe was estimated to range from 1.3 to 7 PWh/y for 2030. Moreover, as discussed in for example Azar et al. (2003) and Faaij (2006), if the implementation of biomass is realised for co-firing in coal power plants, for the production of transportation fuel as well as for replacing fossil feedstocks in the industry, it would require more biomass resources than may be available. Therefore, it is not unrealistic to assume that woody biomass will be subject to competition from various existing and future biomass users. When evaluating the GHG balance from biomass use, the GHG effect should always be compared to an alternative use of the biomass, since the use of biomass for a new application results in less biomass being available for other purposes. This is further described in 6.3.

Considering capturing and storing CO_2 emissions, the analysis in this thesis presupposes a functioning transportation system as well as safe storage sites for CO_2. It is further assumed that the CCS option has gained public acceptance and that all legal obstacles and uncertainties have been resolved. The latter assumptions have also been employed in the analysis of BG-to-FT fuel and BG-to-H_2 production. The cost estimates for transportation and storage are taken from (CCS Skagerrak - Kattegat, 2011) and presuppose transportation by pipeline. The cost has been calculated on full utilisation of the pipelines, i.e. that all the considered sources are equipped with CCS at the same time.

Currently, lack of space is a common problem at many refineries. Therefore, both the FT fuels production units and the post-combustion CO_2 capture units are assumed to be located outside today's primary process area. The estimated piping distance for utility systems, has been doubled to calculate for deviations from straight piping. For the CO_2 capture case, the flue gases are assumed to be transported to a centralized capture plant, with the cost for flue gas channels included in the analysis.

Furthermore, when evaluating the potential for implementing CCS in BG-to-FT fuel production, **Paper IV**, the captured CO_2 is assumed to originate from biomass feedstock. Currently, CO_2 emissions from biomass are not included in the EU ETS[6]. However, the same effects on global CO_2 emissions are obtained if CO_2 emissions are captured, regardless of origin. Therefore, it is assumed that in future policy systems, captured and stored CO_2 originating from biomass will be granted the same economic compensation as CO_2 originating from fossil fuels. The same view-point is discussed in more detail in for examples studies by Grönkvist et al. (2006), who argue for the importance of including credits also for biotic CCS in an international framework, i.e. not specifically within the EU ETS.

---

[6] Emission Trading System

3

The European oil refining sector

Even if the main focus of this thesis is on a case study level it is relevant to know the characteristics of the whole industry, i.e. where refineries similar to the case refineries are located and the future demands for this industry. Therefore, this chapter gives an introduction to the European oil refining industry, describing the configurations and locations of the oil refineries. Overviews of current and future trends in the petroleum market, as well as a brief description of important adjacent infrastructures are also given.

The European oil refining industry (EU-27 + Norway) currently (2011) consists of 114 refineries, see Figure 1, with a combined capacity of approximately 770 Mt crude oil/y. Refineries can be found in 22 of the 27 EU Member states and the oil refining stock in Europe consists of a variety of types of oil refineries, from base oil refineries, which are limited to the production of heavy fuel oil, to high conversion cracking refineries (IPPC, 2003). Oil refineries are often divided into different categories, depending on the configuration of a refinery. In this thesis the refineries are categorised based on the descriptions by the IPPC (2003) and Reinaud (2005), see Table 2. The case study refineries in the appended papers are categorised as Configuration 1 and Configuration 4.

Table 2. Refinery configurations, based on description in (IPPC, 2003; Reinaud, 2005)

Configuration 1 including Base oil refineries	The simplest type of oil refinery. These refineries are equipped with a distillation unit, naphtha reformer, and some type of necessary treatment facility. The main product is often gasoline. Base oil refineries do not have conversion units, which makes these refineries limited to the production of heavy fuel oil.
Configuration 2	Can convert fuel oil to a more valuable fuel by adding a vacuum distillation unit and a catalytic cracker to the units in Configuration 1.
Configuration 3	Configuration 3 adds a hydro cracker, which maximises the production of gasoline and middle distillates and allows the production of high-quality diesel.
Configuration 4	Has both hydro cracking and catalytic cracking units. Some refineries have an IGCC unit, which converts solids and heavy fuels to power and co-generation steam. Eleven refineries in the EU27 countries have a coker unit, which reduces heavy fuel oil to lighter fuels (i.e. diesel) and produces a low-value by-product, coke.

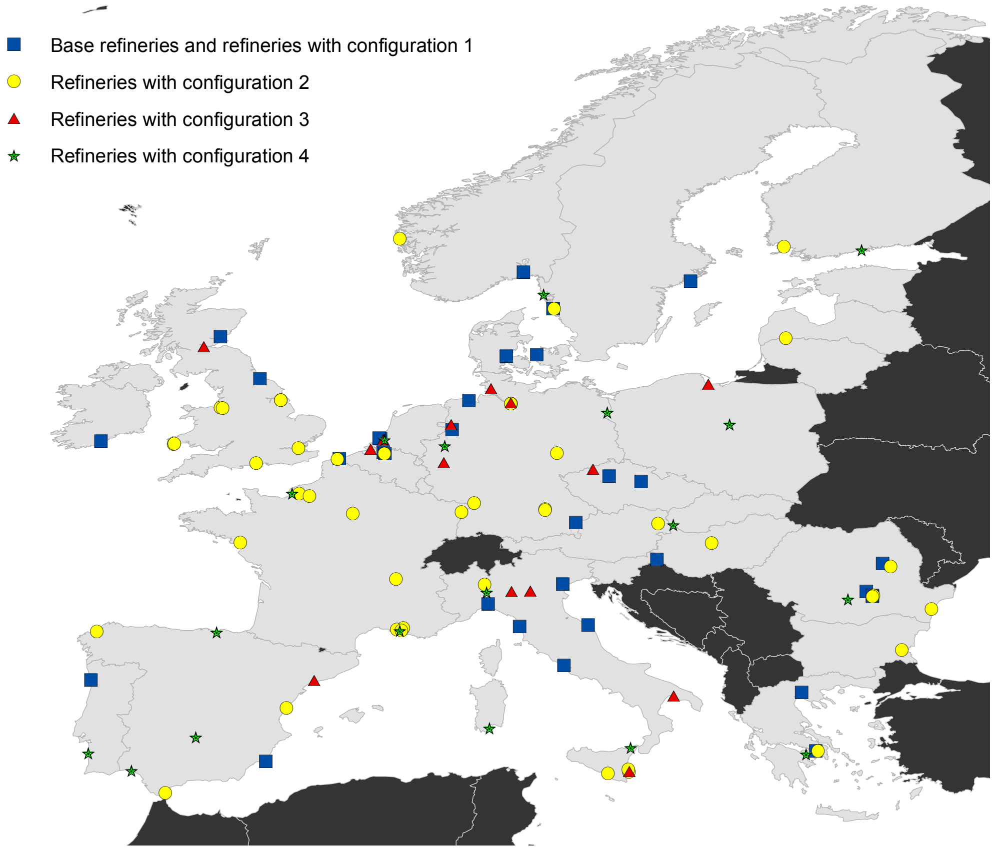

Figure 1. Map of all refineries in EU 27 + Norway. The green stars represent refineries with Configuration 4. Red triangles represent refineries with Configuration 3 and yellow circles represent refineries with Configuration 2. The blue rectangles present the base refineries and refineries with Configuration 1. *This map includes Intellectual Property from European National Mapping and Cadastral Agencies and licensed on behalf of these by EuroGeographics.*

3.1 Future predictions of the petroleum market and associated CO_2 emissions

Currently, the European refineries struggle to meet the demand for a mixture of petroleum products and the tighter specifications for sulfur content in petroleum products. The demand for diesel and gasoil exceeds production while a surplus of gasoline is produced at European refineries (EUROPIA, 2012). The total production of petroleum products is predicted to be relatively stable over the coming decades and the gap between the demands for gasoline and diesel is expected to continue to grow (EUROPIA, 2011). However, the forecast demand for fossil fuels in the transportation sector and other end-use sectors varies significantly between different sources and future scenarios (EC, 2011b; EUROPIA, 2011; IEA, 2010a). It is primarily fuels for domestic heating and fuels for cars (gasoline, diesel/gasoil) that will be replaced by alternative fuels or powertrains, while the demand for aviation fuel and marine fuel is predicted to remain constant or to grow slowly (EUROPIA, 2011). The main strategy for refineries to meet the uncertainties in the petroleum market would be to increase the complexity of the refinery, as discussed by e.g. Castelo Branco et al. (2010). Speight (2011) describes the refinery of the future as:

a technologically focused refinery with energy-efficient installations, which has the ability to accept a variety of feedstocks, including biomass. Thus, it is reasonable to assume that the refineries with the highest complexity are among those with the potential to adjust to changing product demands.

Changes in fuel demand, heavier crude oil and tighter specifications for sulfur content in petroleum products result in a rise in the demand for energy and hydrogen in the European petroleum sector (CONCAWE, 2012). Hydrogen is produced as a by-product from gasoline production. Refineries with a large number of conversion units usually have a hydrogen production plant, but for refineries that primarily produce gasoline the hydrogen from the gasoline production has previously been sufficient to cover the hydrogen demand in the refining processes. However, if those refineries are to adapt to a changing market, their diesel production must increase. This will lead a deficit of hydrogen that must be covered by a new hydrogen plant or the import of hydrogen. The prediction for future CO_2 emissions heavily depends on the development of alternative transportation fuels in the transportation sector as well as the development and implementation of CO_2 mitigation technologies.

3.2 Adjacent infrastructures that could facilitate CO_2 mitigation

The choice of technologies and system solutions to be implemented in the future is likely to depend on the development and transition of other parts of the European energy system. For example, the possibility to implement CCS at a refinery depends on the development of a future infrastructure for CO_2 transportation and storage, and the possibility to export heat from a refinery is highly dependent on the distance to a district heating network. Thus, it is necessary to have good knowledge about the surrounding energy infrastructures to be able to describe the overall potential for CO_2 emission reduction in the oil refining industry. Infrastructures that in this thesis are considered important for future CO_2 mitigation potential related to the oil refining industry are:

- *District heating network*s that could facilitate the utilization of excess heat from the refining process and reduce global CO_2 emissions (i.e. no reduction of CO_2 emissions at the refinery), but compete with post-combustion CCS.
- *A natural gas grid* that could facilitate fuel substitution and reduce on-site CO_2 emissions.
- *Nearby industries* (i.e. petrochemical industries) that could create opportunities for site-wide energy mitigation measures and reductions in global CO_2 emissions.
- *Adjacent CO_2 storage site and CO_2 capture clusters*[7] that could facilitate CO_2 capture and storage at refineries.

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[7] Chapter clusters are, in this thesis, defined as regions where emissions from large stationary point sources (also including emissions from power plants and pulp and paper plants) exceed 20 $MtCO_2$ annually. These areas are assumed to be particular interesting for CO_2 capture and transportation.

4

CO_2 mitigation measures for the oil refining industry – related work

This chapter gives an introduction to different CO_2 mitigation measures for the oil refining industry, both commercially available and emerging technologies. In addition, this chapter presents related work with a focus on energy efficiency at oil refineries and related work connected to the studied development routes as well as the related systems studies.

4.1 Energy efficiency measures and fuel shift

During recent years, a large number of studies have been published focusing on energy abatement options for the oil refining industry. These range from energy measures in specific unit operations to site-wide energy measures such as process integration. The focus in this thesis is on the potential for energy measures at a refinery at an aggregated level. The potential for fuel shift aims at substituting the liquid fuels that are used in furnaces or boilers at refineries for fuels with lower total GHG emissions (i.e. natural gas or biomass).

4.1.1 Energy efficiency opportunities

A number of opportunities exist within the petroleum refinery industry to reduce energy consumption while maintaining the same production in the refinery. Several energy audits have been conducted that show significant potential for energy savings in oil refineries. For example, an energy assessment study conducted in 2001 at a refinery in California identified energy measures that could save 12% of the total energy used at the plant (US-DOE-OIT, 2002). Large energy savings can be achieved by means of heat integration. Most heat integration studies have been conducted on parts of the refinery process. For example, Al-Riyami et al. (2001) found possibilities for energy savings in the fluidised catalytic cracking (FCC) process of 9 MW (19%), with a payback period of 1.5 years. For the same process in a Romanian refinery, Jara-Morante et al. (2009) showed that 15% of the energy could be saved by means of process integration. Bulasara et al. (2009) and Valino et al. (2008) have reported significant heat integration potential in a crude distillation unit. The sum of the energy saving potential for all subsystems is not always equal to the energy saving potential for the whole system, as illustrated e.g. by (Feng et al., 2011; Markusson et al., 2012). During the years, total site analysis has been applied by refineries all over the world to identify optimal utility levels. Brown (1999) reported that typical energy savings from total site analyses are in the range of 20-30%, which was limited to 10-15% when economic potential was considered.

Table 3 summaries the energy saving potential for a selected number of measures for oil refineries described in the literature. Worrell and Galitsky (2005) indicate that an energy saving potential of 10-20 % is economically feasible for most U.S. refineries. Petrick and Pellegrino (1999), who also studied the energy saving potential for the US refining industry, reported that an energy saving potential of 15-20% is feasible to achieve in a medium to long-term perspective. For Brazilian oil refineries, Szklo and Schaeffer (2007) have estimated the potential for energy savings with a focus on two alternatives: the reduction of primary energy use and the implementation of non-hydrogen consuming technologies for sulphur removal. They suggest a near-to-medium-term energy saving potential ranging from 10% to 20%. Alsema (2001) discuss the energy saving potential by energy-efficiency measures for Dutch refineries.

Table 3. Energy saving potential of total fuel consumption for a selection of measures described in the literature.

Energy saving measures	**Szklo & Schaeffer (2007)**	**Alsema (2001)**	**Petrick & Pellegrino (1999)**	**Worrell & Galitsky (2005)**
Heat integration and waste heat recovery	10%[a]	6%[b]	10% (20–40%)[c]	10–30%[d]
Fouling mitigation	2%	2%	2%	0.7–3.4%[e]
Advanced process control	2%	2.5%		2–18%
Combustion efficiency improvements			5%[f]	13–33%[g]
Advanced turbine systems/Co-generation			2 %	
Thermal cracking	17%	18%	3-5%[f]	
Membrane technology[h]	-	-	-	-
Pumps	1%[i]			2–17%[k]
Biodesulfurisation	70–80%[j]	4.4%[l]	2%[f]	
Oxidative desulfurisation process	40%[m]			
Catalytic distillation process	62%[n]			
Advanced catalyst for Fluid Catalytic Cracker and hydroprocessing		15%		
Distillation			4-7%	35%[o]

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[a] Includes: use of waste heat in absorption refrigeration systems; use of waste heat to pre-heat feeds; heat and/or mass (water and hydrogen) integration using Pinch techniques; improvement of furnace efficiencies in combination with computer-controlled combustion; direct feeding of "intermediary products" to process without cooling or storage; use of heat pumps; decreased film temperature and increased turbulence on heat transfer surfaces; insulation of buildings and process units; and adoption of steam management.

[b] Includes: improved heat management and waste heat recovery; and process integration and cross-industry optimisation.

[c] This number is based on energy savings reported in several pinch analyses (Petrick and Pellegrino, 1999).

[d] Based on results from several pinch analyses conducted at different refineries reported by Linnhoff March (2000) (currently KBC-Energy services).
[e] The second number is from (Panchal and Huangfu, 2000), translated into the percentage of the specific energy consumption of Brazilian refineries (Szklo and Schaeffer, 2007).
[f] Savings of fuel for heat and power.
[g] Based on fuels saved in boilers and includes: improved process control; reduced flue gas quality; improved insulation; boiler maintenance, and flue gas heat recovery.
[h] The actual savings achieved with membrane separation are unclear. None of the studies found this option to be very promising. Petrick and Pellegrino (1999) do not even mention this option.
[i] Percentage of electricity consumption.
[j] Reported by Linguist and Pacheco (1999), in (Szklo and Schaeffer, 2007). Percentage relative to conventional hydrodesulphurisation.
[k] Percentage of energy consumption by pumps.
[l] Savings for diesel production.
[m] Percentage relative to conventional hydrodesulfurisation.
[n] Percentage relative to conventional hydrodesulfurisation.
[o] Percentage relative conventional CDU, based on information from the manufactory.

4.1.2 Fuel shift

The major part of the fuel used in a refinery is a by-product from the refining process (refinery gas) and contains light hydrocarbons (mostly C1-C2). The additional fuel, which on average is 23% of the total energy demand in EU refineries, is today covered by low-value liquid fuels. There is a potential for refineries to use these liquid fuels for other purposes and instead import natural gas. For example, Elkamel et al. (2008) showed that 30% of the CO_2 emissions could be saved by switching all fuels for natural gas. Another alternative would be to use biomass as fuel in the furnaces. Such a shift would however involve major refurbishments to current furnaces and/or boilers. The used system boundary on the system for the use of biomass is important for the assessment of the effect of changing from refinery fuel to biomass. The system boundaries and methodologies are described in Chapter 6.

4.2 CO_2 capture and storage

Although the power sector has been the main focus of CCS research, there is significant potential for the application of CCS to the industry. The International Energy Agency (IEA) (2011)[8] estimates that in order to limit climate warming to between 2°C and 3°C, CCS will have to account for 9% of the total reduction in CO_2 emission to be achieved in the world-wide transportation and industry sectors.

There are in principle four options for CO_2 capture:

- Pre-combustion processes in which CO_2 is removed from the fuel prior to combustion in order to produce hydrogen. The hydrogen is then combusted without the formation of CO_2.
- Post-combustion processes in which CO_2 is removed from the flue gases derived from fossil fuel combustion.

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[8] The IEA report is a roadmap that starts with IEA Energy Technology Perspectives' BLUE Map scenario, which describes how possible energy technologies are transformed by 2050 to achieve the goal of reducing CO_2 emissions to half of that of 2005, on a yearly basis.

- Oxyfuel combustion in which fuel is combusted in oxygen (mixed with recirculated flue gas) instead of air, creating a more or less a pure CO_2 stream in the off gases.
- Chemical-looping combustions, which is a combustion technology with inherent CO_2 separation without any energy penalty from the separation of gases (i.e. the separation of oxygen from air or the separation of CO_2 from flue gases is avoided). This technology is, however, still on the pilot scale.

Currently, around the world, there are 73 active or planned CCS projects (Global CCS Institute, 2012), of which fifteen involve the separation of industrial CO_2 emissions.

Over the past 20 years, the results of several studies of CO_2 capture at industrial facilities have been published. For example, UNIDO[9] (2010) published a technological synthesis report for CCS in industry, which describes the main technology options and highlights case studies and cost estimates for research, demonstration, and commercial projects. IEA (2011) published a technological roadmap for CCS in industry, which shows that CCS could reduce CO_2 emissions by up to 4.0 Gt/y by 2050 (accounting for 12% of the total world-wide CO_2 emissions in 2010). The oil companies′ European association, Concawe, (2011) published a report on the potential for CCS application to EU oil refineries. Their results indicate that 50% of the combusted CO_2 emissions in most refineries could be practically or economically captured. The cost for CO_2 capture for selected industries has been studied by e.g. Kuramochi et al. (2012) who, among others, compare the techno-economic performance of several CO_2 capture techniques for the oil refining sector. The findings of their study indicate that CO_2 capture in oil refineries could be achieved at a cost in the range of 30 to 120 €/t CO_2, depending on the targeted CO_2 emission point source characteristics and the CO_2 capture technology considered.

To date, most focus is on post-combustion and oxyfuel combustion capture. One reason that oxyfuel is frequently discussed is that it generates a cleaner CO_2 stream and less heat is required. However, the oxyfuel process requires oxygen, which is very energy demanding to produce, and it requires the reconstruction of furnaces and/or boilers. No re-construction of existing equipment is necessary for post-combustion CO_2 capture. On the down side, the regeneration in the desorber is a very energy consuming part of the post-combustion process. Nevertheless, according to Kuramochi et al. (2012) and CONCAWE (2011) the post-combustion process is the only feasible option in a short-term perspective. However, the cost for post-combustion CO_2 capture may fall if low-cost LP[10] steam is available. Large process industries often have excess heat available at different heat levels which could be used to partly or fully cover the heat demand in the post-combustion process. For example Hektor and Berntsson (2008) have used this approach to study the potential for reducing the cost for the capture of CO_2 at a kraft pulp mill. The authors show that thermal process integration is possible and that it significantly reduces the cost of CO_2 capture. Further, Hektor (2008) showed that using an NGCC is the most competitive way of supplying heat to a post-combustion CO_2 capture plant at a pulp mill.

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[9] United nations industrial development organization

[10] LP – Low Pressure

Table 4 summarises studies of CO_2 capture costs for oil refineries found in the literature. The cost varies between different studies and different technologies and different studies apply different system boundaries. Some studies include CO_2 capture on the heat and power production, while other does not. However, the most common approach among the studies listed in Table 4 is a refinery perspective, where the avoided CO_2 emissions are corrected with the CO_2 emitted by the heat and power production. Kuramochi et al. (2012), Allam et al. (2005) and Melien (2005) include also CO_2 credits for exported power. The cost in all the papers listed in the table below exclude the cost for CO_2 transportation and storage.

Table 4. Summary of studies of cost for CO_2 capture for oil refineries found in the literature. All costs are in original year but recalculated to €. The cost in original monetary value is found below the table. Abbreviations in the table stand for: PC = post-combustion, OC=oxyfuel combustion, PreC= pre-combustion, CS= combined stack.

Authors	**Capture pant**	**Process**	**Energy source**	**P [bar]**	**CO_2 captured [Mt/y]**	**Cost year**	**Cost [€/t CO_2 captured]**	**Cost [€/t CO_2 avoided**
(Allam et al., 2005)[a]	OC	Boilers & Heaters	Gas turbine[e]	220	1.9	2005	31[j]	35[j]
(Haugen, 2009)[b]	PC	CS	NG-CHP, excess steam[e]	200	0.26 and 0.28	2009	72 and 75[i]	
(Ho et al., 2011)[c]	PC	CS	External NG-CHP[d]	100	0.88	2008		59[f]
(Holmgren and Sternhufvud,	PC	Hydrogen plan				2010		24-29[o]
(Kuramochi et al., 2012)[a]	PC	CS	On-site NG-CHP[d]	110	1.2-1.6	2007		120
	PC	FCC	Steam import[d]		0.6-0.8	2007		70-110
	OC	CS	On-site NGCC or NG-CHP or power import[d]		1.6	2007		50
	OC	FCC	Steam import[d]		0.8	2007		60
	OC	CS	On-site NGCC or NG-CHP or power import[d]		1.6	2007		30
	PreC	CS	WGSMR[d]		1.6	2007		80
(Lindsay et al., 2009)[c]	PC	Hydrogen plant	Auxiliary boiler[e]		1.2	2007		53[h]
(Meerman et al., 2012)[c]	PC	SMR	On-site CHP[e]	110	0.33	2008	35	41

(Melien, 2005)[a]	PC	Heaters & Boilers	NG-CHP[d]		2.19	2003		69[l]
	PreC		NG-CHP[d]		1.99-2.19			43-76[m]
	OC		NG-CHP[d]		2.09			36-43[n]
(Simmonds et al., 2003)[b]	PC	CS	On-site NG-CHP[e]	220	2	2003	45-53[g]	
(van Straelen et al., 2010)[c]	PC	CS	Fuel gas CHP[d]		0.6-1.25	2007		~90
	PC	FCC	Fuel gas CHP[d]		0.4	2007		~110
(Zanganeh et al., 2005)[c]	OC	Boiler	Boiler[d]	250	1.34-1.18	2005		32[k]

[a] Includes also CO_2 emissions from heat and/or power production plus including CO_2 emissions from exported electricity.

[b] Does not include CO_2 emissions from offsite power and heat generation.

[c] Includes CO_2 emissions from facility that generates the heat but do not consider the saved CO_2 emissions due to saved marginal electricity.

[d] Capture CO_2 from heat and power production.

[e] Does not consider CO_2 capture of emissions from heat and power production.

[f] 87$\$_{2008}$/t CO_2 avoided, [i] 50-60\$/t CO_2 captured, [j] 71\$/t CO_2avoided, [k] 596NOK/t CO_2 captured ;625NOK/t CO_2 captured, [l] 38\$/t CO_2 captured, 43\$/t CO_2 avoided, [m] 45\$CAD/t CO_2 avoided, [n] 78\$/t CO_2 avoided , [o] 48 – 85\$/t CO_2 avoided, [p] 41-49\$/t CO_2 avoided, [q] 200-243 SEK/t CO_2

In the table above it can be seen that oxyfuel combustion is reported to have a lower avoidance cost than other CO_2 capture technologies. Still, none of the mentioned studies have used excess heat to supply the post-combustion process. Van Straelen et al. (2010) mention excess heat from the refining process as an option, but state that the possibility for significant heat exchange is marginal.

4.3 Biomass gasification (BG)

Gasification is a process that converts carbonaceous materials into an energy-rich gas (syngas/product gas). Gasification of fossil material is commercially available, while gasification of biomass material is still under development. There are several possible routes for the gasification of biomass and conversion of syngas into a variety of products, which is illustrated in Figure 2. As described in Section 2.2, the focus in this thesis is on hydrogen and FT fuel production.

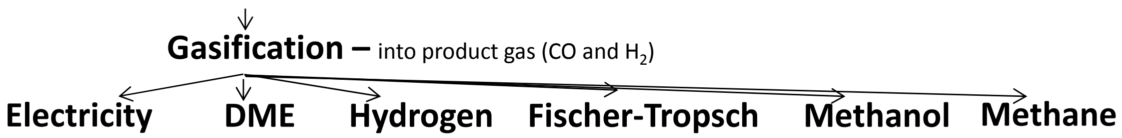

Figure 2. Different routes for BG into various products.

Gasification of fossil-based feedstock is a mature technology and in 2010 over 400 gasifiers were in operation globally, with a combined capacity of over 70 GW_{th} product gas output (NETL, 2010b). According to IEA Bioenergy (2012), there are currently 14 biomass gasifiers worldwide. Kirkels and Verbong (2011) report a world-wide BG capacity of 1.4 GW_{th}. However, BG is still far from commercialization and most of the projects are still on the pilot scale only. In addition, several of the larger projects were recently stopped, which is mainly due to economic reasons. Table 5 shows the current status of BG projects in Europe.

Several coal-to-liquid and gas-to-liquid FT facilities are currently in operation or planned for operation (Andrews and Logan, 2008). FT fuel production from biomass is, however, still on a pilot plant level (Damartzis and Zabaniotou, 2011; IEA Bioenergy, 2012). Around 20 gasification plants and gas-to-liquid plants with hydrogen production are up and running, using petroleum or natural gas as feedstock NETL (2010a). No plants for hydrogen production that run on biomass are yet in operation.

Table 5. Operational, planned and stopped BG plants in Europe, based on (Hellsmark, 2010; IEA Bioenergy, 2012; Wetterlund, 2012).

Company	**Location**	**Type of gasifier**	**Size**		**Products**	**Year of start up**
Vienna University of Technology	Güssing, Austria	Indirect	8 MW	pilot	FT, SNG	2001
Chemrec/LTU	Piteå, Sweden	EF	1 MW	pilot	DME	2011
ECN	Petten, the Netherlands	Milena (BFB)		pilot	CHP	2007
Chalmers University of Technology	Göteborg, Sweden	Indirect gasifier	6 MW	Pilot	Syngas for SNG	2007
Karlsruhe Institute of Technology (KIT)	Karlsruhe, Germany	EF	5 MW	Pilot	Methanol	2013[a]
Göteborg Energi	Göteborg, Sweden	Indirect	20 MW_{SNG}	Demo	SNG	2013[a]
ECN	Alkmaar, the Netherlands	Milena (BFB)	12 MW	Demo	SNG	2013[b]
NSE Biofuels Oy, Neste Oil and Stora Enso JV	Varkaus, Finland	Fluidised bed	12 MW	Pilot	FT	2009[c]
CHOREN Industries GmbH	Schwedt, Germany	Carbo-V	640 MW	Commercial	FT	[c]
CHOREN Fuel Freiberg GmbH & Co. KG	Freiberg, Germany	Carbo-V	45 MW	Demo	FT	2012[c]

[a] under construction, [b] planned, [c] stopped

4.3.1 Biomass gasification to hydrogen

During recent years, several studies have been published on BG-to-H_2, e.g.(Hamelick and Faaij, 2002; Mueller-Langer et al., 2007; Müller et al., 2011; Spath et al., 2005; Wright and Brown, 2007). Spath and Dayton (2003) conducted a review study of the technical and economic feasibility of a number of processes for biomass-derived syngas conversions to fuels and chemicals. They found hydrogen production to be one of the most favorable products. Later Spath et al. (2005) proceeded from those results and published detailed data for hydrogen production in terms of performance, process design parameters and costs based on the Battelle Colombus Laboratory Indirectly-Heated Gasifier. Hamelinck and Faaij (2002) conducted a review of different technological aspects for methanol and hydrogen production. Those authors also included a techno-economic comparison of several production routes for hydrogen based on flowsheet modeling. While the studies mentioned above primarily focus on the techno-economic performance of gasification to hydrogen process, there are studies that mainly focus on exergy and exergyeconomic analysis, e.g. (Abuadala and Dincer, 2011; Cohce et al., 2011; Kalinci et al., 2012a) and others with an emphasis on environmental aspects, e.g. (Kalinci et al., 2012b; Koroneos et al., 2004).

Concerning hydrogen production for utilization in oil refineries, Sarkar and Kumar (2010; 2009) have studied hydrogen production for upgrading of bitumen in oil sands in Canada. The authors consider stand-alone production of hydrogen and a pipeline transport between the hydrogen plant and the refinery.

4.3.2 Biomass gasification to Fischer-Tropsch fuels

Techno-economic studies of FT fuel production via BG have been conducted by several authors, e.g. (Bechtel, 1998; Hamelinck et al., 2004; Swanson et al., 2010; Tijmensen et al., 2002; Tock et al., 2010; van Vliet et al., 2009). There are several possible process routes for FT fuel production and there is no full agreement as to which FT gasification system is the best, since it depends on how the system is evaluated and which parameters are considered most critical. For example, Tijmensen et al. (2002) investigated different routes for BG to FT fuels. Those authors showed that pressurized gasification can reach a higher overall energy efficiency and better economic performance than atmospheric systems. Hamelinck et al. (2004) proceeded from the results in (Tijmensen et al., 2002) and analysed the technological and economic performance of different cleaning and upgrading steps for different pressurized gasifiers. The authors of that study showed that the most profitable gasification concept consists of: a 25-bar oxygen-fired gasifier, followed by a tar cracker and wet gas cleaning and a solid–bed FT reactor with 70% once-through conversion. In contrast to the results by Tijmensen et al. (2002) and Hamelinck et al. (2004), Tock et al. (2010) found the atmospheric indirect gasification system followed by steam methane reforming to be both more economic and more efficient than indirectly heated circulated fludized-bed (CFB) gasifiers and indirect or direct heated entrained-flow (EF) gasifiers. Table 6 summarises the results from a number of techno-economic FT fuel studies found in the scientific literature.

Table 6. Summary of techno-economic FT fuel studies found in the scientific litterature. The price of fuel reported in the different papers are recalculated per liter of fuel but the currancy and the monetary value are remained in orignal units.

Reference	Size	Feed-to-Fuel efficiency	Type of Biomass	Gasifier	Price of fuel
(Boerrigter, 2006)	50-8500MW input	56%	Forest biomass	EF	1.1-0.54 €/l_{diesel}
(Ekbom et al., 2008)	289, 611MW input	44,46%$_{LHV}$	Forest residues	CFB	0.49,0.81 €$_{2008}$/$l_{jet\ fuel}$
(Hamelinck et al., 2004)	400 MW_{HHV} input	40-45%$_{HHV}$	Willow wood (15%moist.)	Direct CFB	0.61 €$_{2004}$/$l_{diesel\ (HHV)}$
(Kreutz et al., 2008)	548 MW_{LHV} input		Switch grass (15% moist.)	CFB	0.84-0.89[a] \$$_{2007}$/$l_{gasoline\ eq.}$
(Ng and Sadhukhan, 2011)	1350MW_{LHV} input	39-51%	Bio-oil	EF	78.7 €/MWh
(Martín and Grossmann, 2011)	227 ML/y ($FT_{gasoline}$ and FT_{diesel})	0.24 kg_{bifuel} /$kg_{biomass}$	Switchgrass	Ind. CFB	0.19\$$_{2011}$/$l_{biofuel}$
(Sarkar et al., 2011)	2000 dry t/d input		Forest residues	Ind. CFB[b] &CFB[c]	0.78-1.22 \$$_{2011}$/$l_{diesel}$
(Swanson et al., 2010)	2000 dry t/d input	39-50%$_{LHV}$	Corn stover	CFB or EF	1.06-1.32\$$_{2007}$/$l_{gasoline\ eqv.}$
(Tock et al., 2010)	20MW, 400MW input	59.8%$_{(FT\ crudel)}$	Wood (50% moist.)	Ind. CFB	53-106 €$_{2007}$/$MWh_{FT-crude,}$
(Tijmensen et al., 2002)	367 MW_{LHVwet} input	33-50%$_{LHV}$	Poplar wood (30% moist.)	Ind. and direct. CFB	0.32-1.08 \$$_{2002}$/$l_{diesel\ (LHV)}$
(van Vliet et al., 2009)	300, 400 and 2000 MW input	44-52%$_{LHV}$	Salix or eucalyptus	EF[d] and BFB	0.49-0.85 €$_{2005}$/$l_{FT\ fuel}$
(Wright and Brown, 2007)	569 $ML_{gasoline\ eq.}$/y	46%	Lignocellulosic feedstock	CFB	0.47\$$_{2005}$/$l_{gasoline}$

[a] with 0\$/t CO_2 eqvivalents, [b]the Silvergas technology,[c]The Renugas technology [d] Carbo-V technology

Several authors have studied the Fischer-Tropsch process based on exergy analysis, e.g. (Prins et al., 2005; Ptasinski, 2008; Sues et al., 2010), who have all shown that the largest exergy losses are found in the gasification section. Moreover, Kirkinen et al. (2009) have compared the greenhouse impact of FT fuel from forest-residues to fossil diesel. The authors showed that the greenhouse impact of forest residue-based FT is approx. one-third less than that of fossil diesel.

4.4 System studies of post-combustion CO_2 capture and biomass gasification to hydrogen and Fischer-Tropsch fuels

The potential for the integration of the development routes, considered in this thesis, with different industries has been studied by a number of authors. This section provides an overview of related system studies of post-combustion CO_2 capture and systems studies of BG-to-H_2 and BG-to-FT fuels.

Despite the high energy demand of the post-combustion CO_2 capture process, only a few studies have been found that include process integration, e.g.(Calin-Cristian, 2010; Harkin et al., 2010; Harkin et al., 2012; Hektor and Berntsson, 2007, 2008). Of those, Calin-Cristian (2010) focused on CO_2 capture at an IGCC and Harkin et al. (2011;2012) focused on CO_2 capture at coal power plants, while Hektor and Berntsson (2007;2008) focused on CO_2 capture in pulp mills. System aspects of integrating a post-combustion CO_2 capture plant with a pulp-mill have been studied in several papers by Hektor (2008), who includes the changes in the surrounding energy system when evaluating the cost and the avoided CO_2 emissions of CO_2 capture.

Several studies have been published on system studies of hydrogen via BG. Andersson and Harvey (2006, 2007) have published several papers on hydrogen production from black liquor integrated with a pulp mill. In those studies pulp-mill-integrated hydrogen production is evaluated with respect to CO_2 emissions and energy use. In the first paper the authors compared hydrogen production from gasified black liquor with electricity and methanol production from gasified black liquor. In the second paper the authors compared hydrogen production from gasified black liquor with stand-alone hydrogen production from gasified biomass. Rodin (2008) compared two gasification concepts for H_2 at a refinery with regard to global CO_2 emissions. The potential for substitution of a refinery hydrogen production unit with BG was recently studied by Brau et al. (2012), who evaluated the potential for gradual substitution based on energy use and CO_2 emissions. Different degrees of integration with the refinery and different polygeneration concepts were included in the analysis. The results in (Brau et al., 2012) show that process integration of the studied concepts increased both energy and exergy efficiencies.

System analyses of FT fuel production integrated with industries or other heat sinks such as district heating systems have recently been conducted by a number of authors, e.g. (Consonni et al., 2009; Egeskog et al., 2009; Ekbom et al., 2008; Isaksson et al., 2012; Joelsson and Gustavsson, 2012; Kirkinen et al., 2010; Walter and Ensinas, 2010). Most of those studies focus on the integration of BG-to-FT in the pulp and paper industry, whereas Walter and Ensinas (2010) have studied the integration of BG-to-FT fuel in an ethanol distillery. Ekbom et al. (2008) compared two routes for FT jet fuel in connection with two different district heating systems. The first route produced jet fuel at the gasification site, while the second approach produced an intermediate that was upgraded to FT jet fuel at an existing refinery. With energy prices at the time of the study, the authors concluded that production of an intermediate product for upgrading at a refinery BG-to-FT jet fuel would be an economically feasible option. Several authors have published studies on the integration potential of FT syncrude upgrading within a refinery, utilizing existing refining units (Gregor, 1990; Schablitzky et al., 2011).

4.5 Related work at industry level and interactions with adjacent infrastructures

The potential for CO_2 mitigation for the refining sector have been studied by several authors, e.g. (Alfke et al., 1999; Bernstein et al., 2007; CONCAWE, 2008, 2011; Rootzén, 2012). For example, Concawe (2008) has modeled the affect and consequences of legislation and expected demand changes in the EU refining industry by 2020, in terms of investments, total cost, energy and CO_2 emissions. The mitigation potential related to energy efficiency improvements, fuel switch and biofuel introduction was evaluated and compared to the targeted refinery emissions when following the overall ETS reduction goals. The results showed that these measures were not enough to reach the ETS goals for reduction in the petroleum sector. A similar approach has been applied by Rootzén (2012), who assessed the prospects for currently available abatement technologies to achieve significant reductions in CO_2 emissions from large stationary sources of CO_2 emissions, among them the petroleum refining sector. The authors concluded that unless a major breakthrough for new low-carbon process technologies is made, the CO_2 emissions from the petroleum sector will exceed the EU goal for reductions in greenhouse gas emissions (GHG) both in the medium term (2030) and in the long term (2050). Furthermore, Bernstein et al. (2007) assessed the CO_2 mitigation potential for the global petroleum refining industry in 2030 mainly based on the results from (Worrell and Galitsky, 2005). The abatement cost of CO_2 and/or the mitigation potential for petroleum industries at the national level can be found in, e.g. (Borba et al., 2012; Castelo Branco et al., 2010; Castelo Branco et al., 2011; Elkamel et al., 2008, Gomes et al., 2009; Holmgren and Sternhufvud, 2008; Park et al., 2010). Moreover, Murphy and Jaccard (2011) used the CIMS[11] method to explore how industrial (e.g. the petroleum industry) energy consumption and GHG emissions in Canada would be affected if a compulsory GHG emission reduction policy was implemented. The results showed that the reduction in GHG emissions in the petroleum industry when a CO_2 charge was introduced was primarily due to actions that reduced process heat and steam.

When evaluating the potential for CO_2 mitigation in European industry it is important to also consider the geographical location of a plant and the potential to use adjacent infrastructures. This importance has for example been stressed by, e.g. Wetterlund et al.(2012) , who uses a model, originally developed by Leduc et al.(2009), for identifying the optimal location of biofuel production plants. The potential for CO_2 capture in European energy-intensive industries has been studied by Rootzén et al. (2011), who also discuss how the geographical distribution of large CO_2 emitters and the distance to storage sites could affect the development of CO_2 transportation networks. In future work it is important to compare the findings from this thesis with findings in related work in other industries. For example, Jönsson and Berntsson (2012) compared the geographical position of large CO_2 emission point sources related to the Pulp and paper industry and Egeskog et al. (2009) investigated the European potential for the implementation of biofuel production based on BG in a district heating system.

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[11] CIMS is a hybrid model, which combines technological explicitness (bottom-up model) with economic realism (top-down model).

5 Studied systems and input data

This chapter describes the system studies presented in the appended papers. The different system boundaries are illustrated with figures in which material and energy flows are shown. A brief description of the input data for the different systems is also included. Economic input data, however, is described and presented in Chapter 7.

5.1 The EU oil refining sector

The general potential for CO_2 mitigation related to on-site emissions from EU refineries (+ Norway) was evaluated in **Paper I**. The CO_2 mitigation measures are however limited to measures that primarily reduce CO_2 emissions produced inside the refinery gates and thus are covered within the current EU ETS. For example, the production of renewable fuels is not covered in **Paper I**. Most of the refineries in the EU belong to the two simplest configurations described in Table 2. Only 18 EU refineries have been built with a large number of converting units and high complexity. The amount of on-site CO_2 emissions from the EU27 (+ Norway) refineries along with the distribution of refineries by configurations are presented in Table 7.

Table 7. Key characteristics of the EU and Norway refinery stock.

	Number of refineries divided by configuration					**Number of refineries**	**Crude oil [Mb/d]**	**CO_2 [Mt/y]**
	Base +1	**2**	**3**	**4**	**unidentified**			
EU + Norway	34	45	14	18	3	114	16	155

For the assessment and mapping of associated infrastructure in **Paper I**, the analysis was limited to refineries with CO_2 emissions exceeding 1Mt CO_2/y. These refineries (a total of 58 refineries) represent 83% of the total CO_2 emissions from EU refineries. Moreover, all refineries with Configuration 4 (i.e. the most complex refineries) were included among these 58 refineries. Data for the EU (+ Norway) petroleum industry was gathered from Chalmers Industry Database (see also 6.1.2). The data for the associated infrastructure was gathered from district heating associations, energy companies, company reports, national statistic, scientific papers and the Chalmers Industry Database.

5.2 The studied case refineries

Two different case refineries have been used in this thesis; a hydroskimming refinery, **Papers II** and **III**, and a complex refinery, **Papers III-V**. These refineries correspond to Configurations 1 and 4, which were described in Chapter 3.

The hydroskimming case refinery had a crude oil capacity of 6 Mt/y and emitted approximately 0.5 Mt CO2 per year. The complex case refinery had a crude capacity of 11.4 Mt/y and emitted approximately 1.8 MtCO2 per year. CO2 emissions from the oil refining process originated from several sources. In the analysis of post-combustion CO2 capture, only the largest CO2 emission sources have been selected for capture, see Table 9. The motivation for the key assumptions and input data in each of the appended papers (II-V) are described in the following text. Table 8 presents the type of case refinery and key assumptions in each of the appended papers.

Table 8. Overview of the case refineries and key assumptions used in the appended papers.

Paper	Type of case refinery/refineries	Status of the case refinery	Change
II	Hydroskimming	Future	The refinery is changed to produce maximum diesel and minimum gasoline, which leads to a deficit of hydrogen that must be met by a new hydrogen production plant.
III	Hydroskimming and complex	Current	None
IV	Complex	Future	The production is kept constant but the refinery is optimized for energy utilization.
V	Complex	Current	None

5.2.1 A future hydroskimming case refinery in Paper II

The aim of **Paper II** was to study the effect on the global CO_2 emissions if a future hydrogen demand is met by hydrogen production via BG. Several BG technologies were investigated and the results were presented as changes relative to a reference refinery in which the future hydrogen demand was met using conventional SMR. See Section 3.1 for the reasons for a hydrogen deficit. The complex case refinery has a hydrogen production plant, which makes it less sensitive to changes in the demand for petroleum products. The hydroskimming case refinery, however, is more sensitive to such changes since it does not have a separate hydrogen production facility and, therefore, needs to cover its hydrogen demand with hydrogen produced as a by-product in the production of gasoline. The hydroskimming case refinery was chosen in **Paper II** because it had a shortage of hydrogen and without a new hydrogen plant the refinery would not be able to meet the increased demand for diesel on the petroleum market. At the beginning of this research the intention was to include a study of BG to hydrogen in the complex case refinery, although in the context of making hydrogen production renewable. However, in the meantime a new research project was developed that focused on BG for hydrogen production in a complex refinery.

*Changes made to the hydroskimming case refinery in **Paper II***: The case refinery was assumed to adapt to the changes on the petroleum market by maximizing the production of diesel and minimizing the production of gasoline. The adjustment resulted in an increased deficit of hydrogen at the refinery. The crude oil capacity remained constant, while the production changes affected the steam demand and the electricity demand at the refinery. The reference case was the adjusted refinery in which an SMR was installed to cover hydrogen demand. Input data for the reference refinery was given from simulation models created by refinery personnel. Data for the excess heat from the refinery has been gathered by the author of this thesis and adjusted to future changes in consultation with refinery personnel. For key data for the case refinery in **Paper II**, the reader is referred to the appended paper.

5.2.2 A current hydroskimming case refinery in Paper III

In **Paper III** the cost for post-combustion CO_2 capture at a hydroskimming refinery was investigated. The reference refinery was the hydroskimming case refinery with a current product mix. The reason for keeping the current operation was the high investment costs that are related to both a new hydrogen plant and a CO_2 capture plant, and the assumption that it may not be profitable to combine two investments associated with high costs at the same time. Currently, most of the excess heat from this case refinery was heat exchanged with two district heating systems: one municipal district heating system and one company-owned heating system. In **Paper III**, it was assumed that for a future in which CCS is a commercially viable solution, the refinery will be able to choose the best option for the use of excess heat, whereby supplying CCS would be one of the options. However, the use of excess heat for CO_2 capture must be compared to other options such as district heating or drying biomass.

5.2.3 Current complex case refinery in Paper III and Paper V

The reference refinery in **Papers III** and **V** was the complex case refinery without any modifications. The current conditions were used since the complex case refinery is rather energy-efficient. There are, however, dissimilarities in **Papers III** and **V**, which are related to the input data for the refinery. Throughout the progress of this thesis the data for process streams in the complex case refinery was continuously updated. For this reason, the data for the refinery process is slightly different in **Paper III** than in **Paper V**.

In **Paper III** the data pertaining to the excess heat levels originated from the previous work by Berntsson et al. (2008). Complementary data for the excess heat as well as data for CO_2 emissions was collected by the refinery personnel. In **Paper V**, on the other hand, data for both CO_2 emissions and the process streams originated from a Pinch analysis recently performed at the case refinery (Andersson et al., 2013a).

5.2.4 A future complex refinery in Paper IV

In **Paper IV**, the analysis of the integration of an FT fuel production in an oil refinery was conducted assuming a future energy optimized refinery. The assumption originated from the fact that the integration of a FT fuel production is unlikely to occur in the near future. It is therefore reasonable to assume that before or in combination with such an implementation, extensive efforts will be put into making the existing refinery as energy efficient as possible. Therefore, the reference case for the analysis in **Paper IV** was an optimal heat integrated refinery, in which heat integration is performed within refinery process units but not between different refinery units. The basis for the analysis was the results from the energy audit that was used in **Paper V**. Since FT syncrude production has large amounts of excess heat, the key data relevant for the analysis in **Paper IV** is related to the heat demand in the energy optimized refinery. The reader is referred to the report by Andersson et al. (2013a) for more information and details about the assumptions and the optimized refinery.

5.2.5 CO_2 emissions considered for capture in Papers III and V as well as excess heat levels in Papers II, III and V

This section presents the levels of excess heat from the refining process used in each paper. **Paper IV** is not included since in that paper the refinery constitutes the heat sink and the gasification process constitutes the major heat source. The refinery constitutes a heat sink also in **Paper II**, but there the biomass is dried with excess heat from the refining process. The excess heat from the current and future hydroskimming case refinery is illustrated in Figure 3, while the excess heat from the current complex refinery in **Paper III** and **Paper V** is illustrated in Figure 4. In Figure 3 it can be seen that the excess heat from the hydroskimming refinery is only marginally affected by the change in production. Despite the difference in the origins of input data, there are only marginal variations in the amount of excess heat between **Paper III** and **Paper V**, which is illustrated in Figure 4. As can be seen in Figure 4, the amount of excess heat is similar at temperature levels above 50°C, which shows robustness in the excess heat data for the case refinery.

Table 9 represents the key assumptions pertaining to the emissions considered for the development route (post-combustion CO_2 capture) in **Papers III** and **V**. There are eight CO_2 emission sources in **Paper III** but only five in **Paper V**. The reason for this is that future emission sources connected to a planned coker unit were included in **Paper III** but not in **Paper V**. The emissions considered for capture, however, are only at the largest point sources (four in both papers) and these are the same in both papers, i.e the emissions from the planned coker are small compared to the other sources.

Table 9. Key Characteristics for CO_2 capture in **Papers III** and **V**.

Key Characteristics	**Hydroskimming case refinery**	**Complex case refinery**	
	Paper III	**Paper III**	**Paper V**
Crude oil capacity [Mt/y]	6	11.4	11.4
Total CO_2 emissions [Mt/y]	0.5	1.97	1.80
CO_2 emissions considered for capture [Mt/y]	0.48	1.74	1.71
Chimneys (CO_2 emission sources)	4	8	5
Number of sources considered for CO_2 capture	2	4	4

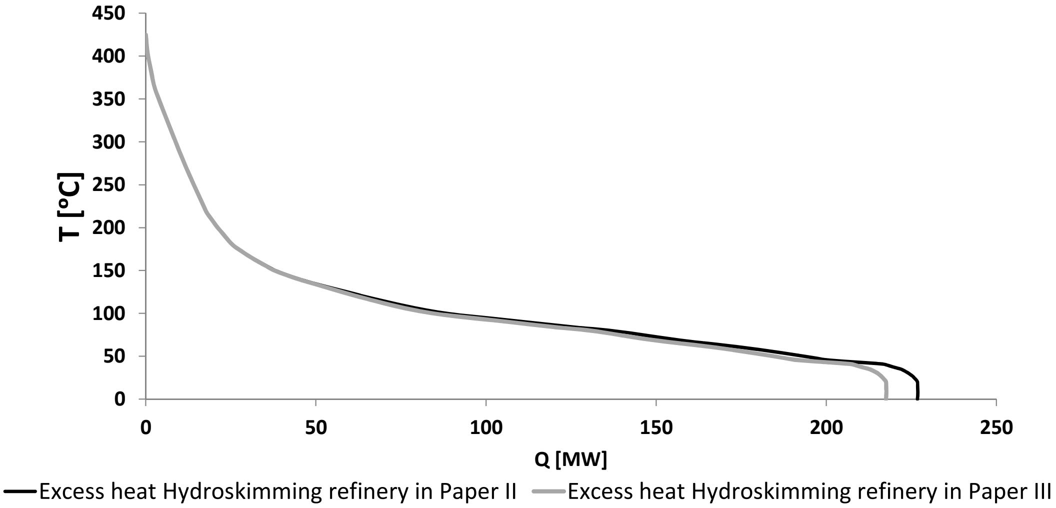

Figure 3. Excess heat levels from the hydroskimming case refinery in **Papers II** and **III**.

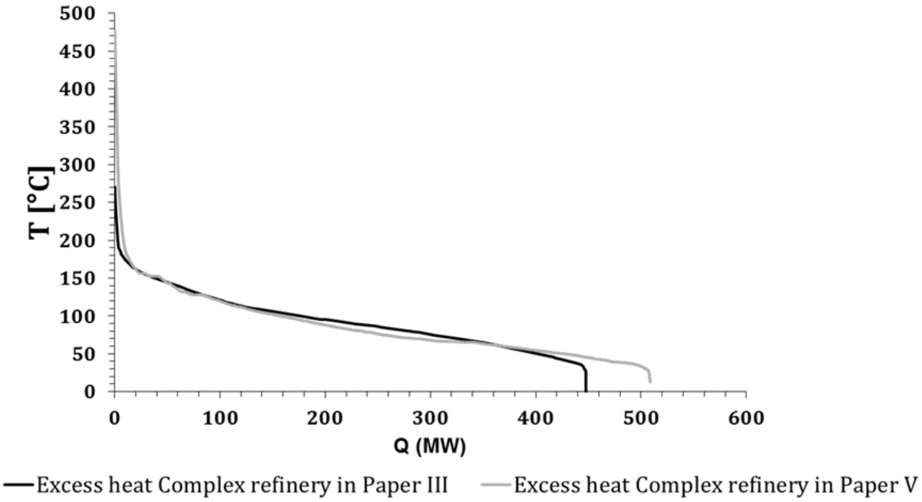

Figure 4. Excess heat levels from the complex case refinery in **Papers III** and **V**.

5.3 General aspects of the studied biomass gasification systems

There are a number of different types of biomass gasifiers: fixed-bed, bubble-bed, fluidized-bed and entrained-flow gasifiers. The gasifiers can also be directly or indirectly heated, pressurized or atmospheric. An indirectly heated gasifier is often reported in the literature as limited in scale, and several units must be built if a large-scale gasifier is needed. However, there are recently built direct CFB boilers that use the same technology as indirect gasifiers, with a capacity of 447-550 MW_{th} input (Finland OPET, 2012; Nevalainen et al., 2012). In this thesis three gasification technologies have been included; direct EF gasifier, pressurised direct CFB gasifier and indirect, atmospheric double-bed gasification (called DB hereinafter).

The different gasifiers have different setups, which result in different qualities of the product gas. Each technology has its pros and cons. During recent years several review articles, reports and books have been published describing the advantages and disadvantages of different gasification setups, e.g. (Basu, 2010a; Damartzis and Zabaniotou, 2011; Olofsson et al., 2005; Spath and Dayton, 2003). The following text gives a short summary of the gasification technologies and the gas cleaning steps that are valid for both hydrogen production and FT syncrude production, see Figure 5.

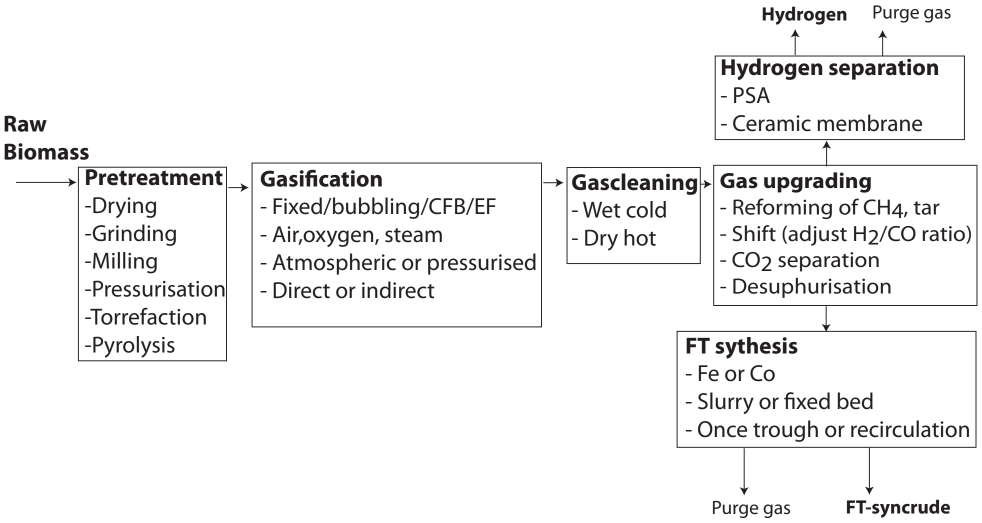

Figure 5. Overview of the gasification route from biomass to hydrogen and FT.

5.3.1 Pre-treatment and gasification

Biomass is dried prior to gasification to increase the efficiency of the gasifier. There are three main options for biomass drying; air drying, steam drying and flue gas drying. All three drying technologies are included in **Paper II**, while steam drying is considered for the FT fuel production in **Papers IV** and **V**.

The CFB gasifier is feed-flexible and suitable for large-scale production. However, the CFB gasifier brings about higher tar content than both the fixed-bed and the EF gasifiers (Olofsson et al., 2005). The fixed-bed gasifier is limited to small-scale gasification only (E4tech, 2009). The syngas from the EF gasifier has a high heating value and low tar content, but extensive pre-treatment of the biomass is needed in order to feed it into the gasifier, which may decrease the overall efficiency of the technology. There remain uncertainties about the efficiency of the pre-treatment technologies. Torrefaction and pyrolysis are two pre-treatment options. The pyrolysis process decomposes the biomass into pyrolysis oil that is fed to the gasifier with a yield up to $75\%_{wt}$ on a dry basis (Bridgewater, 2007). Gas and coke are produced as by-products in the pyrolysis process. The torrefaction process is a thermo-chemical treatment (in the absence of oxgen) that creates a solid uniform product with very low moisture content. Torrefied wood is shown to be easier to grind than untreated wood and it demands less energy for a given sizing (E4tech, 2009).

In addition to the gasification technology, the gasification medium is important for the quality of the gas. To obtain high gas quality (energy content, no nitrogen) oxygen is preferable as a gasification medium for the CFB and the EF gasifier (Basu, 2010a). The

CFB gasifier also allows for indirect gasification (which here is called double-bed (DB) gasification), where heat to the gasification process is provided by circulating bed material. The bed material is heated in a separated combustion chamber. One possible disadvantage of oxygen gasifiers is an increase in the energy penalty for oxygen production.

The gasification systems can be either atmospheric or pressurised. Hydrogen separation and synthesis to FT syncrude are commonly carried out at high pressure, which favours a pressurised gasification process. A pressurised system requires smaller equipment, but gives a more complicated process (e.g. more difficult to feed the biomass and needs more advanced material to withstand the pressure). Atmospheric gasification, on the other hand, is less complicated but requires larger equipment.

5.3.2 Gas cleaning

There are several cleaning and upgrading steps for the syngas for the gasification technologies. A tar cracker is preferable after the gasifier in order to increase the energy content in the gas and to convert the tar and remaining hydrocarbons into carbon monoxide and hydrogen. Three methods can be considered for this step; thermal cracking, catalytic cracking or scrubbing. A catalytic cracker gives a better gas yield and better tar reduction (Basu, 2010a). More about these methods and the formation of tar can be found in e.g. (Basu, 2010b).

There are several options for cleaning the gas from dust or particles: cyclones, fabrics or barrier filters, electrostatic filters or solvent scrubbers. The last cleaning step is a wet cleaning system or a hot cleaning system. Wet cleaning is better known than hot gas cleaning. The disadvantage of wet cleaning, however, is the large amount of steam needed (which gives lower energy efficiency). However, according to (Hamelinck and Faaij, 2001) the wet gas cleaning system is cheaper than the hot gas cleaning system, which compensates for the increase in steam supply.

5.3.3 Gas upgrading

If the syngas has relatively high methane content, the gas has to be reformed to increase the H_2 and CO_2 yield prior to hydrogen separation and FT synthesis. Two main processes are available for this purpose: autothermal reforming and steam reforming. Autothermal reforming has a simpler design and is possibly cheaper (Hamelinck and Faaij, 2001). The disadvantage is that oxygen is needed.

In order to achieve the preferred H_2/CO ratio, the final step before the synthesis to FT syncrude or hydrogen separation is a water shift reaction that adjusts this ratio. The shift reactions can occur in one step or in a dual step reactor. A dual reactor is preferable to shift as much hydrogen as possible (Hamelinck and Faaij, 2001).

An FT reactor catalyst is sensitive to impurities. Therefore, all sulphur must be removed prior to FT synthesis to avoid poisoning the FT reactor catalysts. This is typically achieved in a process based on a physical solvent that separates the acid gases and the CO_2 from the product gas which also has a negative influence on the FT catalyst.

5.3.4 Hydrogen separation

Hydrogen can be separated in a Pressure Swing Adsorption (PSA) unit or through a membrane. The PSA unit is a well-known process, where molecules are bound (adsorbed) to a surface at high pressure and released at low pressure. Examples of adsorbents are: activated carbon, silica gel, alumina and zeolite. For hydrogen purification, two steps of PSA beds are placed in series. The first PSA bed removes all the CO_2 and H_2O from the syngas, while the second bed removes all the residual gases from the hydrogen.

5.3.5 Fischer-Tropsch synthesis

In FT synthesis, (first invented by Franz Fischer and Hans Tropsch in 1938), a set of chemical reactions convert the syngas mixture of carbon monoxide and hydrogen into a range of liquid hydrocarbons at a pressure of 10-40 bar. The synthesis can be classified into two categories, the high-temperature Fischer-Tropsch (HTFT) and the low-temperature Fischer-Tropsch (LTFT). HTFT operates at high temperatures (300-350°C) and favours the production of low molecular weight olefins (C3-C11). HTFT syncrude is the easiest to refine, and is preferable if motor gasoline is the main product. LTFT operates at lower temperatures (200-250°C) and yields high molecular weight linear waxes (>20 carbon molecules). LTFT syncrude is easier to refine into jet and diesel fuels (de Klerk, 2011). There are three main types of FT reactors: the fluidized reactor, the fixed bed reactor and the slurry phase reactor. The fixed bed and the slurry phase reactor are design for production of waxes and diesel, while the fluidized bed reactor is developed for gasoline and light olefin production (Olofsson et al., 2005). Mainly two catalysts are used in FT synthesis; Fe-catalyst or Co-catalyst. The Co-catalyst is more expensive than the Fe-catalyst, but due to its higher activity and longer lifetime, it is often the preferable LTFT catalyst (Dry, 2002; Kumar et al., 2009).

Recycling FT off gases maximise the production of FT syncrude. However, a once-through conversion approach[12] can combine fuel and electricty generation and thereby higher efficiency and is assumed to have lower investment costs (Hamelinck et al., 2004).

In upgrading to FT fuels, the FT syncrude is distilled to produce fractions of gas, naphtha, diesel and wax. These fractions are further processed through a series of refining steps, such as hydro cracking, hydro treating, isomerisation and reforming, before the final FT gasoline and FT diesel are produced.

5.3.6 Specific aspects of the hydrogen production in Paper II

Part of the aim in **Paper II** was to compare different BG routes for hydrogen production. Figure 6 illustrates the studied biomass routes.

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[12] Here, the FT off gases are not recycled back to the FT synthesis.

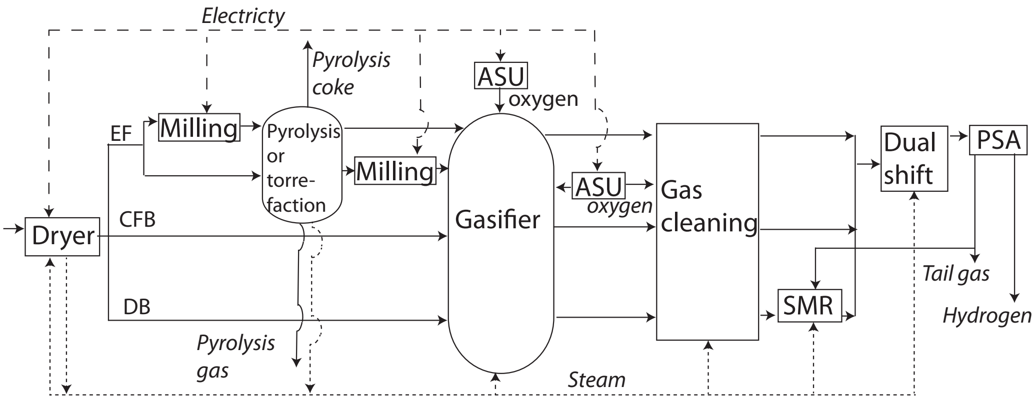

Figure 6. Schematic representation of the studied gasification routes for hydrogen production in **Paper II**.

In **Paper II,** the different systems were simulated in Aspen Plus®. All input data for the gas cleaning to the final product was taken from (Hamelinck and Faaij, 2001) and the gasification processes were simulated by Eva Larsson at TPS (2010). The system boundaries in **Paper II** are illustrated in Figure 7. Hydrogen and steam from the SMR and the BGs were utilized in the refining processes, and replaced natural gas in the boilers. All data, with original sources, is further described in the appended paper (**II**).

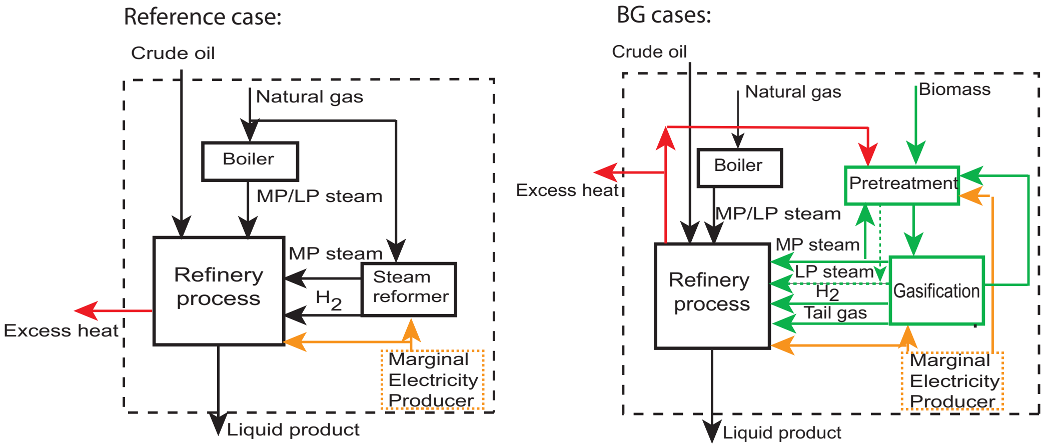

Figure 7. System boundaries for hydrogen production in comparison with an SMR at a hydroskimming case refinery.

5.3.7 Specific aspects of FT fuel production in Paper IV and Paper V

In contrast to the BG-to-H_2 analysis in **Paper II** where several BG routes were considered, only one BG system was evaluated in **Papers IV** and **V**. The BG system in **Papers IV** and **V** is based on a simulation model by Isaksson et al. (2012), who based their model on results from different steps from several authors (Ekbom et al., 2008; Hamelinck and Faaij, 2001; Larson, 2006; Spath et al., 2005). Figure 8 presents a schematic overview of the FT syncrude production and upgrading used in this thesis. For a more thorough description, the reader is referred to the appended paper (**IV**).

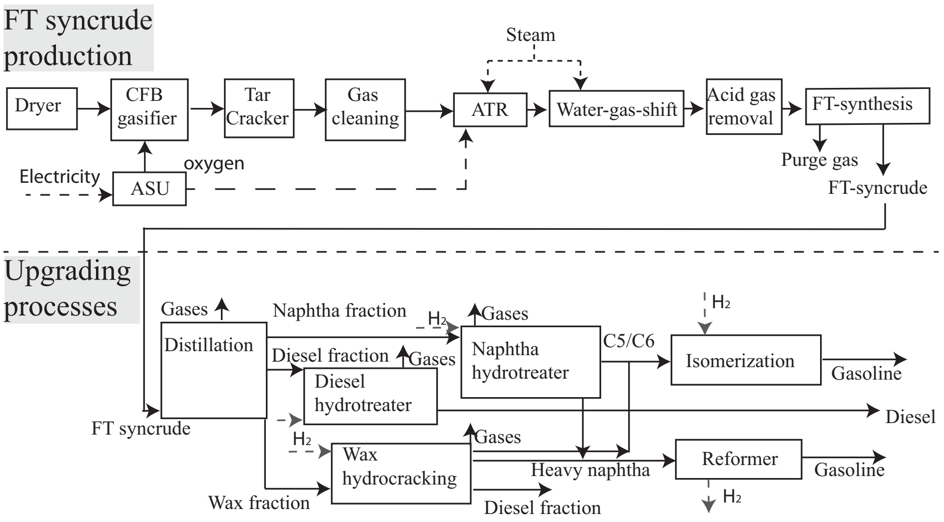

Figure 8. Schematic picture of the FT syncrude production and upgrading of the FT syncrude in **Papers IV** and **V**.

FT fuel production is evaluated for two cases:

- Case heat integrated (HI): The FT fuel is produced at the refinery, i.e. the FT syncrude production is heat integrated with the refinery.
- Case stand-alone (SA) : The FT syncrude is produced in a stand-alone facility but the FT syncrude is upgraded at the refinery in existing or in new units.

The gasification process constitutes a heat source that provides the oil refinery with steam and heat via a molten salt heat transfer system. Figure 9 presents an overview of the flows and the system boundaries for the FT fuel production in **Papers IV** and **V**. The illustration represents both the HI and the SA case. For further information the reader is referred to the appended papers (**IV** and **V**).

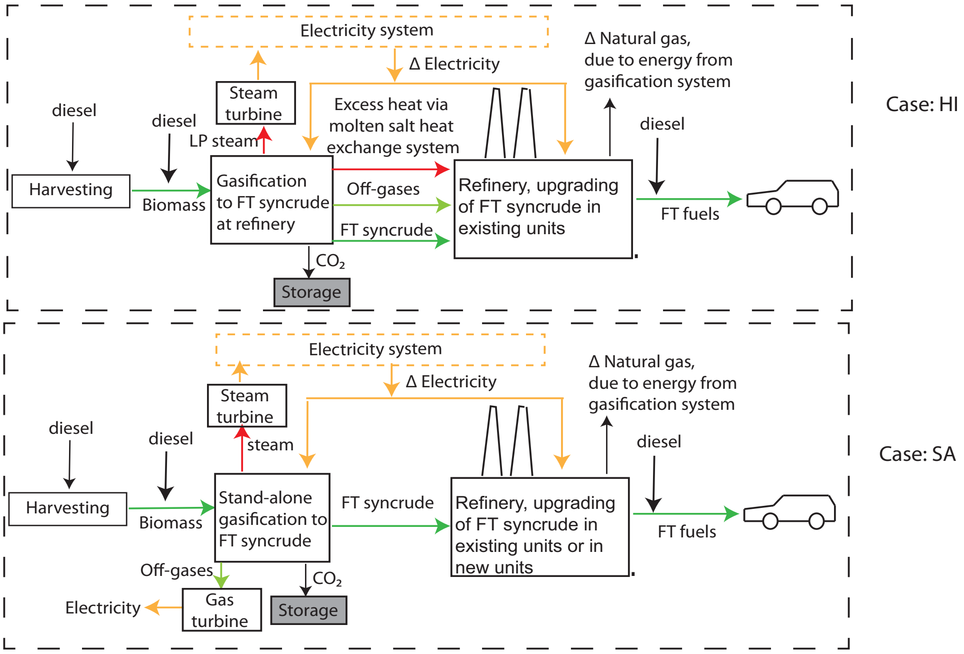

Figure 9. Overview of the material flows in the HI and the SA cases.

5.4 Post-combustion CO_2 capture and heat supply options

Currently, the most common technique for post-combustion capture of CO_2 is chemical absorption with amines as the solvent (Pires et al., 2011). Several chemical solvents are applicable to post-combustion capture, including monoethanolamine (MEA), chilled ammonia, KS solvents, aqueos ammonia, methyl diethanolamine and diethanolamine. For a review the reader is referred to the paper by (Calin-Cristian, 2010). MEA was used as the chemical absorbent for the post-combustion process in this thesis because it is both commercially available and applicable to large-scale CO_2 capture (Hardisty et al., 2011).

The post-combustion process removes CO_2 emissions from flue gases after the combustion of fuels. CO_2 emissions are captured from large point sources and gathered. The CO_2 molecules are then absorbed[13] to an MEA solution in an absorption column. A CO_2 lean stream (15% of the CO_2 emissions) leaves the column, while a CO_2 rich solution is heat exchanged and fed to a desorption column. Here, the CO_2 molecules are separated from the solution by the regeneration of the absorbent, which is a very heat demanding step. The top stream from the desorption column consists of CO_2 and water and after condensing the water, the pure CO_2 stream is compressed to between 75 to 200 bar.

There are uncertainties about the heat demand needed per kg CO_2 captured. Typically, MEA systems are reported to have a regeneration requirement around 3700 kJ/kg CO_2 (Abu Zhara, 2009, Karimi et al., 2011; Kim et al., 2011; Moser et al., 2011; Pfaff et al., 2010). This number is used in **Paper V** where the post-combustion process is compared to the FT fuel process in an oil refinery. There are, however, researchers that have reported both lower and higher energy requirements. For example, a recent study by Karimi et al, revealed that post-combustion capture with MEA could reduce regeneration requirements down to 2722 kJ/kg CO_2 if the capture unit was built with a vapour recompression unit. There are previous studies that have reported less optimistic values, up to 4700 kJ/kg CO_2 (Haugen, 2009). Due to this uncertainty, two levels of heat demands are investigated in **Paper III** (2800 kJ/kg CO_2 and 4700 kJ/kg CO_2). Further, the reboiler in the desorption unit in the post-combustion process is, here assumed to have a temperature of 120℃. A lower temperature level amounts to a higher specific heat demand in the capture process, see studies from, e.g. (Abu-Zahra et al., 2007; Andersson et al., 2013b). It is further assumed that the specific heat demand for the capture process is constant for the CO_2 concentration in the appended papers (5- 12 $\%_{vol}$). This is in accordance with Li et al. (2011), who have shown that the specific reboiler duty decreases sharply between 1 to approx. 6 $\%_{mol}$ but beyond this range the duty continues to decrease at a much lower rate. Furthermore, a higher concentration of CO_2 results in a smaller absorber tower, and a decrease in investment costs.

One of the main focuses of this thesis is the evaluation of different options for supplying the heat needed for the regeneration of the absorbent in the CO_2 capture process. The different heat supply options studied are:

- Production of steam in a natural gas combined cycle (NGCC)

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[13] With an assumed efficiency of 85%.

- Production of steam in a natural gas boiler with power production (NB)
- Production of steam in a biomass boiler with power production (BB)
- Production of steam from excess heat from the refinery (directly or with the aid of a heat pump) (EH &HP)

The different heat supply options together with a schematic picture of the post-combustion process are provided in Figure 10. For a thorough description of the heat supply options and the corresponding input data the reader is referred to **Paper III**

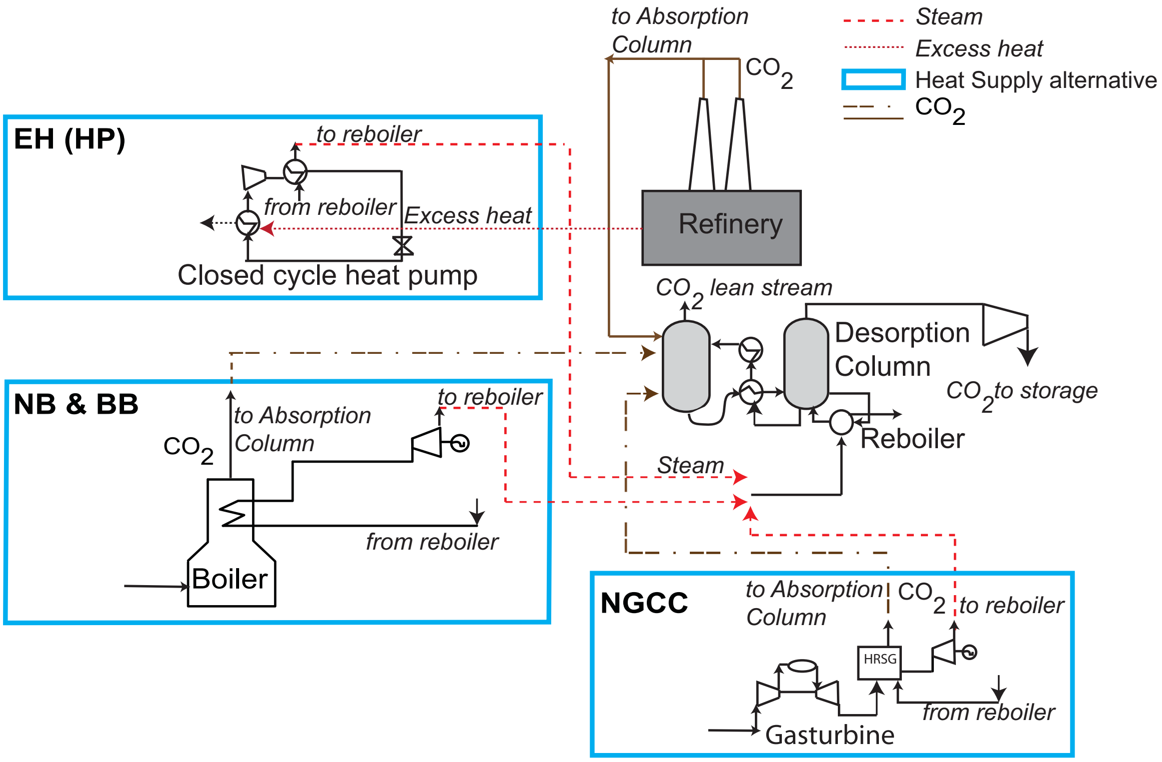

Figure 10. Schematic presentation of the different heat supply options together with a schematic picture of the post-combustion process. The red dotted lines represent the heat delivered to the capture unit reboiler, the brown lines represent the CO_2 streams, and the blue boxes represent the different heat supply alternatives.

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6

Methodologies

This chapter describes the general methodology of this thesis, and the specific methodologies used in the different appended papers. The methodology in the first paper is of a more general character with a higher systems perspective, while the methodology in the following papers is of a more specific systems analysis character.

All papers are based on a bottom-up approach represented by current oil refineries, or a case refinery, and of emerging technologies. The methodology in **Paper I** differs from the methodology in the other papers. While **Paper I** assesses the potential for CO_2 abatement for the entire EU petroleum industry stock, the other papers assess the potential for CO_2 or GHG emissions reduction for specific technologies at a case refinery.

All the papers appended to this thesis include an assessment of the impact on global GHG emissions. **Papers I-III** focuse on CO_2 emissions only, while the two latter papers expand the analysis to include GHG emissions such as CH_4 and N_2O. Two of the papers (**III** and **V**) include a techno-economic evaluation of the integration of CO_2 capture and the integration of FT fuel production at a refinery. In these studies a scenario approach is used for the evaluation of costs and GHG emissions. In three of the papers (**I, III** and **IV**) the current refinery structure is the basis for the integration and evaluation of the studied technologies. In **Paper II** the reference refinery is a refinery with a future production mixture that has been changed in order to meet future demand for fuel. In **Paper IV** an otimal heat integrated refinery is the basis. The methodology in **Paper I** was developed by the authors, while the methodology used in the other papers was a combination of methods and methodologies developed by others. An overview of the methodologies used in the different papers is found in Table 10.

Table 10. Overview of the methodology assessment in the appended papers.

Methodology approach	**Papers**				
	I	**II**	**III**	**IV**	**V**
Assessment of the whole sector	x				
Assessment of a case refinery		x	x	x	x
CO_2 analysis	x	x	x		
GHG analysis				x	x
Techno-economic assessment			x		x
Current refinery approach	x		x		x
Future refinery approach		x		x	

6.1 Methodology approach for analysing the oil refining industry at sector level

In **Paper I**, the assessment of CO_2 mitigation potential for the petroleum sector is limited to the change in CO_2 emissions related to the refining process (on-site emissions covered within the current EU ETS), and includes mapping of access to infrastructures such as district heating networks, natural gas grids, chemical industries, possible CO_2 storage sites and possible CO_2 capture clusters[14]. Figure 11 illustrates the methodology approach in **Paper I**.

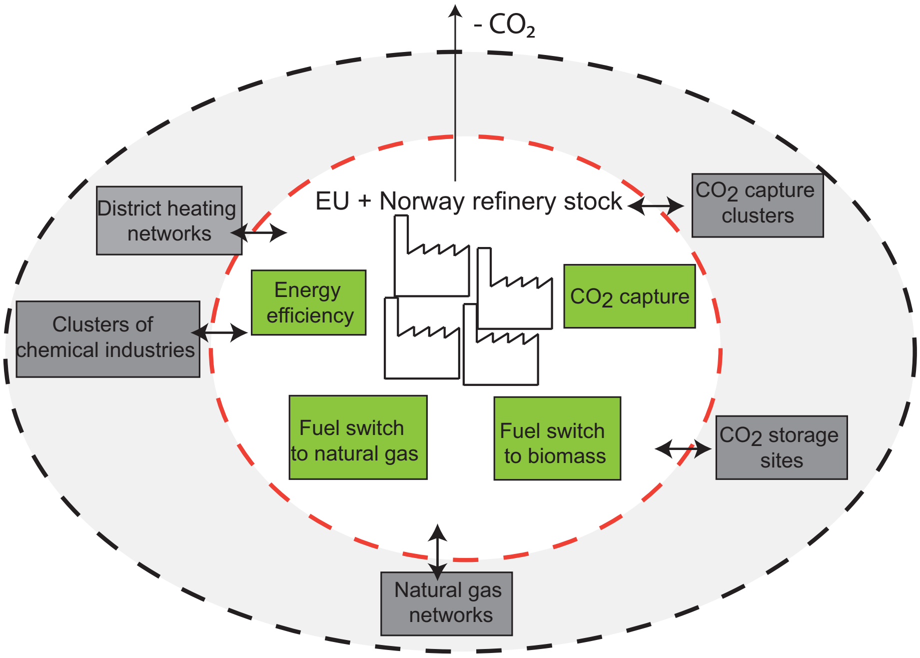

Figure 11. An illustration of the systems approach used in **Paper I**, where the CO_2 mitigation potential is assessed for several abatement options and where mapping of access to adjacent infrastructure is included. The change in CO_2 is only related to on-site emissions.

6.1.1 General methodology approach for Paper I

In **Paper I**, the potential for CO_2 mitigation in the EU refining industry is assessed in terms of improving energy efficiency, switching fuel and CCS. The general methodology approach involves:

- ***Assessment of key characteristics*** of the EU refinery sector (e.g. CO_2 emissions, crude oil capacity, complexity)
- ***Assessment of future trends*** in the petroleum market (e.g. fuel demand and fuel quality requirements)
- ***Identification of suitable mitigation options***

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[14] For the definition of a CO_2 capture cluster, the reader is referred back to 3.2.

- ***Assessment of adjacent infrastructures*** (district heating networks, natural gas grids, possible CO_2 storage sites, potential CCS clusters and chemical industries)
- ***Analysis of the aggregated potential*** for on-site CO_2 reduction at EU refineries

The assessment of the adjacent infrastructure entailed mapping adjacent infrastructure with the potential to facilitate on-site or global CO_2 mitigation, The adjacent infrastructures with the potential to influence on-site CO_2 emissions were considered when analysing the aggregated potential for on-site CO_2 reduction, i.e. possible CO_2 storage sites, potential CCS clusters and natural gas grids.

6.1.2 Chalmers Industrial Energy Database

As part of **Paper I**, the description and the assessment of the EU petroleum refining industry and the adjacent energy infrastructures were based on the Chalmers Industry Energy database (as described in bullet points one and four in previous section). The database was designed to cover both the supply side and the demand side of the European energy system (Kjärstad and Johnsson, 2007) and is divided into a set of sub-databases: the Chalmers power plant database (Chalmers PP db), the Chalmers fuel database (Chalmers FU db), the Chalmers CO_2 storage database (Chalmers CS db), the Chalmers member states database (Chalmers MS db) and Chalmers industry database (Chalmers IN. db).

6.2 Methodology approach for analysing the oil refining industry at case refinery level

6.2.1 General methodology approach in Papers II-V

To varying extents, these papers are based on the methodological approach of system expansion for all flows from a life cycle perspective. The general methodology steps are:

1. ***Define the system boundaries and the surrounding*** systems that affect the studied system. Systems left out do not affect the studied system, nor are they affected by streams leaving the studied system.

2. ***Define the reference system*** with which the GHG emissions and the economic performance is compared (see Chapter 5 for the different reference systems).

3. ***Choose methodology*** to evaluate the effect of the surrounding systems (in this thesis a scenario and marginal technology approach is used).
4. ***Determine the current level of excess heat*** at the case refinery that can be made available for heat integration with a CO_2 capture plant or ***identify the heating demand*** in the refinery processes for heat integration with a BG-to-H_2 process or a BG-to-FT process.
5. ***Calculate energy balances*** for the studied development routes (in **Paper II**, simulation models were built for this purpose while the other papers use information found in the literature).

6. ***Assess the potential*** for the studied development routes to be heat integrated with the case refinery.
7. ***Calculate energy balances*** for the studied systems after heat integration of the development routes with the case refinery .
8. ***Analyse the net annual profit and/or GHG emission reduction*** by using future energy market scenarios.
9. ***Vary key parameters*** to analyse how sensitive the results are to changes in these parameters.

The methodology approach has been adapted from several sources and adjusted to the assessment of GHG emissions and economic performance in an oil refinery. For example, Jönsson (2011) uses a similar methodology to analyse different technology pathways for a future pulp and paper mill. However, Jönsson (2011) uses, in contrast to the methodology in this thesis, an optimisation tool to estimate the potentials for CO_2 emission reduction and revenues for different future plausible options. Pettersson (2011) is another example of an author that has used a similar methodology approach, but again for a pulp and paper mill. The methodology approach also builds on ideas from Hektor (2008), who developed a methodology for the integration of a post-combustion CO_2 capture plant in the pulp and paper industry. Steps 3 and 8 are a methodology originally developed by Ådahl and Harvey (2007) and most recently updated by Axelsson and Harvey (2010). Within our research group[15] this methodology has been used for analysing profitability and CO_2 emissions especially for the integration of future biorefinery concepts. Andersson (2007), Axelsson (2008), Hektor (2008), Jönsson (2011), Pettersson (2011), and Svensson (2012) are examples of authors that partly base their work on the same methodology (steps 3 and 8) as this thesis. Steps 4, 6 and 7 build on the idea of the pinch analysis method, which was originally developed by Linnhoff and Flower (1978).

6.2.2 System analysis of GHG emission balances and economic performance

In the appended papers (**II-V**) an expanded systems approach was used for the evaluation of the three development routes and their GHG emissions and economic profitability. This approach has previously been used by several authors, for example by Jönsson (2011) and Pettersson (2011) for comparing various technology pathways in the pulp and paper industry and by Wetterlund (2012) and Andersson (2007) for comparing different products from the gasification of biomass. By using an expanded systems approach, focus is on interactions and the arrangement of components in the system and between the system and its surroundings. Therefore, all energy streams entering or leaving the studied system are assumed to cause a change in surrounding systems, such as the electricity system or the natural gas system, as illustrated in Figure 12.

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[15] Heat and Power Technology, Chalmers University of Technology

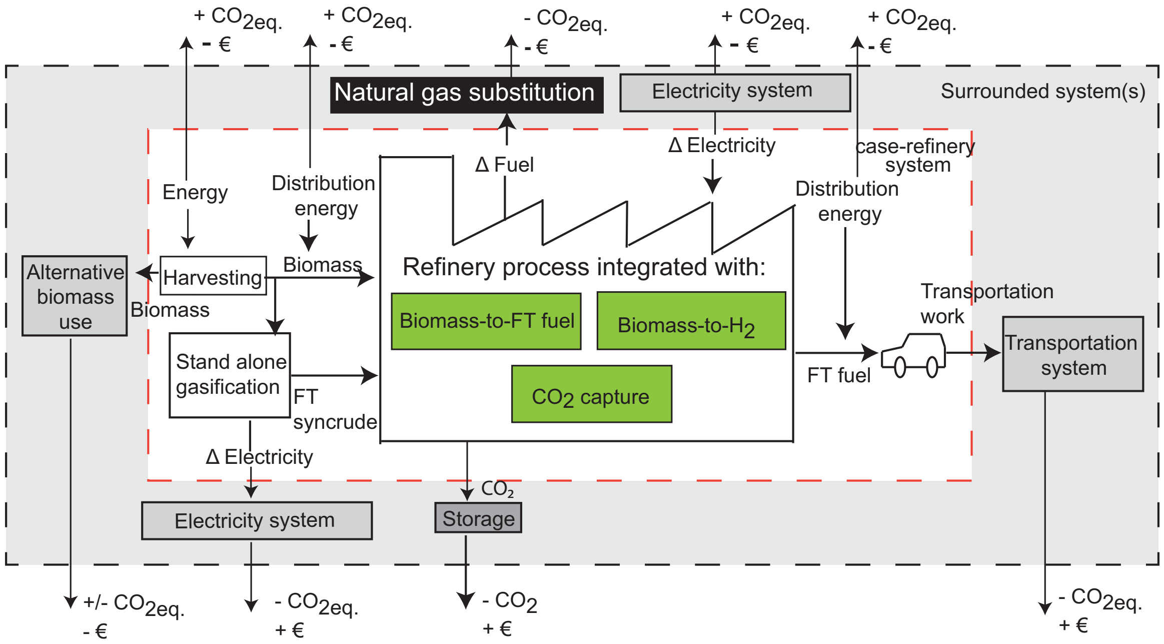

Figure 12. An illustration of the systems approach used in **Papers II-IV**. The arrows represent the net energy streams entering or leaving the system and how these interact with the surrounding systems. The GHG emission effect and the economic value of each flow are indicated with +/-, where + means an increase in $CO_{2eq.}$ or a revenue and – means a decrease in $CO_{2eq.}$ or a cost.

The implementation of development routes in a case refinery was evaluated for several conditions in the surrounding energy systems. These conditions were given by using energy market scenarios, which are described in more detail in Sections 6.3 and 7.2. For the evaluation of both GHG emissions and economic performance, a European perspective was used. In the following text, the reference surrounding systems are briefly described.

Electricity produced or consumed in the studied systems affects marginal electricity production. Since the timeframe in the present study is relatively long, base load build margin rather than operating margin is considered. The base load build margin approach determines a likely type of electricity generation facility that will be built when new generation capacity is installed. Despite a large share of renewables and increasing development of solar and wind power capacity, it is assumed that the base load build margin electricity produced in the modelled time period (2030) will still occur in condensing power plants fired with fossil fuels. In **Papers II**, **III** and **V** the marginal electricity is assumed to be coal power plants or coal power plants with CCS as a possible marginal electricity producer, while **Paper IV** also includes NGCC as a possible marginal electricity producer in the analysis. Solar and wind power are not considered as build margin due to the fluctuation in capacity.

The reference system for the transportation market (used in **Papers IV-V**) is fossil fuel based. It is assumed that the produced FT diesel and FT gasoline will replace otherwise fossil-based diesel and gasoline with a 1:1 ratio (based on energy content).

In two of the papers (**II** and **IV**) and in the discussion part of **Paper V** the system has been expanded to include alternative users of biomass. In those papers, it was assumed that biomass will become a limited resource in the future, thus additional demand for biomass in new processes will lead to less biomass being available for other parts of society. The alternative user was assumed to be the large-volume user with the highest willingness to pay. Co-combustion of biomass in coal-power plants was regarded as the only alternative biomass user in **Paper IV**. **Paper II** also regards DME production as an alternative biomass user, while **Paper V** regards FT fuel production as an alternative biomass user along with coal power plants.

6.3 Energy market scenarios

There are strong links between a number of energy market parameters, such as fuel prices, the price of electricity, marginal electricity production technology and its associated GHG emissions, the charge for CO_2 as well as different policy instruments. An approach to accounting for consistent interrelations between energy market parameters is to use energy market scenarios (Ådahl and Harvey, 2007). Energy market scenarios constitute a packaged sensitivity analysis with respect to energy-related costs, revenues and GHG emissions. The following text will give a brief introduction to the scenario tool used in the appended papers. For a more thorough description of the tool and the assumptions behind it, the reader is referred to the report by Axelsson and Harvey (2010).

In **Papers II**, **III** and **IV** the energy market scenarios are used to reflect a variety of plausible future energy market conditions that will affect the revenues and the reduction in GHG emission of the studied technologies. In those papers, a calculation tool, the ENPAC[16] tool, developed by (Axelsson and Harvey, 2010) based on (Axelsson et al., 2009; Ådahl and Harvey, 2007) is used. The ENPAC tool calculates future energy market prices for large-volume customers based on world market fossil fuel price and assumed values for energy and climate policy instruments. The user input to the tool are the fossil fuel prices on the world market, policy instruments for CO_2 emissions, renewable electricity and motor fuel production as well as the assumption of the availability of a CCS structure. The output is consistent energy prices as well as associated GHG emissions. An overview of the ENPAC tool is given in Figure 13.

The ENPAC tool also includes a number of parameters associated with a number of energy conversion technologies that are possible candidates for the marginal production of electricity and alternative use of woody biomass. For example the tool includes data for investment costs, transportation and operation costs, efficiencies and annual production capacities.

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[16] Energy Price and Carbon Balance Scenario tool

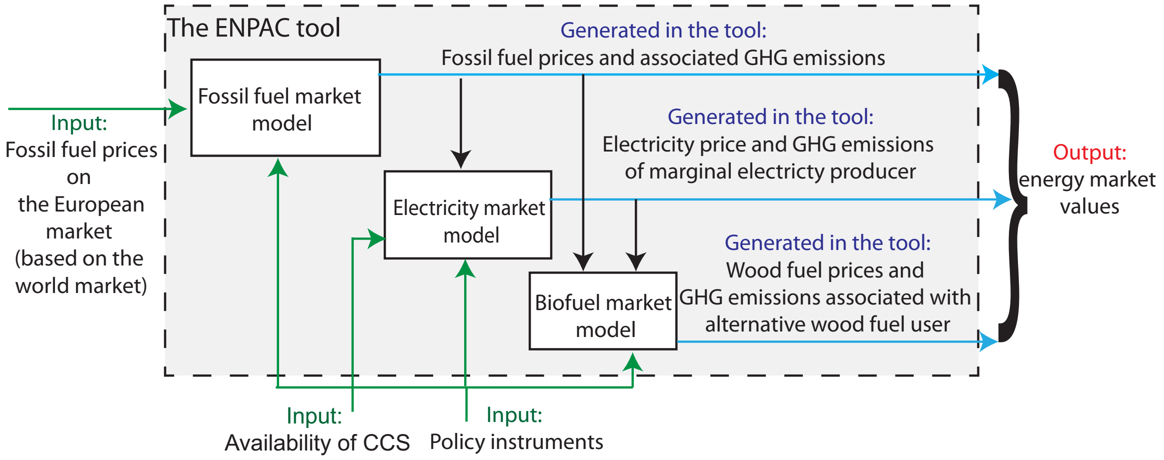

Figure 13. Overview of the ENPAC tool; input requirements, generated parameters and output values. (Adapted from (Axelsson and Harvey, 2010), p.5, Fig.2).

6.3.1 Additional sensitivity analysis

In **Papers III** and **V**, an additional sensitivity analysis of the results is conducted with respect to a number of parameters that are identified as having potentially strong influences on the economics. Examples of such parameters are: the capital recovery factor, the heat demand for the CO_2 capture process, the level of support for renewable electricity and motor fuel production. See the appended papers for the specific parameters investigated in the different papers.

6.4 Techno-economic evaluation

In **Papers III** and **V**, a techno-economic evaluation has been included. Projected commercial ("Nth plant") performance and costs are assumed. The investment costs are taken from previous studies investigating similar systems. In the cases where cost data was presented as equipment costs, the data was adjusted by an installation factor[17] to represent the investment costs. There is significant uncertainty about the investment cost for new, emerging technologies. In general, methods for estimating investments result in uncertainties about the final costs by approximately +/-30%.

The base year of 2010 is used for the techno-economic evaluation, and all investment data has been adjusted accordingly using the Chemical Engineering Plant Cost Index (Chemical Engineering, 2011) and currency indexes. The operating and maintenance (O&M) costs are assumed as 4% of the capital cost for all investments except for the CO_2 capture plant where the O&M costs are taken from previous studies at the case refineries. However, the costs in general (either equipment cost or investment cost) are scaled from the base cost and base size of the unit as follows:

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[17] By using an installation factor the direct costs for the installation of a new plant are included. These are, in addition to equipment cost: 1) equipment erection, 2) piping, 3) electrical, power, lighting, 4) instruments and automatic control systems, 5) process buildings and structures, 6) ancillary building, offices, laboratory buildings, workshops, 7) storage, 8) utility services, 9) site preparation.

$C=C_0*(S/S_0)^f$ (1)

where f is the cost scaling factor, typically ranging between 0.6 and 0.8, C is the cost of components, S is the size of the component. In the cases where the cost scale factor was not given a scaling factor of 0.7 was used.

To calculate the prices on a yearly basis, an annualisation method was used. As a base case, a capital recovery factor (CRF), also termed the annuity factor, of 0.1 was used[18]. This CRF has been based on the assumption that investing in a BG-to-FT fuels or CO_2 capture plant can be viewed as strategic investments and therefore is allowed to have a longer payback period. The same argument was used in, e.g. (Haugen, 2009; Hektor, 2008; Svensson, 2012). A detailed analysis of the interest rate and the economic lifetime is beyond the scope of this thesis, but the appended papers contain a sensitivity analysis where the CRF is changed from 0.1 to 0.2[19]. The change represents a shorter economic lifetime but can also be seen as a change in the investment costs. Both the investments and the capital costs are uncertain, therefore a large span has been used to study the influence. The net annual profit was used to evaluate economic performance, see Section 5.2.

In **Paper III**, the profitability of a CO_2 capture plant is evaluated by calculating the avoidance cost and comparing the resulting price with the CO_2 charge. The avoidance cost from a refinery perspective is in that paper defined as Eq. 2 and the avoidance cost from a global perspective is defined as Eq. 3:

$$C_{\text{avoided (on-site)}} = C_{\text{annual}} / CO_{2\ \text{refinery-avioded}} =$$
$$= C_{\text{annual}} / (CO_{2\ \text{from refinery before CCS}} - CO_{2\ \text{from refinery after CCS}})\ [€/t\ CO_2] \quad (2)$$

$$C_{\text{avoided (globally)}} = C_{\text{annual}} / CO_{2\ \text{gobally-avioded}} =$$
$$C_{\text{annual}} / (CO_{2\ \text{from refinery before CCS}} - CO_{2\ \text{from refinery after CCS}} +/- \Delta CO_{2\ \text{from electricity export/import}})\ [€/t\ CO_2] \quad (3)$$

where C_{annual} is the annualised capital and operation cost for the capture plant.

6.5 Process integration

Process integration was defined by the International Energy Agency (IEA) at an expert meeting in Berlin in 1993 as "*systematic and general methods for designing integrated production systems ranging from individual processes to total sites, with special emphasis on the efficient use of energy and reducing environmental effects*" (IEA, 2007).

All of the appended papers include thermal process integration to some extent. In all of the papers, heat integration refers to the concept of thermally heat integrating heat sources and heat sinks of a process or a system in order to reduce the external hot and cold utility,

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[18] A CRF of 0.1 is equal to e.g. an economic lifetime of 25 years and an interest rate of 8% or an economic lifetime of 15 years and an interest rate of 5%.

[19] A CRF of 0.2 is equal to e.g. an economic lifetime of 6 years and an interest rate of 5% or an economic lifetime of 10 years and an interest rate of 15%.

(i.e. to save primary energy). The pinch methodology[20] is a systematic tool for process integration that has been used for decades and for several applications, varying from small isolated processes to large clusters of industries. For a thorough description of this methodology, the reader is referred to recent publications by Kemp (2007), Smith (2005), and Klemeš et al. (2011). The aim of a pinch analysis is often to minimize the use of energy by maximizing internal heat recovery in a process or between processes. The methodology can be applied to both existing processes and grass roots designs. What separates the pinch methodology from other integration methodologies is that it is a graphical tool that facilitates easy understanding. The amount of internal heat exchange possible in a stream network or the total heat and cooling demand is, in the pinch analysis methodology, illustrated with composite curves (CC) or grand composite curves (GCC). The GCC is plotted as the net heat flow against shifted interval temperatures. The interval temperatures are shifted to include the temperature difference (driving force) necessary for heat exchange between streams, see Figure 14. In **Papers II**, **III** and **V** where one purpose has been to identify the amount of excess heat from the case refinery the Actual Cooling Load Curve (ACLC) was considered for the analysis. The ACLC represents the streams that currently are cooled with utility (e.g. air, water) at real temperatures. For more information about these curves, the reader is referred to (Nordman, 2005). In **Paper IV**, however, a theoretical approach was assumed. In this paper the reference refinery was an optimal heat integrated refinery, represented by the GCC of each unit at the refinery (16 in total).

To study the potential steam generation in the BG-to-H_2 process in **Paper II** and to evaluate the electricity generation and the excess heat from the BG-to-FT syncrude process in **Papers IV-V** the pinch analysis approach, called background/foreground (BG/FG) analysis (for split GCCs), has been used. A BG/FG is typically conducted to find solutions for process integration. The sub-process of interest is presented by one GCC curve (FG), while the main process is represented by another GCC curve (BG) in the same plot. Using this approach, it is possible to determine and find process integration possibilities. Figure 14 shows an example of a BG/FG analysis. The red line represents the background process (in this case all streams in the FT syncrude process), while the dashed line represents a steam power cycle.

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[20] First developed by Linnhoff and Flower (1978).

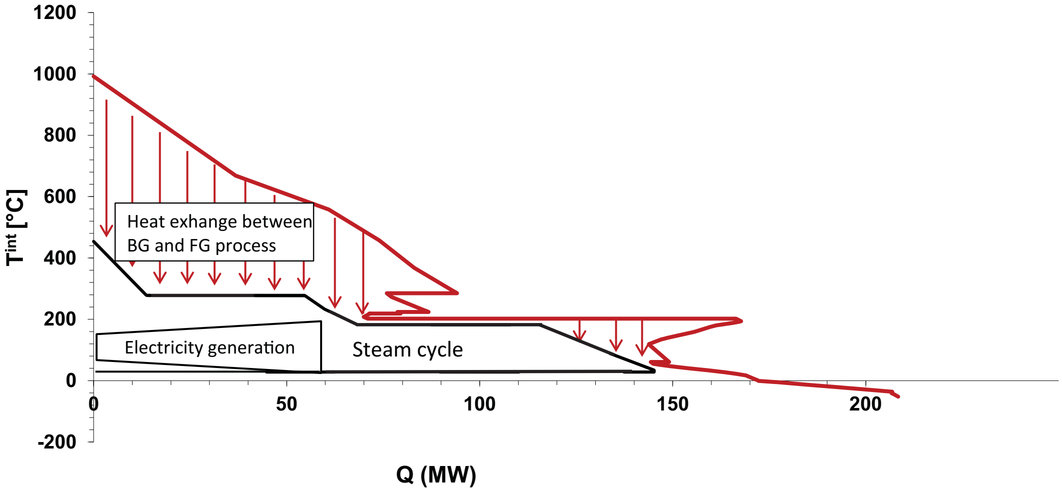

Figure 14. An example of a BG/FG GCC, representing the FT syncrude process (BG) and a steam power cycle (FG).

In all of the papers (**II-V**) a pinch targeting analysis rather than a complete retrofit design has been conducted. This is because the aim of this thesis is to present the economic and environmental (here: GHG emissions) potential for integration opportunities of the studied technologies in a refinery rather than calculating the exact costs.

7

Economic input data

This chapter presents the input data for the economic evaluation, including investment costs and energy prices. This chapter also presents the similarities and dissimilarities between the economic input data in the concerned papers.

7.1 Investment cost data

The technical and economic data for the studied technologies has been based on information and data from the literature. Table 11 presents the cost data for the technologies included in this thesis and in the appended papers (**III** and **V**). The investment costs for the BG-to-FT fuel plant and the CO_2 capture plant are described separately.

Table 11. Summary of the investment costs for the different technologies studied in this thesis.

Equipment	Cost year	Equipment cost	Scaling Unit	Factor [21]	Source
Heat exchanger	2007	$24{,}000+46*A^{1.2}$ [$]	m^2	3.5	(Sinnott and Towel, 2009)
	2002	$3.28*10^4*(A/80)^{0.68}$[$]	m^2	3.5	(Peters et al., 2003)
Heat pump	2010		MW_{th}	2	Based on vendor information[a]
NGCC	2007	$1.82*10^6*P_{el}^{0.79}$ [$]	MW_{el}	2	(Gas Turbine World, 2007)
NB	2007	$2719.5*S^{0.763}$ [$]	kg/h	2	(Matche, 2011; Milligan, 2011)
BB	2007	$1.27*10^6*Q^{0.72}$ [€]	MW_{th}	Incl.	(Pihl et al., 2010)
Steam turbine	2007	$1.9*10^6*W^{0.72}$ [€]	W_{el}	Incl.	(Axelsson, 2008)
Piping	2002	See Fig. 12.4 in (Peters et al., 2003) [€/m][b]	m	2.5	(Peters et al., 2003)

[a]The cost for equipment was subdivided into the cost for the evaporator, compressor and condenser (equal parts) and scaled using vendor information.

[b] The scaling coefficients for Stainless steel (316) and carbon steel have been used.

Several studies can be found that investigate the techno-economic performance of Fischer-Tropsch from biomass systems (Ekbom et al., 2008; Hamelick and Faaij, 2002; Hamelinck et al., 2004; Kreutz et al., 2008; Larson et al., 2009; Ng and Sadhukhan, 2011; Swanson et al., 2010; Tijmensen et al., 2002; Trippe et al., 2011; van Vliet et al., 2009).

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[21] To include direct costs, see Section 6.4.

Figure 15 shows the investment cost for several studies with a BG-to-FT fuel production system similar to the one in this thesis. The cost data was transformed into year 2010 monetary value and represented as investment costs (M€) versus biomass input (kg dry substance/s).

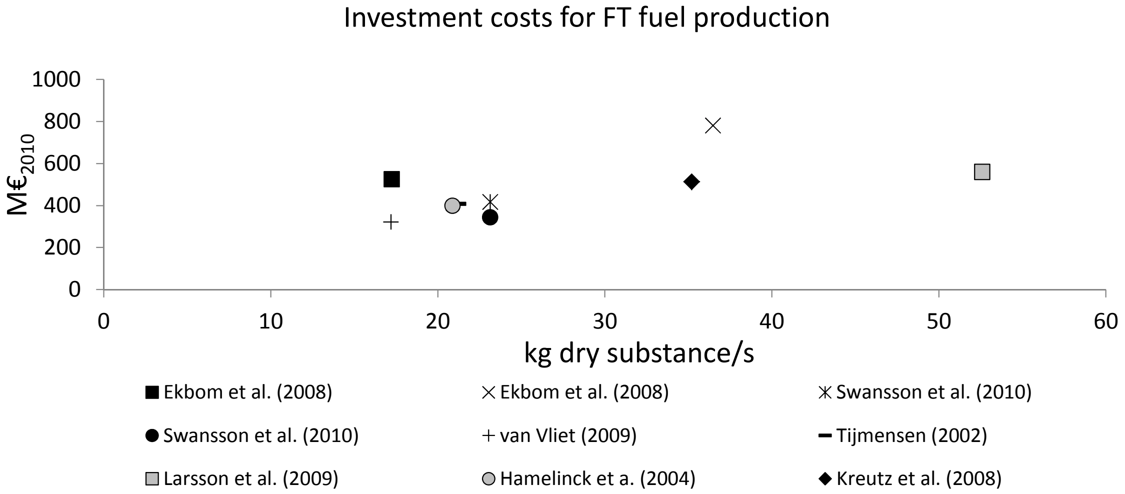

Figure 15. Investment costs for FT fuel production, with a fluidized bed gasifier, reported in the literature.

As can be seen in Figure 15, the investment costs for an FT plant vary significantly between different sources, which depends on several factors such as process layout and equipment choice, production phase (first-of-its-kind process or a “nth” plant), and assumptions pertaining to direct and indirect costs. This makes the costs not directly comparable. The costs for the BG-to-FT syncrude process have mainly been taken from detailed cost data reported by (Kreutz et al., 2008) and the investment costs have been calculated by adding the indirect and direct costs to the equipment cost, see **Paper V** for more details.

In contrast to the investment costs for the FT fuel system, less detailed cost estimates were found in the literature for the CO_2 capture investment costs. In **Paper III** the cost estimates were based on information from the case refineries. In **Paper V** the cost data for the CO_2 capture plant has been estimated by the curve fit of investment cost data from a number of studies on MEA post-combustion CO_2 capture, see Figure 16. All cost data is reported without energy plant investment. The curve fit for the data gives CO_2 capture investment cost (in million Euros, M€) as:

$$\text{Investment cost for the } CO_2 \text{ capture plant} = 0.0053x^{0.75} \quad (4)$$

for interval 370,000 <x< 2,000,000, where x is tonne CO_2 captured per year.

The curve fit was a way to validate the cost-data used in **Paper III**. The red dot in the figure represents the input data used in **Paper III**. As can be seen in the figure, the data corresponds to the curve-fit very well and it can be concluded that data for the investment costs in **Paper III** and **Paper V** is quite consistent.

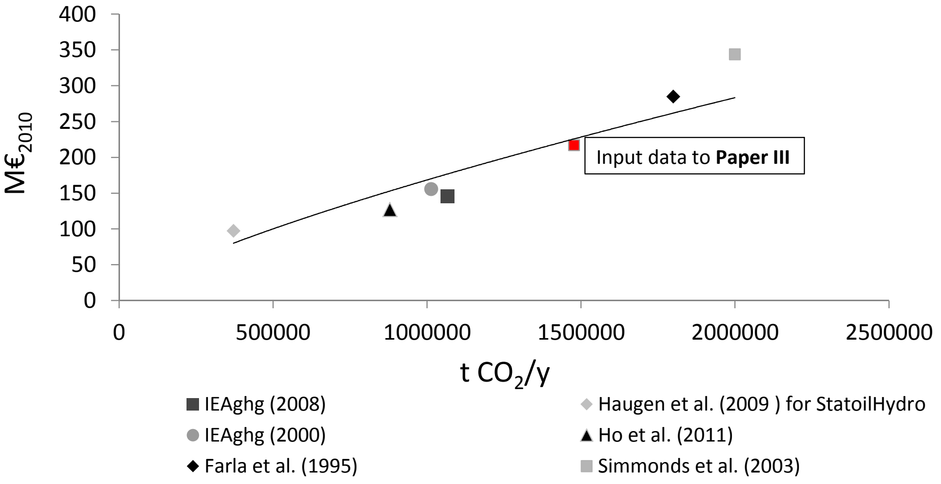

Figure 16. Total investment costs for CO_2 post-combustion plants found in the literature. The red dot represents the input data used in **Paper III.**

7.2 Data for the energy market scenarios

As described in 6.3, the prices and GHG emissions for energy market parameters were generated using the ENPAC tool. Because of a continuous development of the tool, which was conducted in parallel with this thesis work, two different versions of the tool have been used. Despite somewhat different price and emission levels, the general concept of the tool remains. In the following text, a description of the differences between the two versions of the data (presented in Table 12 and Table 13) and a short description of the key assumptions in the tool are presented.

The scenarios have been based on combinations of different levels of fossil fuel prices and CO_2 charges, which are illustrated in Figure 17. The fossil fuel prices represent different developments in the fossil fuel market, and the CO_2 charges represent weak to strong ambitions to decrease CO_2 emissions.

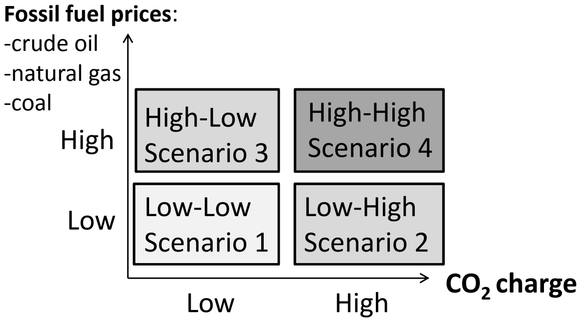

Figure 17. Illustration of input to the ENPAC tool.

The input data for the fossil fuels has been taken from World Energy Outlook 2010 (IEA, 2010b) and represent the current policies scenario and the 450 ppm scenario. The two levels of charges for CO_2 emissions have been based on reports that predict the future (in 2030) charge for CO_2 emissions (EC, 2008; Europelectric, 2009; IEA, 2010b). The high charge for CO_2 emissions represents the highest value from these studies, while the low charge for CO_2 emissions reflects the median value from the same studies. The lowest charge for CO_2 represents a value of 45 €/t CO_2, which could be questionable, see Section 9.6. The level of support for renewable electricity and renewable fuel production are additional inputs to the tool, see Figure 13. The levels of support for renewable electricity and renewable fuel have been set to represent an average value for Europe.

Since neither FT fuel production nor CO_2 capture is a mature technology, but rather a technology that is under development, these technologies will most likely not be implemented before 2020. It was therefore judged appropriate to use energy market conditions for 2030, representing an average year on which to base estimations of cash flows related to the different investment options.

Table 12. Energy market parameters for the different scenarios in **Paper III**. The prices are for year 2030 but in the monetary value of 2010.

Input data to ENPAC				
Fossil fuel price level	Low	Low	High	High
CO_2 charge level	Low	High	Low	High
Fossil fuel price	[a]	[a]	[b]	[b]
- Crude oil [\$/barrel]	92	92	132	132
- Natural gas [\$/Mbtu $_{HHV}$]	11	11	14	14
- Coal [\$/t]	67	67	114	114
CO_2 charge [€/t CO_2]	45	106	45	106
Support for renewable electricity [€/MWh_{el}]	25	25	25	25
Scenario tool output	1	2	3	4
Electricity price [€/MWh_{el}]	66	75	75	87
CO_2 from electricity generation [kg/MWh_{el}]	679	129	679	129
Build marginal technology for electricity production	Coal	Coal w.CCS	Coal	Coal w.CCS
Price of low grade wood fuel [€/MWh_{fuel}]	35	56	40	61
Alternative user of wood fuel	Coal power plants			
Natural gas price[c] [€/MWh_{fuel}]	33	33	41	41

[a] From the 450 ppm scenario in (IEA, 2010b).

[b] From the current policies scenario in (IEA, 2010b).

[c] Price on the European market including transit and distribution costs.

Table 13. Energy market parameters for the different scenarios in **Paper V**. The prices are for year 2030 but in the monetary value of 2010

Input data to ENPAC				
Fossil fuel price level	Low	Low	High	High
CO_2 charge level	Low	High	Low	High
Fossil fuel price	[a]	[a]	[b]	[b]
- Crude oil [\$/barrel]	92	92	132	132
- Natural gas $[\$/\text{Mbtu}_{\text{HHV}}]$	11	11	14	14
- Coal [\$/t]	67	67	114	114
CO_2 charge [€/t CO_2]	45	106	45	106
Support for renewable electricity [€/MWh$_{el}$]	20	20	20	20
Support for renewable fuel (diesel) [€/MWh$_{fuel}$]	26	26	26	26
Support for renewable fuel (gasoline) [€/MWh$_{fuel}$]	35	35	35	35
Scenario tool output	1	2	3	4
Electricity price [€/MWh-el]]	69	77	79	89
GHG from electricity generation [kg $CO_{2\text{-eq}}$ /MWh$_{el}$]	259	259	805	259
Build marginal technology for electricity production	Coal w. CCS	Coal w.CCS	Coal	Coal w. CCS
Price of low grade wood fuel [€/MWh$_{fuel}$]	27	49	38	53
Alternative user of wood fuel	FT CCS	Coal	FT CCS	Coal
Natural gas price[c] [€/MWh$_{fuel}$]	37	37	45	45
FT Diesel gate price [€/MWh$_{fuel}$]	69	86	90	107
FT Diesel gate price [€/l$_{fuel}$]	0.89	1.11	1.17	1.39
FT gasoline gate price [€/MWh$_{fuel}$]	62	79	81	98
FT gasoline gate price [€/l$_{fuel}$]	0.73	0.93	0.95	1.15

[a] From the 450 ppm scenario in (IEA, 2010b).

[b] From the current policies scenario in (IEA, 2010b).

[c] Price on the European market including transit and distribution costs. The difference from the figures in table above was caused by the update of the tool.

The latter version of the tool, used in **Paper V**, includes GHG emissions other than CO_2 (CO_{2eq}). In the latter version of the tool a support for renewable diesel and gasoline was included, see Table 13. No support for renewable fuels was needed in **Paper III** and was therefore not included.

The electricity price has been assumed to be equal to the total cost of electricity generation for a new base-load plant, and the technology with the lowest cost was assumed to constitute the base load build margin technology in each scenario. As can be seen in Table 12 (**Paper III**), in scenarios with a low charge for CO_2, the build margin is coal power, while in scenarios with a high charge for CO_2, coal power with CCS has the lowest production cost and, thus, is the build margin technology of choice. In Table 13 (**Paper V**) the build margin technology is coal power with CCS in all scenarios except in the scenario with high fossil fuel prices combined with a low charge for CO_2. In both versions of the tool, the electricity generation cost for a coal power plant and a coal power plant with CCS are very close in the scenario with a low charge for CO_2 and a low price for fossil fuels. Therefore when the investment costs was updated in the latter version of the tool; the build margin technology was changed. This does not affect the price on the future energy market, but will affect the GHG emissions connected to the marginal technology. The tables show how the electricity price, which is dependent on both CO_2 charge and coal price, increases from Scenario Low/Low to Scenario High/High.

It is assumed that the high-volume user of wood fuel with the highest willingness to pay is the marginal price-setting user for wood fuel. The price of wood fuel has been set based on the assumption of a high-volume user with the highest willingness to pay. As can be seen in Table 12 (**Paper III**), the alternative use of wood was co-firing in coal power plants in all scenarios. Between **Paper III** and **Paper V**, the ENPAC tool was developed to also include FT fuel production as an alternative use of biomass, which is the main reason for the difference in alternative biomass users between **Paper III** and **Paper V**. In the scenarios with a low charge for CO_2 in **Paper V (**Table 13**)** FT facilities are estimated to have the highest willingness to pay for wood fuel, while in scenarios with a high charge for CO_2 , coal power plants will have the highest willingness to pay for wood fuel. The ENPAC tool presupposes a harmonized European bioenergy market that has been developed due to increasing requirements on the share of renewables in the EU. In 2011 the price of wood fuel in Sweden was around 20 €/MWh. In the ENPAC tool the price of wood fuel is higher in all scenarios, which is due to the fact that a future with a subsidy for renewable electricity generation in coal power plants, subsidies for renewable motor fuels and a higher CO_2 charge increases the willingness to pay for wood fuel. The price of biomass is higher in the scenarios used in **Paper III** than in the scenarios used in **Paper V**. The reason is that a higher support for renewable electricity was used in **Paper III**.

For FT diesel and gasoline the gate price has been set so that the end user will have the same price per unit of fuel energy for FT diesel or gasoline as for fossil-based diesel and gasoline. Energy tax and VAT are assumed to be the same for FT diesel and gasoline as for fossil-based diesel and gasoline, but fossil fuels are subject to the charge for CO_2 emissions. Distributions costs are assumed to be the same for renewable diesel and gasoline as for fossil-based diesel and gasoline.

8

Results

This chapter summarises the main results of the thesis.

This chapter presents the results connected to the research questions described in Section 1.1. The answers to some of the research questions are a combination of results from several of the appended papers. For a more thorough description and discussion of the results the reader is referred to each specific paper.

Short summary to facilitate the reading of the results:

In this thesis biomass is considered a limited resource. Under this condition the use of biomass in a refinery decreases the amount of biomass available for other applications. By assuming an alternative user of biomass, the impact on GHG emissions under these conditions can be quantified. In ***Paper II****, this impact is illustrated by adding the emissions from the use of biomass. In* ***Paper IV****, however, the impact is illustrated by comparing the emission reduction with the emission reduction for the alternative user of biomass. All results concerning GHG emission reduction in relation to BG are here illustrated as in* ***Paper IV*** *(i.e. all results in Paper* ***II*** *are recalculated). Different alternative uses of biomass are considered in the appended papers, including co-firing biomass with coal in coal power plants, stand-alone DME and FT fuel production. It should be stated that the reduction potential for co-firing biomass with coal in coal power plants does not take into account the practical aspects of co-firing biomass with coal, see also Chapter 9.*

Moreover, as discussed in Section 6.3, a future build margin approach is used for electricity generation. In ***Paper II****, two future build margins for power generation have been used; coal condensing power and coal condensing power with CCS. In* ***Paper IV*** *an additional possible build margin is included; an NGCC. In this chapter the results from* ***Paper II*** *are updated with an additional marginal technology, the NGCC, to be able to compare the results with the results of* ***Paper IV****. For* ***Papers III*** *and* ***V****, the marginal electricity is generated by the scenario tool used in these studies, see 7.2.*

*Finally, for the papers that include cost calculations (****Paper III and V****), the results are shown for four scenarios. To understand these results, it is necessary to understand the difference in the scenarios, which are discussed in Section 7.2.*

8.1 CO_2 mitigation potential for the European oil refining industry

1. *What is the potential for on-site CO_2 mitigation for the European oil refining industry and where are refineries located that have good access to adjacent infrastructures?*

In **Paper I** the prospect for future CO_2 abatement in European oil refining industry has been assessed in relation to the associated infrastructure. Figure 18 shows the 58 most CO_2 emitting refineries (exceeding 1 Mt CO_2/y) and their current adjacent infrastructure. (For the infrastructure included in this thesis see Section 5.1.) The analysis gave that most refineries are located close to at least one energy infrastructural system that could facilitate CO_2 mitigation. This represents the reliability of creating connections with the potential to facilitate larger reductions in CO_2 emissions, on-site or globally. For example, almost all refineries in the EU (+ Norway) have a nearby natural gas grid.

CCS technology is currently under development and there is no appropriate infrastructure either for CO_2 transportation or CO_2 storage. As concerns the predictions for the implementation of CCS technology at refineries, the refineries would most likely benefit if they could coordinate CO_2 transportation with other large CO_2 point sources, such as the power industry. In Figure 18, the areas highlighted in grey represent regions with several large point CO_2 sources that have been assumed to be areas favourable for CCS. The finding shows that only 14 of the 58 refineries with the highest CO_2 emissions are located in these areas. Possible CO_2 storage sites are also shown on the map.

Regarding the possibilities for energy efficiency measures, refineries could benefit from coordinating energy flows and process streams with other industries. Clusters of industries could also be attractive for emerging biorefineries, with a focus on large-scale conversion of biomass to high-grade materials and fuels. Most of the refineries in Europe are located close to other chemical industries. The largest chemical clusters, including refineries, are located in the Netherlands, Belgium, France, Germany, and the UK.

As to the prospect for excess heat utilisation, district heat delivery is dependent on access to available district heating networks. Refineries with potential access to available district heating systems are, obviously, associated with countries that have developed district heating markets, including Sweden, Finland, and Lithuania, as well as countries with high annual district heating growth rates, such as Norway and Austria.

In summary, refineries located along the North Sea coastline and in the west of Germany have the most advantageous locations with respect to adjacent infrastructure. Complex refineries are expected to have the largest potential to withstand changes in fuel demand. These refineries are spread all over Europe and only three out of eighteen complex refineries are located in areas with favourable conditions for the clustering of CO_2 emissions. In this thesis the areas that represent capture clusters include emissions from large point sources exceeding 20 Mt CO_2 annually. However, capture clusters with less CO_2 emissions annually (e.g. 10 Mt CO_2) may also facilitate the transportation of CO_2. If that is a future case, more refineries (from all categories) will be located in such areas. As can be seen in Figure 18 there are also refineries that are located close to a region with a high density of CO_2 sources, which perhaps also could benefit from coordinated CO_2 transportation with large CO_2 point sources.

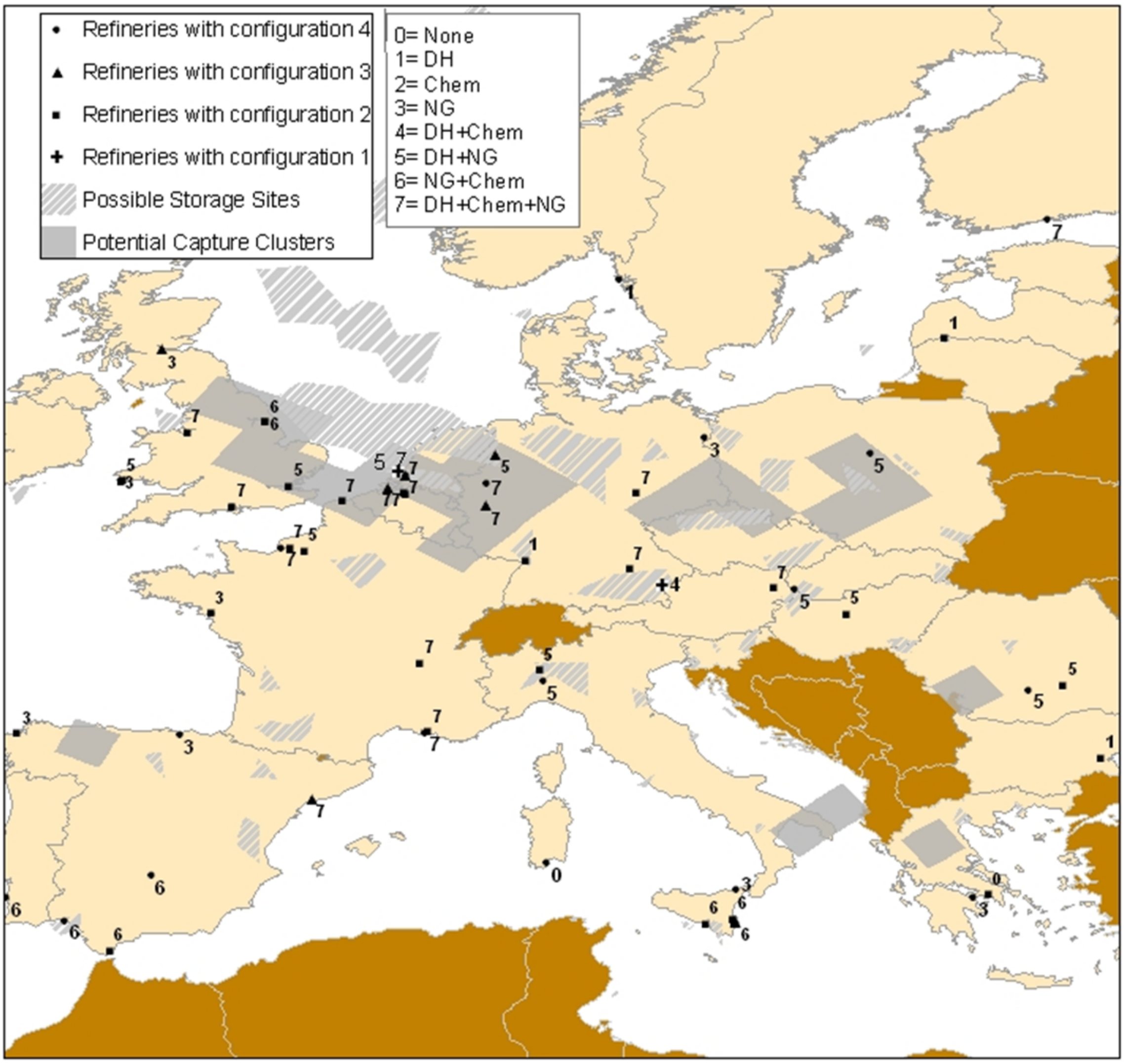

Figure 18. Geographical distribution of refineries with CO_2 emissions >1 Mt/year in relation to district heating systems (DH), chemical clusters (CC), and natural gas grids (NG). The numbering indicates the combination of different adjacent infrastructures. Possible CO_2 storage sites are represented by grey lines. Potential capture clusters, regions where emissions from large stationary point sources (including emissions from power plants, iron and steel industries, cements plants and pulp and paper plants) exceed 20 $MtCO_2$ annually, are highlighted in grey. *This map includes the intellectual property of the European National Mapping and Cadastral agencies and is licensed on behalf of these agencies by EuroGeographics.*

By combining the above described analysis of adjacent infrastructure with that of the current status of the refinery industry and review of the potential for various mitigation options, the potential for CO_2 mitigation was assessed in **Paper I**, see Figure 19. The results from the detailed analysis above were scaled to include all refineries in EU27 and Norway. The mitigation options included were: three different levels for energy efficiency improvements, fuel substitution (with biomass or natural gas), CCS at hydrogen plants, CCS at FCC units, CCS with oxyfuel combustion and CCS with post-combustion capture from combined stacks. For the CCS option, the four different alternatives were further limited to only include those refineries that were located in CO_2 capture clusters. The reader is referred to **Paper I** for a more thorough discussion of the underlying assumptions for the different mitigation options.

The results show that from a short-term perspective, the possible abatement options are energy efficiency improvements by 5-10% and fuel substitution of liquid fuel for natural gas[22]. For these alternatives the overall abatement potential was shown to be relatively low. From a long term perspective, introducing biomass for direct use in boilers and more advanced energy efficiency improvements could possibly increase the potential for mitigation. The introduction of CCS has the potential to achieve more substantial reductions in the refining sector, considering the conditions used in **Paper I**. It should be noted that **Paper I** considers CO_2 emissions from a refinery perspective (i.e. on-site emissions), which for example does not include the introduction of renewable fuel production. The estimated potential for CO_2 capture varied depending on the capture technology and the target source for CO_2 capture. The post-combustion process may have the largest potential for the capture of large amounts of CO_2. In **Paper I** it was also found that if CCS is only implemented at refineries located in CO_2 clusters, the abatement potential will be significantly lower. Moreover, it is not feasible to simply add all the CO_2 mitigation possibilities shown in Figure 19, since each individual measure often has an impact on the potential for other measures.

The potential for reduction in on-site CO_2 emissions for EU refineries was in **Paper I** compared to potential increases in CO_2 emissions reported by (CONCAWE, 2008). The possible increase in CO_2 due to changes in quality and demand has been shown to be in the range of 36-86 Mt CO_2/y for EU refineries (CONCAWE, 2008), which should be compared to the current level of 155 Mt CO_2/y. This highlights a possible future dilemma for the oil refining industry.

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[22] The emission factor for liquid fuel is approx. 0.075 kg CO_2/GJ, while methane has an emission factor of 0.055 kg CO_2/GJ, thus, replacing the liquid fuel with natural gas results in less CO_2 emissions from combustion.

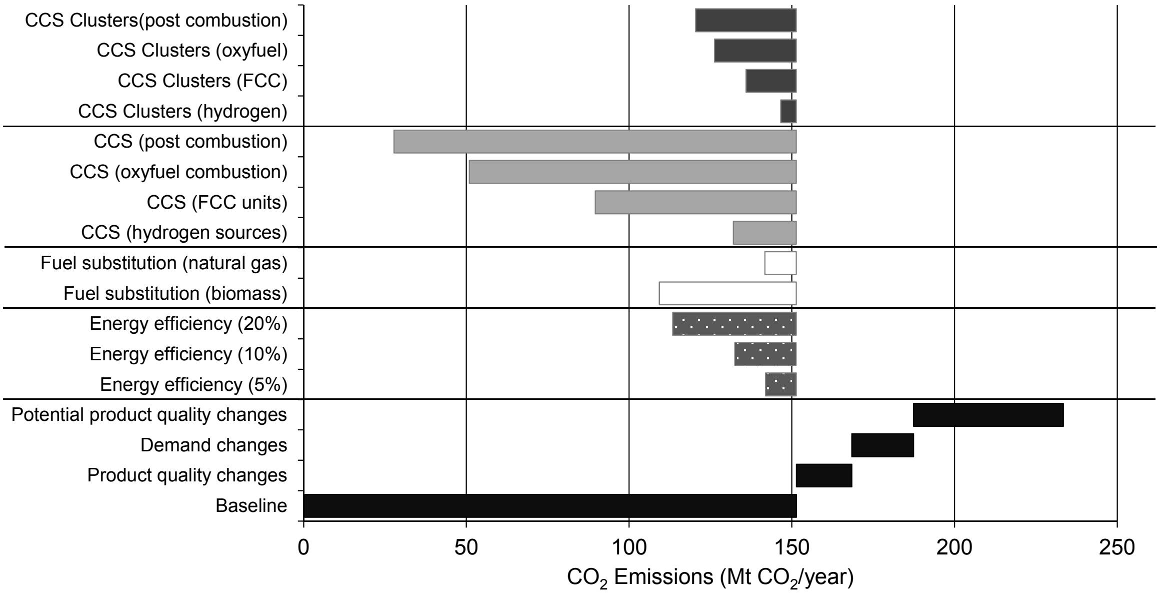

Figure 19. Potential impacts on the aggregated refinery CO_2 emissions of the assessed abatement strategies, and changes in fuel demand and fuel specifications. The dark and light grey, white and dotted bars indicate potential for reductions with the different mitigation options. The black bars, to the right, indicate potential increases in CO_2 emissions (a consequence of demand and product changes). The baseline represents the current CO_2 emissions from the European refining industry (average CO_2 emissions in 2007–2009). All CO_2 mitigation measures are scaled to represent the current EU refining industry.

To summarise the answers to research question one "What is the potential for on-site CO_2 mitigation for the European oil refining industry and where are refineries located that have good access to adjacent infrastructures?":

- *From the mitigation options studied in* ***Paper I****, it was found that the EU oil refining industry has the potential to reduce its CO_2 emissions considerably. However, it was also found that the potential for the currently available mitigation measures in the oil refining industry is relatively limited.*

- *The potential for CO_2 capture varies significantly depending on capture technology and the targeted CO_2 source. Furthermore, the potential for CO_2 capture at EU refineries significantly decreases with the assumption that CCS will be limited to those areas with large CO_2 emission sources.*

- *Refineries located along the North Sea have potentially good access to infrastructures, such as district heating networks, natural gas grids, chemical industries, and possible CCS storage sites that could facilitate CO_2 mitigation measures.*

8.2 Effect of large-scale biomass gasification at a refinery on global GHG emissions

2. *How would the implementation of large-scale biomass gasification (BG) technology with hydrogen or FT fuel production at an oil refinery affect global GHG emissions?*

Three of the appended papers include an evaluation of the potential for GHG emission reduction of large-scale BG integration at an oil refinery. Of those, **Paper II** evaluated different BG technologies for the production of hydrogen, while **Papers IV** and **V** evaluated the integration potential for FT fuel production at a refinery with an integrated and a stand-alone alternative.

The aim of **Paper II** was to show the effect on CO_2 emissions of different BG technologies[23] for hydrogen production, and to compare these alternatives with the conventional production of hydrogen in an SMR. In **Paper II**, the system boundaries were set around the refinery, excluding biomass harvesting and the transportation of biomass and natural gas. For this reason and in order to be comparable to the results of **Papers IV** and **V**, the results of **Paper II** are in this section recalculated to include these parameters. The results are also recalculated for GHG emissions. Since the BG systems studied in this thesis are of different sizes and applied in different types of refineries and reference systems, the resulting reduction in GHG emissions is not directly comparable. Therefore, all results in this section are presented per unit of biomass input.

The results in **Paper II** showed that implementing a BG-to-H_2 process at a hydroskimming refinery could reduce global GHG emissions compared to a reference refinery with an SMR, as illustrated in Figure 20. The specific GHG emissions for two plausible alternatives for the use of biomass, co-firing of biomass with coal in coal power plant and DME production, are shown to the right in Figure 20. As can be seen in the figure, the BG-to-H_2 alternatives always have a lower potential for GHG emission reduction than co-firing of biomass with coal in coal power plants. However, all BG-to-H_2 alternatives (except EF with torrefied biomass) have a greater potential for GHG emission reduction than DME production. From a climate point-of-view and with the conditions stated in **Paper II**, biomass is more efficiently used for co-firing in coal power plants than for hydrogen production in a refinery.

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[23] FT(pyrolysis oil), EF (torrefied biomass (BM)), CFB and DB

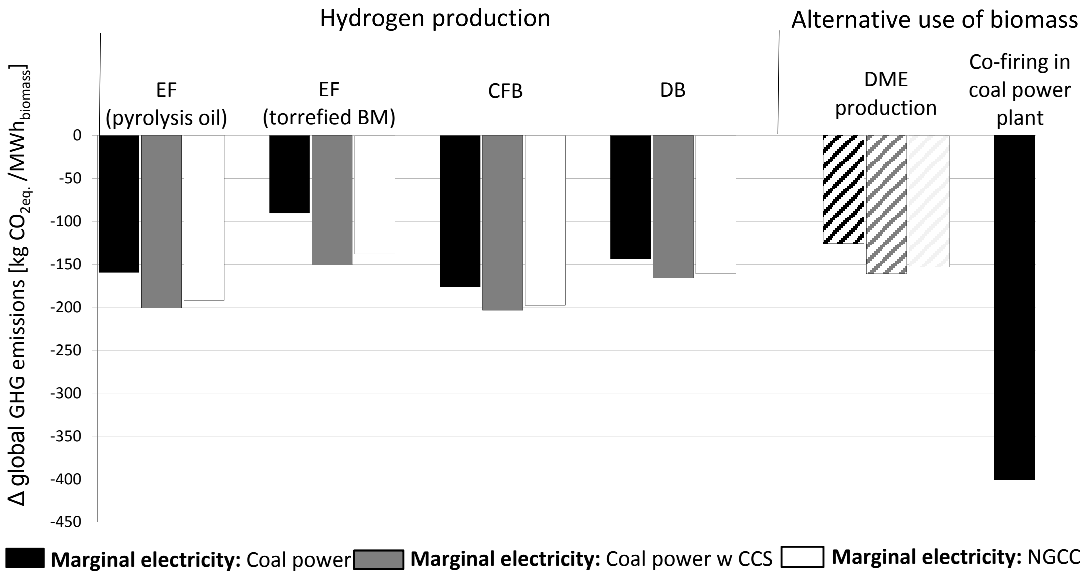


Figure 20. The effect of different BG concepts on global GHG emissions, in comparison to the reference case; an SMR. A comparison of co-firing the same amount of biomass with coal in coal power plants with DME production is also shown on the right in the figure. The abbreviation BM= biomass.

From Figure 20 it can be seen that, from the perspective of an unlimited amount of biomass for all uses, the EF gasification process using pyrolysis oil or the CFB gasification process would be the alternative for hydrogen production that yields the largest reduction in GHG emissions per biomass energy input. The reason the EF (pyrolysis oil) case has greater potential for a reduction in GHG emissions is due to the fact that this system generates extra steam and gas (pyrolysis gas) that replace natural gas at the studied refinery. However, the EF (pyrolysis oil) case has a larger demand for biomass than the other BG cases, but this is compensated by the additional generation of steam. The other alternative that generates greater reductions in GHG emissions than DB and EF (torrefied BM) is the CFB process. This process is the most resource efficient of the studied processes and it is therefore also one of the processes with the greatest potential for specific GHG reduction. The EF process using torrefied biomass has the least potential for GHG emissions reduction, which is due to a loss in efficiency because of the necessary pre-treatment. All gasification processes require a net electricity import. Therefore, in all gasification processes, the marginal electricity generation associated with the lowest GHG emissions (coal power with CCS) gives the greatest potential for GHG reduction. For a more thorough description of the results, the reader is referred to the appended **Paper II**.

The CFB gasifier was the selected gasifier for FT fuel production in **Papers IV** and **V**. The results of **Paper IV** were later applied in **Paper V**. The aim of **Paper IV** was to evaluate the impact on GHG emissions associated with FT fuel production via BG. The analysis was conducted for an FT syncrude process that was integrated with a refinery and the results were compared to an analogous stand-alone case. The reader is referred back to Figure 9 for the energy flows. The results presented in the thesis illustrate the case where heat from the FT syncrude process at high temperatures (>400°C) was utilised for heat integration with the refining processes and that heat at 200°C was utilised for electricity generation in a steam cycle. The other case represented in **Paper IV** was a setup where the heat from the FT syncrude process at 200°C was utilised for heat integration with the refining processes via steam generation. However, when the results from **Paper IV** were applied in **Paper V**, the case with a combination of heat integration and electricity generation was judged to be the most general case, see also **Paper V** and **Paper IV**. The results are also illustrated for the alternative where the FT syncrude is upgraded in new units. For all the cases and the results of **Paper IV**, see the appended paper.

The results in **Paper IV** showed that an FT syncrude production that is heat integrated (HI) with refining processes has greater potential for GHG emission reduction than the stand-alone (SA) alternative in two of the scenarios, when the marginal electricity generation is coal power with CCS and NGCC. The difference is because natural gas could be saved when integrating the FT syncrude process with the studied refinery. The stand-alone case generated more electricity (45 MW compared to 19 MW in the HI case). However, this does not affect the global GHG emissions as much as the amount of natural gas that could be saved in the HI case in the scenarios when the marginal electricity is generated by coal power plants with CCS or an NGCC. The result is here illustrated for the alternatives where FT syncrude is upgraded in new refinery units and for the HI case where electricity is generated. As is illustrated in Figure 21, the resulting GHG emission effects for the studied alternatives are compared to alternative use of the same amount of biomass for co-firing with coal in coal power plants. The figure shows that the production of FT fuels will not offset the same potential for GHG reduction as co-firing the same amount of biomass in a coal-power plant. Therefore, from an environmental point-of-view the biomass is, in this case, more efficiently used for power generation in a coal power plant than for FT fuel production. As for the results in **Paper II**, the greatest reduction in GHG emissions was obtained when the marginal electricity generation was a coal power plant with CCS. The reason is, again, that this technology is associated with the lowest GHG emissions. From the figure it is clear that the HI alternative is more sensitive to variation in marginal electricity technology than the stand-alone case, which is due to the larger net import of electricity. The results also show that capturing the relatively clean CO_2 stream leaving the gasification process significantly improves the potential for reduction in GHG emissions.

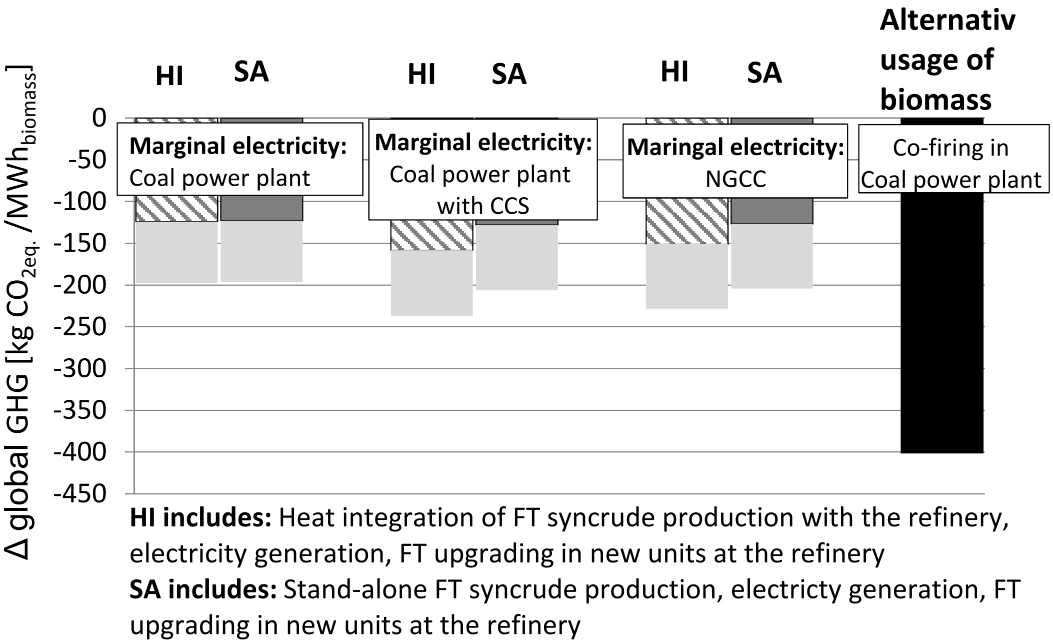


Figure 21. GHG emission reduction for an integrated FT production and a stand-alone FT production. The grey bars represent the increase in GHG emission reduction if CO_2 capture is considered in the BG process. A comparison with co-firing the same amount of biomass with coal in coal power plants is also shown (black bar).

The focus in this thesis has been on integrating FT fuel production with a refinery in order to compare the potential for reduction in GHG emissions with stand-alone FT fuel production. However, the FT syncrude process could be heat integrated with other industries that have a deficit of heat at suitable temperatures. Therefore, in **Paper IV**, the results were also compared to the results from a study by Isaksson et al. (2012), who, among other BG concepts, have evaluated the CO_2 effect of the integration of an FT syncrude process in a pulp and paper mill. As can be seen in Figure 22, the results from Isaksson et al. (2012) show a greater CO_2 reduction per extra biomass input than the results for a refinery. For the refinery, the extra biomass input is the same as the biomass required for the FT syncrude process, which is due to the fact that no biomass is currently used for heating at the refinery. In the pulp mill, however, the extra biomass is less than the amount required for the FT syncrude process, since part of the biomass demand is taken from the biomass saved due to the integration with the FT plant, see Isaksson et al. (2012).

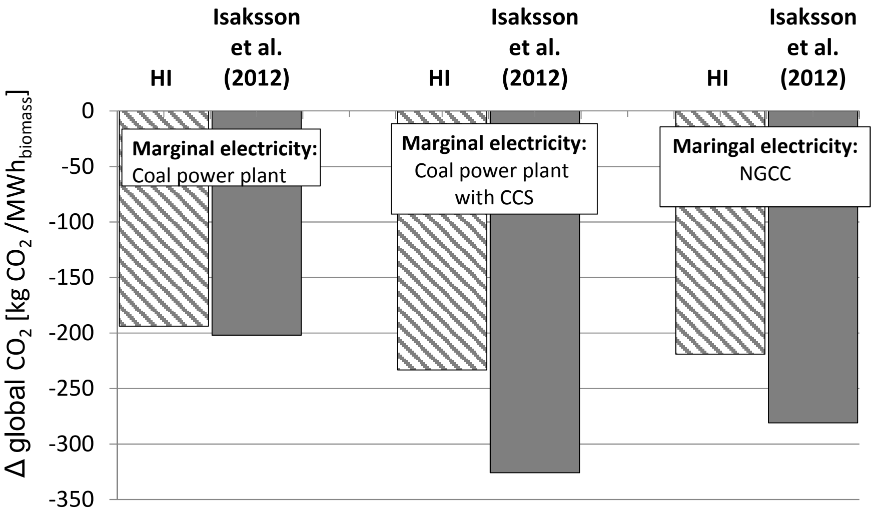


Figure 22. Comparison of CO_2 emission reduction when integrating FT syncrude production in a refinery and a pulp mill.

Capturing the CO_2 stream leaving the gasifier was considered a reasonable option for the BG processes in **Papers IV** and **V**. This option was not considered in **Paper II.** For that reason the results in **Paper II** have been recalculated to include the possibility for CO_2 capture of the CO_2 stream leaving the BG processes. Figure 23 summarizes the studied BG concepts in this thesis for a comparison of the potential for GHG emission per unit of biomass. The two plausible alternative uses for biomass, DME production and co-firing in coal power plants, are presented as a comparison to the levels of GHG emission reduction obtained for the studied BG alternatives.

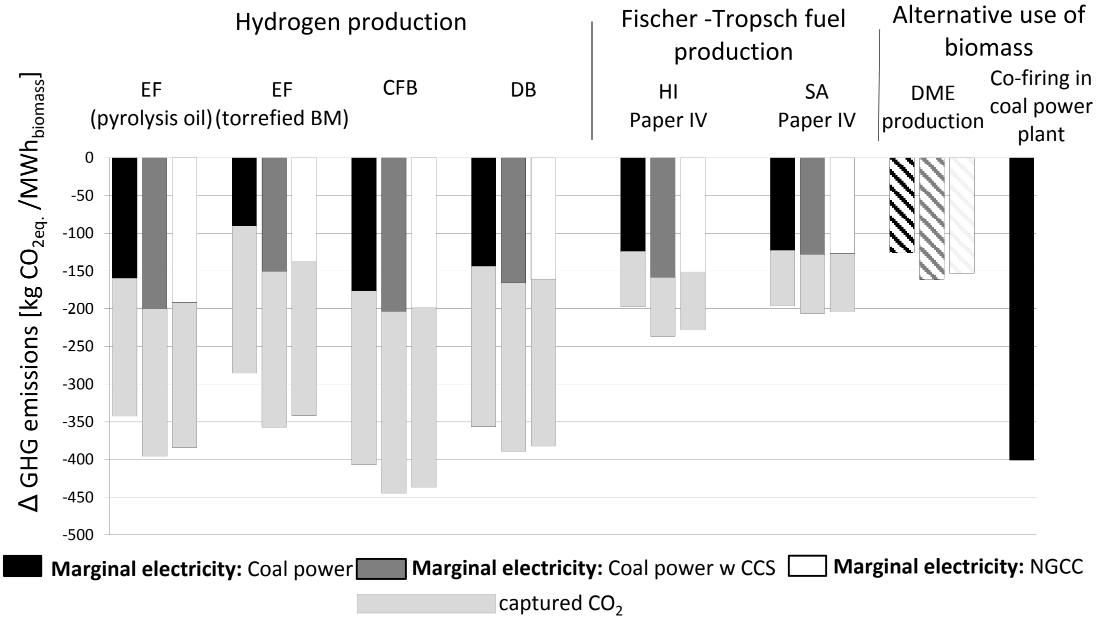


Figure 23. GHG emission per unit of biomass for the studied BG options for three alternatives of plausible build margin electricity generation. The light grey bars represent the increase in GHG emission reductions if CO_2 capture is considered in the BG processes. The GHG emissions corresponding to alternative use of biomass are also included in the figure, representing DME production and co-firing in coal power plants.

In the figure it can be seen that the hydrogen production via BG generates a greater reduction in emissions than the FT cases. This is due to the fact that the results shown here for the BG-to-H_2 cases are compared to a reference system consisting of a refinery and an SMR. In addition to the reduction in natural gas due to steam generation in the BG process, these cases benefit from a major reduction in natural gas feedstock to the SMR. In addition, the net electricity demand is larger in the BG-to-FT cases than in the BG-to-H_2 cases.

Moreover, compared to the use of biomass in coal power plants and in terms of GHG emission reduction, only the hydrogen production cases can compete, presupposing that the CO_2 stream from the gasification is captured. The ability to capture CO_2 from the BG cases leads to a greater reduction in GHG emissions per unit of biomass for the BG-to-H_2 cases than in the BG-to-FT cases. The reason for this is mainly due to the fact that a BG-to-H_2 process generates larger CO_2 streams. In addition, the produced hydrogen replaces hydrogen from natural gas and thus saves additional GHG emissions. Moreover, the BG-to-H_2 process generates larger flows of CO_2 than the reference case refinery with an SMR, which is due to the composition of the feedstock. Reductions in GHG emissions are also obtained in the integration between the gasification and the refinery, which is found in both hydrogen and FT production.

To summarise the answers to research question two:" How would the implementation of large-scale biomass gasification (BG) technology with hydrogen or FT fuel production at an oil refinery affect global GHG emissions?

- *Implementation of a large-scale BG technology at an oil refinery could reduce global GHG emissions. However, the reduction in GHG emissions is, for all studied cases without the possibility to capture the CO_2 stream from the gasification, lower than co-firing the same amount of biomass with coal in a coal power plant.*

- *When considering the effect on GHG emissions of integrating a biomass gasification process for either hydrogen or FT fuel production at a refinery, capturing the CO_2 stream leaving the gasification process significantly increases the reduction in GHG emissions.*

- *An FT fuel production that is heat integrated with a refinery generates significantly larger GHG emission savings than corresponding stand-alone alternative in the scenarios when the marginal electricity is coal power with CCS or an NGCC.*

- *With CO_2 capture available at the gasification process, FT fuel production at a refinery results in greater potential for a reduction in GHG emissions per unit of biomass than stand-alone FT fuel production and DME production. Yet, the potential for reduction is lower than co-firing in coal power plants.*

- *With CO_2 capture available at the gasification process, BG-to-H_2 production with a CFB gasifier in a refinery results in greater potential for a reduction in GHG emissions than both the use of biomass for DME production or co-firing with coal in coal power plants. The reason for this is the large flow of CO_2 that is separated in the BG-to-H_2 process, and the substitution of natural gas feedstock.*

- *The results also show that the potential for GHG mitigation of integrating an FT plant in a refinery compared to an alternative where a similar process is integrated in a pulp mill depends on the origin of the marginal electricity. The GHG effect is shown to be significantly greater in a pulp mill when the marginal electricity is coal power with CCS and an NGCC.*

- *Consequently, from a climate point-of-view and with the conditions stated in this thesis, biomass is shown to be more efficiently used for co-firing in coal power plants than for biomass gasification in a refinery for all the studied biomass concepts with the exception of hydrogen production with a CFB gasifier connected to CCS.*

8.3 Heat supply alternatives for post-combustion CO_2 capture at a refinery

3. *What is the most cost-effective and robust heat supply alternative for a post-combustion CO_2 capture plant?*

Research question three was addressed in **Paper III**. The question was answered by evaluating the avoidance cost for post-combustion CO_2 capture at two case refineries. The focus in **Paper III** was on different heat supply options to cover the heat requirements in the capture plant. Four different heat supply options were considered (see also Table 10):

- Steam produced by excess heat from the refinery process (directly or via a heat pump). (EH+HP)
- Steam produced by an NGCC with electricity generation. (NGCC)
- Steam produced by an NB with electricity generation. (NB)
- Steam produced by a BB with electricity generation. (BB)

The profitability of investing in a post-combustion CO_2 capture unit was evaluated as the cost for avoided CO_2 emissions on-site at the case refineries. The avoidance cost was then compared to the price of CO_2 emissions allowances in order to assess whether or not the alternative is cost-effective. An avoidance cost much lower than the charge for CO_2 emissions indicates potential profitability for investing in a CO_2 capture unit. The final cost must incorporate additional costs, such as the costs for transportation and storage, which are not included in **Paper III**. However, previous studies have estimated the price of transportation and storage to be in the range of 15- 25 €/t CO_2 captured (Campbell, 2008; CCS skagerrak - Kattegat, 2011).

Paper III was the only paper that included both case refineries at the same time. Figure 24 presents the resulting avoidance cost for the studied case refineries. The avoidance cost was evaluated for two different levels of heat demand in the capture process, as well as under four different future energy market conditions (see Section 7.2). In order to cover uncertainties about the demand for heat of the post-combustion CO_2 capture process that still exists, this thesis evaluates two levels of heat demand (2800 kJ/kg CO_2 and 4700 kJ/kg CO_2). For each of the scenarios the charge for CO_2 emission is marked as a straight line.

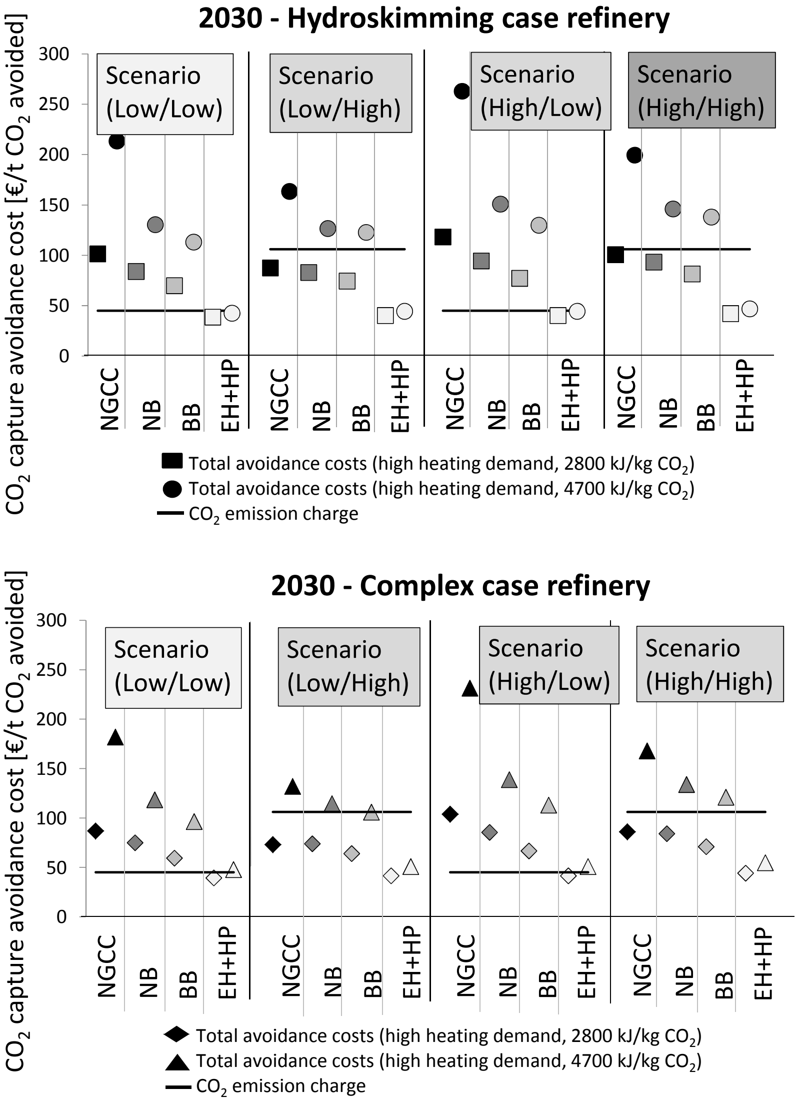


Figure 24 Capture avoidance costs for the two case refineries. In scenarios with a low charge for CO_2 the marginal technology for electricity is coal power, while in scenarios with a high CO_2 charge the marginal technology coal power with CCS.

The most robust heat supply option is defined as the option least sensitive to changes in energy requirements in the CO_2 capture plant and energy market parameters; such as for example fuel and electricity prices. The figure shows that the EH+HP alternative is robust to such changes. This alternative indicates an avoidance cost of around 50 €/t CO_2 in all scenarios. Assuming an additional cost of 25 €/t CO_2 (CCS skagerrak - Kattegat, 2011) for CO_2 transportation and storage, post-combustion CO_2 capture using excess heat could be profitable for scenarios characterised by a high charge for CO_2 emissions. The cost for the whole CO_2 capture chain would add up to around 75 €/t CO_2, which in the scenarios with a high charge for CO_2 emissions creates a significant margin to the CO_2 charge level at 106 €/t CO_2. Moreover, for scenarios with low fossil fuel prices and a high charge for CO_2 emissions, several other alternatives could be promising, pre-supposing that the CO_2 capture process has low heating requirements. However, several of these options will add up to an avoidance cost close to or above the CO_2 charge level of 106 €/t CO_2 when the whole capture chain is considered.

For all options, the avoidance cost increases when the energy requirement increases. The difference is significant for all heat supply options, except for the EH+HP. This can be explained by the fact that the excess heat from the refinery is not associated with any fuel costs, except electricity to the heat pump. The flue gases produced while generating heat for the regeneration of the absorbent are also captured, which results in larger absolute energy demands and higher investment costs for the NGCC, the NB and the BB cases. Besides this, the avoided amount of on-site fossil CO_2 emissions will decrease for the NGCC and the NB alternatives, which is due to the increase in CO_2 emissions that are emitted during heat generation (i.e. the efficiency in the capture plant was assumed to be 85%).

The reason for studying two different types of refineries was to determine whether or not there were differences in the available amount of excess heat that could influence the avoidance cost for the EH + HP alternative. However, the results showed that the amount of excess heat, with the aid of a heat pump, was sufficient to cover the heat demand in the CO_2 capture process in both refineries. The part that is covered with the aid of a heat pump, however, differs between the case refineries and the different energy requirements. For the hydroskimming refinery, excess heat above 130°C is enough to cover the entire demand for heat in the case with a low energy demand, while a high demand for energy requires that 15% of the energy demand is covered with the aid of a heat pump. For the complex refinery, in the case with a low energy demand, 40% of the energy demand must be covered with the aid of a heat pump, while with a large demand for energy, 60% of the demand for heat needs to be covered with the aid of a heat pump. From the results it is clear that the size of the heat pump and the increase in the demand for electricity affect the avoidance cost. However, this is only marginal compared to the effect of a higher demand for heat of the other heat supply options. Furthermore, the results show slightly lower avoidance costs for the complex case refinery than for the hydroskimming refinery. This can be explained by lower specific investment costs related to higher levels of emitted CO_2 emissions.

The main focus in **Paper III** was a refinery perspective on the cost and robustness of the implementation of a CO_2 capture plant. The reason for this is that EU refineries are part of the EU ETS, and the refineries are currently allocated CO_2 allowances for on-site CO_2 emissions. However, electricity is both generated and consumed in the CO_2 capture plant and the net electricity generation/usage will affect global CO_2 emissions. Figure 25 shows the change in avoidance cost if the perspective is expanded to include the changes in CO_2 emissions outside the refinery gate, see Section 6.2.2 for the methodology approach. If the CO_2 avoidance cost is viewed from an expanded system approach this cost will be reduced for all heat supply alternatives except in the EH+HP case. The electricity demand of the EH+HP alternative is the net import of electricity, which results in lower levels of avoided CO_2 emissions when the system is expanded to include the CO_2 emissions associated with power generation. The largest change in the CO_2 avoidance cost is related to the option with an NGCC that supplies the required heat. The reason for this is that the NGCC generates large amounts of electricity that decrease marginal electricity production and, consequently, decrease global CO_2 emissions. However, even with a slightly higher avoidance cost for the EH+HP alternative, this is still the most cost-effective option for most scenarios.

The avoidance cost from a global perspective show the importance to have policy instruments that include all global emissions and not only the emissions that are emitted on-site at the industry. Estimating CO_2 reduction from a "global perspective", can be seen as input to legislative bodies when considering political instruments to reduce CO_2 emissions from the process industry.

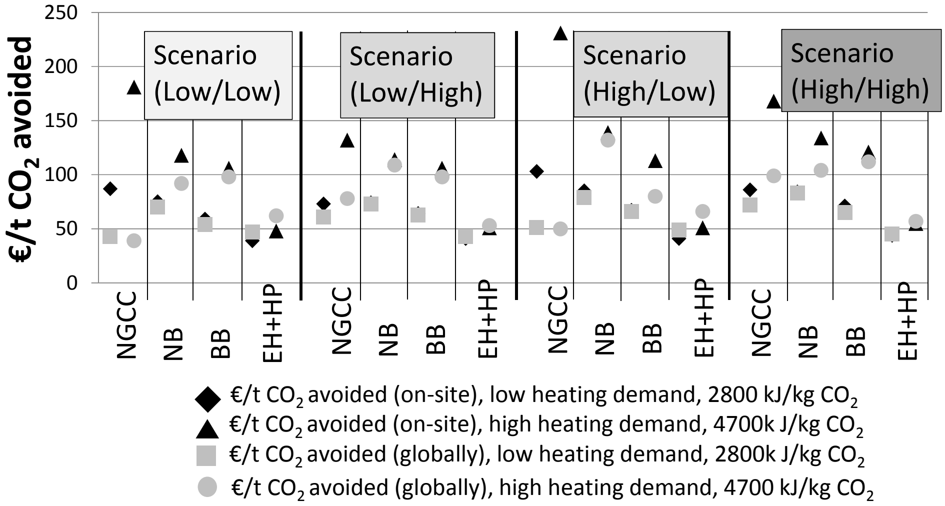


Figure 25. Comparison between CO_2 avoidance cost from a refinery perspective and from an expanded system approach. The comparison is illustrated for the complex refinery only. In scenarios with a low charge for CO_2 the marginal technology for electricity is coal power, while in scenarios with a high CO_2 charge the marginal technology is coal power with CCS.

When considering the avoidance cost for post-combustion capture with heat supplied by natural gas CHP, the findings in this thesis show an avoidance cost (without the cost for transportation and storage) in the range of 70 to 151 €/t CO_2, while the literature reports a cost in the range of 90 to 120 €/t CO_2 avoided from combined stacks, see Table 4. It should be noted that the costs estimated in other studies arise from different assumptions pertaining to the demand for heat for the capture process, and energy prices, and consequently it is difficult to compare the results from different studies. When the avoidance cost is evaluated from an expanded system approach the NGCC alternative is, in most scenarios and for both a low and a high energy requirement, a competitive alternative for supplying the necessary heat for the regeneration of the MEA, which is similar to what was reported by Hektor (2008), who concluded that the NGCC alternative shows lowest avoidance costs for CO_2 capture in a pulp mill.

To summarise the answers to research question three: "What is the most cost-effective and robust heat supply alternative for a post-combustion CO_2 capture plant?"

- *Using excess heat for supplying the necessary heat to the post-combustion CO_2 capture process is shown to be the most robust alternative, considering the prerequisite for sufficient amounts of excess heat (with the aid of a heat pump at reasonable temperature levels) are available at the refinery.*

- *CO_2 capture is only profitable in scenarios with a high charge for CO_2. In this case, the only alternative that shows possible profitability for both a high and a low level of energy requirement is the option of using excess heat.*

- *If the CO_2 avoidance cost is calculated to include the emission changes outside the refinery, the avoidance cost can be significantly decreased for heat supply alternatives that generate a large amount of electricity. Of the alternatives that generate electricity, the NGCC shows the lowest avoidance cost for both a low and a high energy demand. This cost can be seen as input to legislative bodies when comparing different CO_2 mitigation options.*

8.4 The global CO_2 effect of post-combustion CO_2 capture at a refinery

4. *What is the effect of post-combustion CO_2 capture on CO_2 emissions when the perspective is changed from a refinery perspective to a global perspective?*

In Paper III the global CO_2 effect of post-combustion CCS at a refinery was evaluated from two different perspectives. Figure 26 shows the resulting potential for CO_2 mitigation for CO_2 capture at the two studied case refineries. The results are shown both for on-site CO_2 emissions only and the potential for global CO_2 mitigation, i.e. including CO_2 outside the refinery. The mitigation potential is greater for a complex refinery than a hydroskimming refinery, which is due to the fact that the complex refinery emits more CO_2. From the figure it is clear that the impact of system expansion is significant in a future with electricity generation corresponding to high CO_2 emissions (Coal power plant), particularly for the NGCC case. In those scenarios, the CO_2 capture plant with an NGCC entails the greatest reduction in global CO_2 emissions, which is due to the high net surplus of electricity that replaces electricity generated by coal power. The effect is, however, less in a future with electricity generated with low CO_2 emissions (Coal power plant with CCS). The captured CO_2 emissions from the refinery process are, however, the same in each scenario.

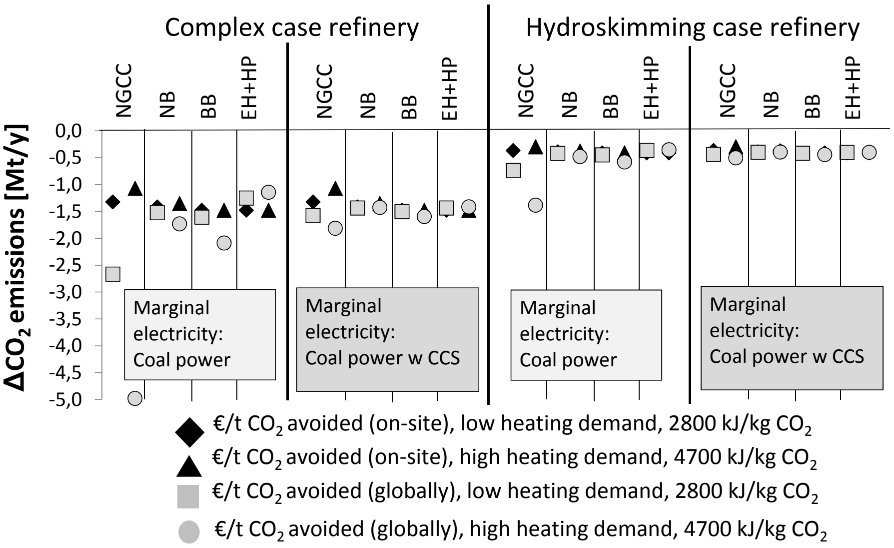


Figure 26. The impact on global CO_2 emissions of the different case refineries and two different marginal electricity technologies. The emissions are calculated both from a refinery-perspective and a global perspective. In scenarios with a low charge for CO_2 the marginal technology for electricity is coal power, while in scenarios with a high charge for CO_2 the marginal technology is coal power with CCS.

In **Paper III** biomass is considered to be available for all purposes. Here, an additional analysis of the results from **Paper III** has been performed, applying the approach where biomass is considered to be a limited resource. The coal power plant is regarded as the alternative user and the results show that the CO_2 reduction significantly decreases for the BB alternative with limited biomass, see Figure 27. For a high heating value, the BB alternative even entails an increase in CO_2 emissions. The results are illustrated for the complex case refinery. However, the effect is the same for the hydroskimming case refinery.

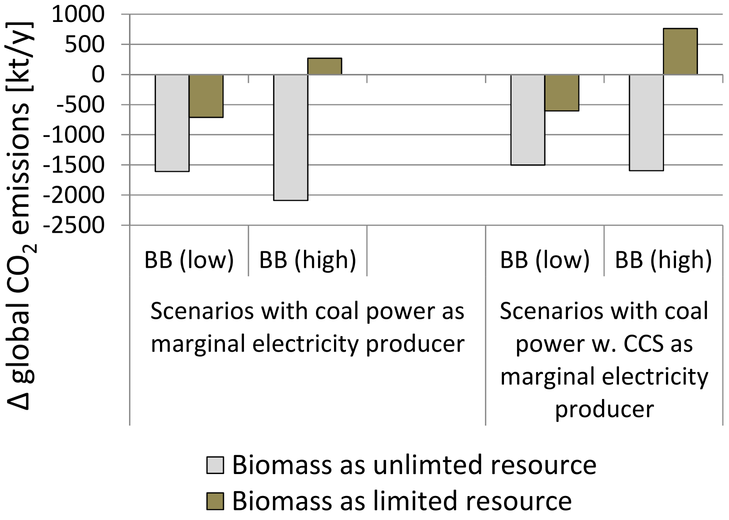


Figure 27. The effect on global CO_2 emissions of the BB case when biomass is viewed as an unlimited and a limited resource.

To summarise the answers to research question four: "What is the effect of post-combustion CO_2 capture on CO emissions when the perspective is changed from a refinery perspective to a global perspective?"

- *If an expanded systems perspective is used that takes into account the changes in the surrounding energy systems, and for scenarios characterized by a coal power plant as marginal electricity generation, the case that utilizes excess heat (with the aid of a heat pump) has less potential for CO_2 emission mitigation than the heat supply options that generate electricity.*

- *If biomass is considered a limited resource, the reduction in CO_2 emission for the BB alternative is significantly less.*

8.5 Comparison of biomass gasification for Fischer-Tropsch fuel production and post-combustion CO_2 capture at a refinery:Economic evaluation and GHG emissions

5. *Under what conditions is it more advantages to invest in a CO_2 capture system (post-combustion technology) than in a large-scale biomass gasification for Fischer-Tropsch fuel production at a complex oil refinery?*

The question of investing in an FT fuel production or a post-combustion CO_2 capture was addressed in **Paper V** and, thereby, unifies this thesis. Both development routs are large strategic investments, which are not likely to be implemented before 2020. For this reason it is particularly interesting to conduct this comparison at a complex refinery which is the type of refinery most likely to withstand change in the transportation sector.

The results discussed in Section 8.3 (for *research question 3*) show that CO_2 capture could be an attractive option under certain conditions; presuming that heat is supplied by excess heat (with the aid of a heat pump), a high level of CO_2 charge and available infrastructure for CCS. The results discussed in Section 8.2 (*for research question 2*) show that FT fuel is more efficiently produced in the heat integrated case than in the stand-alone alternative. However, the previous discussion was limited to GHG emissions and no economic comparison was conducted. Due to this fact and to the uncertainties regarding practical aspects of integrating a BG-to-FT fuel process with a refinery, both the heat integrated case and the stand-alone case are, in this chapter, included in the comparison with the post-combustion CO_2 capture development route. Figure 28 shows the global GHG emissions and the net annual profit for the studied alternatives compared to the reference refinery; see Section 5.2.3. The studied reference refinery is presented by the origo in the figure. The cases that are positioned on the right side of the y-axis result in a higher net annual profit than the reference refinery. These cases could be interesting for the refinery from an economic point of view. Cases positioned on the left side of the y-axis give a lower net annual profit than the reference refinery. These cases are thus not profitable for the refinery. However, these cases show a greater reduction in GHG emissions and could therefore be interesting from an environmental point-of-view.

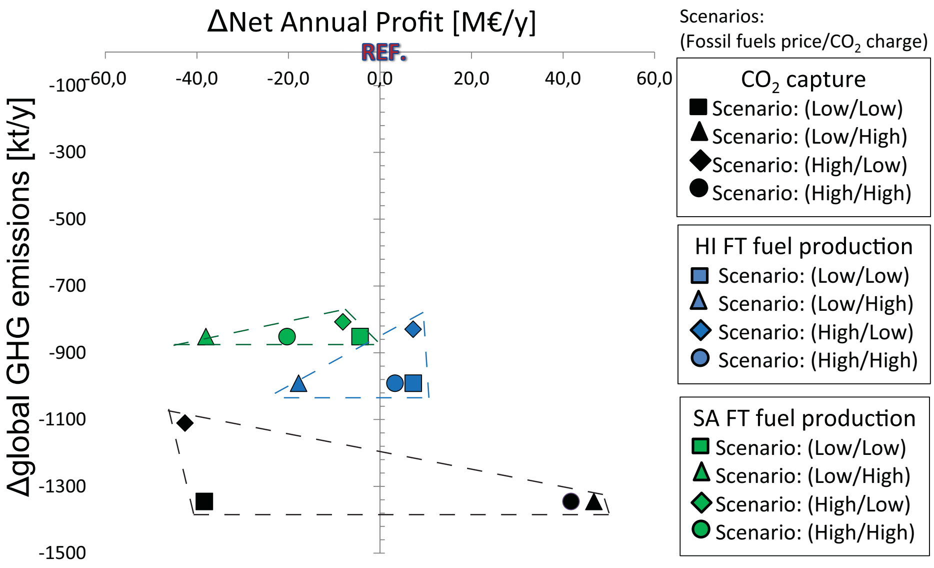


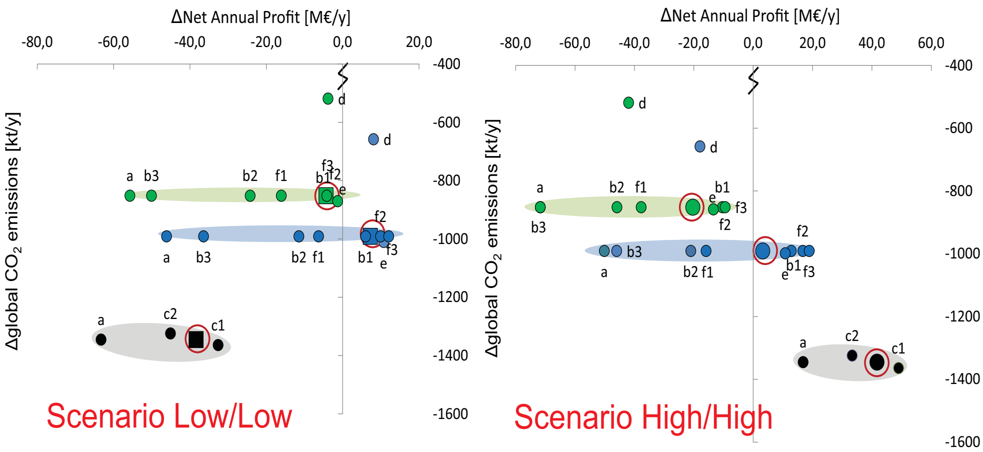
Figure 28. The net annual profit and global GHG emissions for the post-combustion CO_2 capture case, the integrated FT case and the stand-alone FT case, compared to the reference case refinery. For comparison, the total CO_2 emissions from the refinery are approx. 1 800 kt/y.

From the results it is clear that all the studied cases, as expected and discussed in Sections 8.2 and 8.4, result in global reduction in GHG emissions. The post-combustion CO_2 capture case shows, not surprisingly, the greatest global reduction in GHG emissions. The large reduction is solely due to the assumption that all the CO_2 emissions from the major chimneys are captured. The larger reduction in GHG emissions in the integrated FT case than in the stand-alone case is because natural gas could be saved when integrating the FT syncrude process with the refinery, which was discussed in 8.2. The marginal technology for electricity generation is a coal power plant with CCS in all scenarios except in the scenario when the price of fossil fuels is high and the charge for CO_2 is low (Scenario (High/Low)). For that reason the potential for GHG reduction is the same for these scenarios in each of the studied cases. For the Scenario (High/Low) the marginal electricity generation is coal power without CCS. The largest reduction in GHG emissions is associated with the generation of electricity with lowest GHG emissions (Coal power with CCS), as was discussed in Section 8.2.

The CO_2 charge varies from 45 €/t CO_2 to 106 €/t CO_2. In the scenarios where a high CO_2 charge is assumed (Scenario (Low/High) and Scenario (High/High)), post-combustion CO_2 capture is the alternative with the greatest profitability. For these scenarios it should be noted that the charge for CO_2 is very high (106 €/t CO_2), which of course could be questionable. CO_2 emission allowances are the single revenue in the post-combustion CO_2 capture case, which makes it very sensitive to the cost of emitting CO_2. For the CO_2 capture process to be profitable, the charge of emitting CO_2 must exceed the CO_2 avoidance cost. A rough interpolation shows that the CO_2 avoidance cost must at least surpass a value of around 75 €/t CO_2.

Heat integrated FT fuel production (HI) shows profitability in three of the studied scenarios. The profitability is highest in the scenarios with a lower price for wood fuel (in scenarios with a low charge for CO_2). The wood fuel price is very high in scenarios where a high charge for CO_2 has been assumed, which is a consequence of increasing requirements on the share of renewables. However, in the scenario where both high fossil fuel prices and a high charge for CO_2 have been assumed, the high price for wood fuel is compensated by a high selling price for the FT fuels. The stand-alone FT case (SA) does not show profitability in any of the studied scenarios. The explanation is that the large amount of natural gas that can be saved in the heat integrated FT case generates more revenues than the extra electricity that is generated in the stand-alone case.

In the ENPAC tool that was used to generate the energy prices, some parameters are defined by the user, e.g. the support level for the generation of renewable electricity and for renewable fuels. In Figure 29 the results of a sensitivity analysis of the level of subsidy along with a few other parameters that are judged to have significant influence on the results are presented for each scenario. These are: the capital recovery factor, the possibility to capture CO_2 from the BG, the reboiler duty in the capture process as well as upgrading FT syncrude in new units. The studied parameters and the changes are described below the figure. The results are presented for two of the scenarios, (Low/Low and High/High), the results from the remaining scenarios can be found in **Paper V**. For comparison, the results in Figure 28 are in Figure 29 marked with red circles.



[a] Represents a change in capital factor from 0.1 to 0.2

[b] Represents changes in the level of biofuel support (b1=+50%, b2=-50%, b3=no support)

[c] Represents changes in the level of heat demand for the reboiler (c1=2.8 MJ/kg CO_2, c2=4.7 MJ/kg CO_2)

[d] Represents a change in the FT cases where no CO_2 is captured

[e] Represents a change in FT upgrading from existing to new units

[f] Represents changes in the level of renewable electricity support (f1=+50%, f2=-50% , f3=no support)

Figure 29. Result of an additional analysis of parameters that were assumed to have a significant effect on the results. The green symbols represent the FT stand-alone case, the blue symbols represent the heat integrated FT case and the black symbols represent the post-combustion CO_2 capture case. The original results from Figure 28 are represented by a square in Scenario Low/Low and a circle in Scenario High/High.

In Figure 29 it can be seen that the net annual profit significantly decreases when the CFR is changed from 0.1 to 0.2, which can be expected since all studied cases have high investment costs.

The different management control measures (subsidies for renewable fuels and renewable electricity) economically affect FT fuel production in different directions. In both cases, a change in the level of subsidy will have an effect on the price of wood fuel, since that will affect the willingness for a high-volume user to pay for wood fuel, as discussed in 7.2. An increase in the levels of subsidies (b1 and f1) results in a higher wood fuel price. However, in the case with an increase in the level of subsidies for renewable fuel production, the net effect is an increase in the revenues for the refinery (especially when the prices for both fossil fuels and CO_2 is high), while an increase in the level of subsidies for renewable electricity leads to a net decrease in the revenues for the refinery. If the level of support for renewable electricity is decreased (f2 and f3), the price for wood fuel will decrease, which results in an increase in the annual profit for the FT cases. In general, the subsidy for renewable fuel production is a large share of the income from FT fuel production and thus, a drop in level leads to a drop in revenues.

From the sensitivity analysis it can be concluded that support for renewable fuels is necessary for the FT cases to be profitable. The level of support, however, varies depending on the different scenarios. The integrated FT case will be profitable with a support for FT fuels that corresponds to an average value of the current European level, but the revenues go from a positive value to a negative value if the level of support is decreased by 50% (for all scenarios except Scenario (High/Low), see **Paper V**). None of the FT cases can be profitable without support for renewable fuels. For the stand-alone case, the level of support needs to be increased by more than 50% compared to the current level before this case can become economically attractive. The level of support for renewable fuels represents an average value for European countries, but there are countries that already have a higher level of support, which could possibly make FT fuel production profitable in all scenarios (corresponding to the support level in b1). However, there are also countries that need to promote higher levels of support in order for integrated FT fuel production to show promise for becoming profitable.

A decrease in the reboiler duty (c1) for the post-combustion CO_2 capture plant results in, as can be expected, an increase in both revenues and the reduction of GHG emissions. The greater reduction in emissions is due to the drop in power to the heat pump. If the reboiler duty instead is increased, a larger heat pump will be needed and the reduction in GHG emissions will instead be lower (c2).

A change into upgrading FT syncrude in new units instead of using existing refining units gives a better diesel yield that would increase the revenues for the refinery.

5. *To summarise the answers to research question five:" Under what conditions is it more advantages to invest in a CO_2 capture system (post-combustion technology) than in a large-scale biomass gasification for Fischer-Tropsch fuel production at a complex oil refinery?":*

- *A high charge for CO_2 economically favours CO_2 capture at a refinery, while a low charge for CO_2 economically favours FT fuel production.*
- *Economic policy support for renewable fuels is essential for FT fuel production to be profitable.*

- *A high* CO_2 *charge is essential for* CO_2 *capture to be profitable. The results show that the* CO_2 *charge must exceed 75 €/t* CO_2*.*
- *The heat integrated FT fuel production is most profitable in scenarios with a low price for wood fuel. The stand-alone alternative is not profitable in any of the studied scenarios.*
- *When comparing post-combustion* CO_2 *capture and FT fuel production at a refinery, post-combustion* CO_2 *capture shows the greatest potential for GHG emission mitigation.*
- *Without the option to capture and store the concentrated* CO_2 *stream in the FT process the potential for GHG reduction in the FT cases significantly decreases.*
- *The results show that the economic performance of the studied technologies is highly sensitive to several parameters. The net annual profit significantly decreases when the capital recovery factor is changed, which is due to the high investment costs.*

9 Discussion

This chapter provides a discussion of the results, and includes general reflections on the research conducted in this thesis.

9.1 The need for a functioning infrastructure for CO_2 transportation and storage

One major assumption has been that CO_2 capture will become a likely mitigation option in a mid-term future with an operational transportation system and safe storage sites. Today, the CO_2 capture technology struggles with several issues, especially with respect to public acceptance for onshore CO_2 storage and uncertainties about the legal framework for CO_2 capture, transportation and storage. Collaboration between nations and across disciplines is required to overcome these issues which also include technology uncertainties. Such collaboration has begun and must continue to progress.

9.2 Generality of the results

The development routes for heat integration with refining processes studied in this thesis will increase the complexity of the operation at a refinery. The studied development routes are under development and, thus subject to uncertainties with regard to accessibility. In general, a refinery runs with very high accessibility. Therefore, it must be determined how different operating hours affect integration possibilities and how to control a backup system (e.g. furnaces with spare capacity) that can be turned on and off quickly. Before any potential heat integration can be realised all safety and process technical aspects must be investigated along with the construction of an appropriate back-up system. These issues have been beyond the scope of this thesis but would be interesting for further research.

This thesis examines the potential for CO_2/GHG mitigation and the economic performance of the included development routes based on the background foreground principle in the pinch methodology. For example, it was assumed that all available excess heat could be utilized, both concerning the refining processes, the BG-to-H_2 and the BG-to-FT fuel processes. In reality there may be some constraints that have not been considered here, e.g. space, controllability, practicability of the CO_2 capture facility and BG plants, as well as uncertainties pertaining to the utilization of the syngas from the gasification process at high temperatures.

Although the results are largely based on case refineries, the data has been taken from real operation conditions and provides important indications of the level of excess heat from refineries. In practice, however, all refineries are different with respect to process units, operation, size, product mix and feedstock. This fact makes it more difficult to generalise the results in this thesis. However, according to the Solomon Index, the case refineries are among the European refineries with the lowest CO_2, SO_x and NO_x emissions, which indicate that these are among the most energy-efficient refineries. For this reason it is reasonable to assume that the level of excess heat at the present refinery could be regarded as a minimum level of excess heat at refineries with the same complexity in Europe.

9.3 Assumptions regarding CO_2 capture sources and utilization of excess heat

It is probable that CO_2 capture will be placed at target sources with the highest concentration of CO_2. The cost for the capture technology will decrease since an increase in the concentration of CO_2 will require a smaller post-combustion plant. At a refinery, hydrogen production is subject to a very high CO_2 concentration compared to other sources. The most cost-effective alternative would probably be to capture the CO_2 emissions in the flue gases from hydrogen production directly, before these emissions are mixed with flue gases from other sources. However, there are uncertainties pertaining to operational security at this stage. Since a low risk of disturbance of normal operational conditions is of the highest importance, the safest alternative would be to capture the CO_2 emissions from the chimneys.

Excess heat from the refinery process has been regarded as a heat source for the CO_2 capture process. Heat at temperatures below the CO_2 reboiler temperature is generated in the capture process and could be utilised with the aid of a heat pump. This is not considered in this thesis since there is enough heat that could be utilised with the aid of a heat pump from the refining process. Moreover, it is assumed that the CO_2 plant is located outside the current refining process area. There are several options for arrangements for a post-combustion capture plant; however, most refineries suffer from a shortage of space and for this reason an arrangement of the transportation of flue gases in pipelines to a decentralised capture plant is considered feasible.

9.4 System analysis and availability and reliability of input data

The results are based on system analysis. Performing system studies implies many assumptions regarding the studied technologies, the studied systems and the surrounding systems. The scenario tool and additional sensitivity analysis have been used to account for uncertainties in the future energy market. Several alternative users of biomass and marginal electricity generation have been used to account for uncertainties about changes in the surrounding energy system. Data concerning equipment and investment costs has been found in the literature. To account for different assumptions in literature data, a comparison between several sources has been conducted both for the input data and for the final output. For all cost calculations an Nth production plant has been assumed, despite the fact that the studied technologies are all under development and not yet commercially available. These technologies need to be demonstrated in large-scale before commercialisation. Consequently, the estimation methods applied for these investments generally leads to uncertainties about final costs. The influence on investment cost has been studied by changing the capital recovery factor from 0.1 to 0.2. A CRF of 0.1 is viewed as a strategic investment. For the studied development routes it can be concluded that without considering the investments from a strategic point-of-view (with a CRF of 0.1), only the CO_2 capture alternative in scenarios with a high charge for CO_2 emissions would be profitable.

Concerning the CO_2 mitigation analysis on an aggregated level (**Paper I**), the results are based on general technical estimations and there may be contingency with regard to refinery specific potential. Nevertheless, the results show the relative magnitude of the potential for CO_2 reduction. When it comes to exploring the potential for CO_2 mitigation at European refineries and adjacent infrastructures, information has been collected from many different sources, mainly national statistics, company homepages, and different association reports, in order to get as much information as possible. One major issue that appeared during that effort was the difficulty in obtaining consistent data for all refineries and adjacent infrastructures. It was found that the level of public data varies significantly depending on type of data and between countries. Some countries could provide detailed public data, while others keep the same information confidential. To improve energy analysis on an aggregated industrial level the accessibility of data concerning energy infrastructures and industry energy balances must be improved.

9.5 Applicability of the results and relevant comparisons

From a climate point-of-view the use of biomass should be compared to the most reasonable alternative use of biomass with the greatest potential for reduction in CO_2 emissions. Large volume users of biomass are here considered as probable alternatives for a comparison of biomass use. The results in this thesis, and from related research, show that it is, in most cases, preferable from a climate point-of-view to use biomass for co-firing in coal power plants. However, there are more alternatives for renewable electricity generation (e.g. solar, wind etc.) than alternatives for renewable motor production that at the same time are suitable for the current car park. Therefore, the results of FT fuel production in this thesis should be compared to other biomass-based motor fuel production routes in general, and different production routes for FT fuel, in particular.

One key assumption concerning the possibility for CO_2 capture was that the excess heat will be used as a heat supply to the CO_2 capture process. The use of excess heat from the refinery process may be subject to competition between CO_2 capture and different CO_2 mitigation alternatives, such as district heat delivery or drying biomass. Therefore, a thorough analysis of alternative uses of excess heat is needed to determine which strategy is the most beneficial one.

9.6 Using the scenario tool

As stated, the scenario tool (ENPAC) has been developed parallel to the work in this thesis. As a consequence of that, two versions of the tool have been used. However, the same input data has been used. Even if the absolute values differ in the two versions, the structure of the results is the same. In this thesis the lowest CO_2 price was set to 45 €/t CO_2. At this price post-combustion CO_2 capture was shown to be non-profitable. During 2012, the price of CO_2 emission allowances was lower than 10 €/t CO_2. Therefore, at a CO_2 charge of 10 €/t CO_2, the CO_2 capture alternative is not interesting to study. This is the reason why a higher cost for CO_2 is used. However, to study what effect this assumption has on the FT fuel alternatives, the lowest charge for CO_2 emissions was, in the ENPAC tool, set to 10 €/t CO_2. When the price of CO_2 emissions is changed, the price of electricity, the price of wood fuel and the price of renewable fuels are also affected. For the production of FT fuel alternatives, a change in the price of CO_2 allowances does not significantly affect the results. The reason for this is that the lower income from capturing CO_2 emissions from the gasifier is compensated by a lower price for electricity and a lower price for wood fuel. At the same time, the price of the FT fuels would be lower.

Furthermore, co-firing of biomass has been assumed as one alternative user of biomass in this thesis. The ENPAC tool considers an increase in the cost for transportation for biomass compared to coal, based on average transportation costs. However, the ENPAC tool does not consider the difficulties of long distance transportation and the fact that most coal power plants are located in parts of Europe with a low density of biomass and without access to large harbors. Consequently, the emissions from co-firing biomass with coal in coal-power plants should be seen as an optimistic level that might be slightly lower.

10

Conclusions

This thesis presents system analyses of the integration of three development routes in an oil refinery under different future energy market scenarios. The focus has been on biomass gasification to hydrogen and Fischer-Tropsch fuels as well as post-combustion CO_2 capture. The system analysis has been performed with respect to CO_2 or GHG mitigation potential and economic performance. This chapter presents the main conclusions from the research in this thesis and the appended papers, as well as some general conclusions concerning the outcome of this thesis:

- The potential for available short-term on-site CO_2 mitigation options, such as energy efficiency and fuel substitution, is relatively limited. In a long-term perspective, a reduction in on-site CO_2 emissions between 13 to 80% could be achieved by implementing CO_2 capture. The potential for CO_2 capture varies depending on the targeted CO_2 emission point source, the CO_2 capture technology considered and whether CCS is assumed to be limited to areas with large point sources of CO_2 emissions or not.
- It is shown that refineries that are located along the North Sea have nearby infrastructures such as district heating networks, natural gas grids, chemical industries and possible CO_2 clusters and storage sites with the potential to facilitate CO_2 mitigation.
- Implementing biomass gasification for hydrogen production at a refinery could reduce global GHG emissions compared to a reference refinery that produces hydrogen with conventional technology in the range of 85-200 kg CO_2eq. /$MW_{biomass}$. Of the studied alternatives of biomass gasification for hydrogen production, the CFB gasification process shows the greatest potential for GHG mitigation.
- Implementing biomass gasification for Fischer-Tropsch fuel production at a refinery could reduce global GHG emissions in the range of 120-160 kg CO_2eq./$MW_{biomass}$. An integrated Fischer-Tropsch fuel production is preferable over a stand-alone Fischer-Tropsch fuel production, both in terms of economic performance and the potential for GHG mitigation.
- The potential for reductions in global GHG emissions per unit of biomass are greater for hydrogen production at a refinery than Fischer-Tropsch fuel production at a refinery.

- If capturing the CO_2 stream leaving the gasification process in the production of hydrogen and Fischer-tropsch fuels is an option, it can significantly improve the global potential for the reduction in GHG emissions.
- From a climate point-of-view and given the conditions in this thesis, biomass use for co-firing with coal in a coal power plant would lead to greater reductions in GHG emissions than hydrogen or Fischer-Trospsch fuel production at a refinery. The exception is hydrogen production via CFB gasification with CO_2 capture that shows greater potential for reduction in GHG emissions than co-firing biomass with coal in coal power plants.
- The economic performance of the studied development routes depends on future policy instruments and the prices on the future energy market. It is concluded that subsidies for renewable fuels are necessary for Fischer-Tropsch fuel production to be profitable, but the price for subsidies depends on developments on the future energy market. It is further concluded that post-combustion CO_2 capture could be a profitable alternative for a refinery if the charge for emitting CO_2 is high.
- The cost for post-combustion CO_2 capture at a refinery could significantly be reduced if excess heat from the refining process with the aid of a heat pump is used, as compared to post-combustion with other heat supply. It is further concluded that a charge for CO_2 above 75 €/t CO_2 is needed for a CO_2 capture process that utilizes excess heat (with the aid of a heat pump) to become an economically attractive option for an oil refinery.
- Taking into account changes in the surrounding energy systems, if an expanded system is used when evaluating CO_2 capture at a refinery, the potential for CO_2 mitigation increases for a CO_2 capture process that utilizes heat supply options that generate electricity. Given a high charge for CO_2, these systems could be economically attractive from a society perspective. The NGCC alternative is the most interesting alternative for both a low and a high energy demand.
- Future energy prices and policies will strongly affect the investment opportunities of the studied development routes at a refinery. In a comparison between Fischer-Tropsch and post-combustion CO_2 capture, the most profitable option depends on the assumption about the future energy market. Moreover, the development routes are also sensitive to a change in the capital recovery factor, which is due to high investment costs.

11

Future work

In order to better understand the potential for CO_2 mitigation for the European refinery sector, it would be interesting to complement this work with additional case-analyses at similar refineries. The study could at first be limited to refineries with the same types of complexity as the studied refineries in this thesis.

From a refinery perspective, the results in this thesis could form the basis for more detailed investigations of the studied alternatives. It would also be interesting to compare the findings in this thesis with other opportunities for CO_2 or GHG mitigation at a refinery. Examples of alternatives that would be interesting to compare are: district heating delivering, electricity and SNG production from gasified biomass, fuel switch to biomass, electricity generation from gasified petroleum products, renewable diesel production by hydrotreating of bio-oils, as well as other CO_2 capture technologies.

Since this thesis does not consider all practical aspects, it is important to study which constrains that could potentially affect the results in this thesis. It would be interesting to study the accessibility of each process part and how different operation times for different units influence the final mitigation potential and costs. Further, this thesis focuses on maximum heat integration between the CO_2 capture plant and the refining processes. It would be of interest to study how partial integration, which would require a supplementary heat supply from an external heat source, would affect the avoidance cost and the potential for CO_2 mitigation in the alternative for CO_2 capture. It would also be interesting to study the effect on avoidance cost if only the CO_2 sources with the highest CO_2 concentrations were considered.

In this thesis refineries with adjacent district heating networks have been identified. However, neither the potential for utilizing excess heat from the refineries nor the demand for additional district heating has been included. In future research it would be interesting to combine the potential for excess heat delivery from refineries, and other nearby industries, with a future demand for district heating or cooling in order to evaluate the potential for a global reduction in GHG emissions and to compare alternatives for excess heat utilization at refineries.

The global CO_2 effect of integrating Fischer-Tropsch fuel production at a refinery is here compared to a stand-alone alternative and integration with a pulp mill. It would be interesting to broaden this comparison to include the potential for integration with other industries and district heating networks. It would also be interesting to compare at what cost FT fuels can be produced using different alternatives for heat integration.

Abbreviations

BB	Biomass boiler
BM	Biomass
BG	Biomass gasification
CCS	CO_2 capture and storage
CFB	Circulated Fluidized bed
CHP	Combined heat and power
DB	Double bed
EF	Entrained flow gasifier
FCC	Fluidized Catalytic Cracker
FT	Fischer-Tropsch
HI	Heat Integrated
HTFT	High-temperature FT
HP	Heat pump
IGCC	Integrated Gasification Combined Cycle
LTFT	Low-temperature FT
MEA	Monoethanolamine
NB	Natural gas boiler
NGCC	Natural gas combined cycle
SA	Stand-Alone
SMR	Steam Methane Reformer
VGO	Vacuum Gas Oil
WGSMR	Water-gas shift membrane reactor

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