TAMPEREEN TEKNILLINEN YLIOPISTO
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SAMPO KOURI
FEASIBILITY OF CARBON CAPTURE IN KRAFT PULP MILLS

Master of Science thesis

ABSTRACT

SAMPO KOURI: Feasibility of Carbon Capture in Kraft Pulp Mills
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The purpose of this thesis was to assess the feasibility of carbon capture in Finnish Kraft pulp mills. The feasibilities of thirteen different technology options were evaluated based on their technical maturity, capture potential and estimated break-even price for emission allowance including biogenic emissions. Also the role of pulp and paper industry in climate change mitigation was discussed.

The study was conducted by comparing oxy-fuel combustion cases modelled with Balas software and a fuel switch option calculated based on mass and energy balances with results from previous studies. The technical maturity was evaluated based on previous studies and existing commercial activity. The break-even prices from literature were recalculated under the same assumptions for investment costs, commodity costs and supporting policies. Two price estimates were calculated: one including supporting policies and possibly available revenue and another excluding them. The capture potential was estimated using a reference mill scaled to the pulp production rate of 1200 ADt/d. Part of the technologies require large structural changes and were considered available only if the mill is rebuilt. Thus the capture potential in the reference mill was scaled up based on either the capture potential in mills to be rebuilt by 2030 or the capture potential in all Finnish pulp mills. The relevant capture potential was calculated based on the production capacities and the estimated age distribution of the chemical recovery sections in Finnish pulp mills. The results from this thesis were subjected to qualitative and numerical sensitivity analyses.

Carbon capture in pulp mills seems feasible only if other revenues than CCS supporting policies are available. These revenues could include by-products like biofuels, higher energy efficiency or increased pulp production. The cases of lignin separation and black liquor gasification to transportation fuels were most cost efficient, but the capture potentials were limited to 1.45 $Mt(CO_2)/a$ and 0.82 $Mt(CO_2)/a$, respectively. Small scale carbon capture for utilization could be achieved with pre-calcination, as the break-even price was only around 4.5-7.3 $€/t(CO_2)$. Large scale implementation of carbon capture in the Finnish pulp mills with monoethanolamine (MEA) absorption had a capture potential of up to 12 $Mt(CO_2)/a$, but the break-even price was around 70-80 $€/t(CO_2)$ excluding transportation and storage costs. Moreover, barriers in transportation and storage and in the lack of incentives for bio-CCS remain. In addition to CCS, the pulp and paper industry could mitigate the climate change by increasing the amount of carbon based products, sustainable forestry practices and investments in forestation.

TIIVISTELMÄ

SAMPO KOURI: Hiilidioksidin talteenoton sovellettavuus sulfaattisellutehtaisiin
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Avainsanat: bio-CCS, Suomen sellu- ja paperiteollisuus, MEA-absorptio, happi-poltto, happirikastus, vaihtoehtoiset biopolttoaineet, esikalsinointi, kemikaalikierto-poltto, mustalipeän kaasutus, ligniinin erotus

Tämän diplomityön tarkoitus oli arvioida hiilidioksidin talteenoton sovellettavuutta Suomen sellutehtaisiin. Kolmentoista teknologian sovellettavuutta arvioitiin perustuen niiden tekniseen kypsyyteen, talteenottopotentiaaliin ja bioperäisen hiilidioksidin sisältävän päästöoikeuden rajahintaan. Sellu- ja paperiteollisuuden roolia ilmastonmuutoksen hillinnässä käsiteltiin myös.

Tutkimus tehtiin vertailemalla Balas-ohjelmistolla mallinnettuja happipolttotapauksia ja massa- ja energiatasein laskettua polttoaineenvaihtotapausta aiempien tutkimusten tuloksiin. Teknistä kypsyyttä arvioitiin aiempien tutkimusten sekä löytyneen kaupallisen toiminnan perusteella. Päästöoikeuden rajahinnat kirjallisuuslähteistä muokattiin vertailukelpoisiksi käyttäen samoja oletuksia koskien investointikustannuksia, käyttöhyödykkeitä ja poliittisia tukia. Laskettiin kaksi rajahinta-arviota: yksi sisälsi poliittiset tuet ja mahdolliset muut tulot ja toinen ei. Talteenottopotentiaali arvioitiin referenssisellutehtaan avulla, jonka tuotantokapasiteetti oli skaalattu 1200 ADt/d. Eräiden teknologioiden soveltaminen vaatii suuria rakenteellisia muutoksi ja niiden käyttöönoton oletettiin edellyttävän tehtaan uudelleenrakentamista. Siksi talteenottopotentiaali referenssitehtaassa skaalattiin joko niiden tehtaiden potentiaalilla, jotka todennäköisesti rakennettaisiin uudelleen 2030 mennessä tai vaihtoehtoisesti kaikkien Suomen sellutehtaiden potentiaalilla. Kyseiset talteenottopotentiaalit laskettiin Suomen sellutehtaiden tuotantokapasiteettien ja kemikaalikiertojen arvioidun ikäjakauman perusteella. Työn tuloksia tarkasteltiin kvalitatiivisessa ja numeerisessa herkkyysanalyysissä.

Hiilidioksidin talteenotto sellutehtaissa vaikuttaa soveltamiskelpoiselta vain, jos talteenottoa tukevien politiikkojen lisäksi saadaan muita tuloja. Näitä tuloja voivat olla sivutuotteet, kuten biopolttoaineet, energiansäästö tai kasvanut sellutuotanto. Ligniinin erotus ja mustalipeän kaasutus liikennepolttoaineiksi olivat kustannustehokkaimmat teknologiat, mutta niiden talteenottopotentiaalit olivat rajalliset: 1,45 Mt(CO_2)/a ja 0,82 Mt(CO_2)/a. Pieniä määriä hiilidioksidia voitaisiin ottaa hyötykäyttöön esikalsinoinnilla, jonka rajahinta oli vain 4,5–7,3 €/t(CO_2). Laajamittaisella monoetanoliamiiniabsorption soveltamisella Suomen sellutehtaisiin voitaisiin saavuttaa jopa 12 Mt(CO_2)/a talteenotto, mutta rajahinta olisi noin 70–80 €/t(CO_2) ilman kuljetus- ja varastointikustannuksia. Haasteina ovat kuljetus ja varastointi sekä poliittisen tuen puuttuminen bioperäisen hiilidioksidin talteenotolta. Sellu- ja paperiteollisuus voisi hillitä ilmastonmuutosta myös lisäämällä hiilipitoisten tuotteiden määrää, kestävillä metsänhoitokäytännöillä tai investoimalla metsittämiseen.

PREFACE

Tampere, 23.2.2016

Sampo Kouri

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LIST OF SYMBOLS AND ABBREVIATIONS

ADt	Air-dry ton
Air-LK+MEA	Air combustion in the lime kiln with monoethanolamine-absorption
Air-RB+MEA	Air combustion in the recovery boiler with monoethanolamine-absorption
ASU	Air separation unit
BCTMP	Bleached chemi-thermomechanical pulp
BECCS	Biomass energy with carbon capture and storage
BeP	Break-even price for CO_2 emission allowance
Bio-CCS	Biogenic carbon capture and storage
BLG	Black liquor gasification
BLGCC	Black liquor gasification with combined cycle
BLG to DME	Black liquor gasification with motor fuels production
BLS	Black liquor solids
CCS	Carbon capture and storage
CCSP	Carbon capture and storage project
CCU	Carbon capture and utilization
CCUS	Carbon capture, utilization and storage
CLC	Chemical looping combustion
CLC-RB	Chemical looping combustion in the recovery boiler
DARS	Direct alkali regeneration system
EOR	Enhanced oil recovery
EU-ETS	European Union Emissions Trading Scheme
GDP	Gross domestic product
GHG	Greenhouse gas
IEA	International Energy Agency
IEAGHG	IEA Greenhouse Gas R&D Programme
IPCC	Intergovernmental Panel for Climate Change
LK	Lime kiln
LP	Low-pressure
MEA	Monoethanolamine, monoethanolamine-absorption
MeO	Metal oxide
MP	Medium-pressure
Mt	Million tons
MTCI	Manufacturing and Technology Conversion International
MW_{th}	Million watts, thermal
NO_x	Nitrogen oxides, NO and/or NO_2
OE-LK+MEA	Oxy-enrichment in the lime kiln with monoethanolamine-absorption
OE-RB+MEA	Oxy-enrichment in the recovery boiler with monoethanolamine-absorption
OF-LK+SEPA	Oxy-fuel combustion in the lime kiln with physical separation
OF-LK&RB	Oxy-fuel combustion in both the recovery boiler and the lime kiln
OF-RB+SEPA	Oxy-fuel combustion in the recovery boiler with physical separation
PSA	Pressure swing adsorption
RB	Recovery boiler

R&D	Research and development
SEPA	Physical separation unit
SYNTH	Synthetization to DME
Tekes	The Finnish Funding Agency for Innovation
Vol-%	Percent by volume
VTT	Technical Research Centre of Finland
C	Investment cost
C_0	Reference investment cost
i	Interest rate
$\dot{m}$	Mass flow
n	Number of annuities
R	Scale-up factor of investment
S	Size or production volume
S_0	Reference size or production volume
T	Temperature

1. INTRODUCTION

CLIC Innovation Ltd coordinates research within the *Carbon Capture and Storage Project* (CCSP) that supports the development of carbon capture and storage (CCS) technologies [1]. This thesis contributes to the project by evaluating the carbon capture options for Kraft pulp mills and assessing the related bio-CCS potential as well as the effect of political instruments on the economic feasibility of the studied technologies.

Climate change drives governments around the world to reduce *greenhouse gas* (GHG) emissions. The Finnish government aims to reduce the total GHG emissions from Finland by 80-95 % from the 1990 level by 2050, as stated in the government report Energy and Climate Roadmap 2050 [2, p. 9]. According to the report, reaching this goal depends largely on the successful commercialization of *carbon capture and storage* (CCS) technologies. Much research on the subject has already been conducted [3-7], but most of the existing research has been focusing on applications for energy production, especially coal-fired power plants, steel industry, oil refining and cement industry because of their large carbon dioxide (CO_2) emissions and energy consumption. Some of the capture methods developed for other industry sectors could also be implemented in the pulp and paper industry.

In most cases it is currently not economically feasible to capture carbon and therefore market supporting mechanisms are needed. Utilizing the captured CO_2 would be optimal, but the associated capture potential is currently small compared to the emissions. Even in the USA, the industrial need for CO_2 was only 2 % of the emissions and enhanced oil recovery (EOR) accounted for 80 % of that in 2002 [7, p. 42]. An example of a market supporting mechanism for CCS is the *European Union Emission trading system* (EU ETS) [8]. EU ETS regulates the maximal amount of *fossil* CO_2 and other GHG that may be emitted by large energy intensive industries and aviation. To support CCS in pulp mills, the current EU ETS should be modified to include also *biogenic* emissions. The support from EU ETS should also be more reliable, as the emission allowance price has fluctuated from 30 €/t(CO_2) to zero [8, p. 4; 9, p. 17] and between 6 and 8 €/t(CO_2) in 2015 [10]. With the CO_2 reduction goals as strict as Finland has, other political measures may be needed. The effects of the EU ETS and other political instruments are discussed in more detail in Chapter 2.3.

In the Nordic countries the role pulp and paper industry is very significant. As the Finnish public statistics reveal, pulp and paper industry accounted for 50.6 % of the energy consumption [11] and around 30 % of the *fossil* CO_2 emissions [12, p. 291] of the Finnish manufacturing industry in 2012. Even if the fossil CO_2 emissions of the pulp and

paper industry are less than a third of the total manufacturing industry emissions in Finland, the biogenic CO_2 emissions that originates from wood holds a much greater capture potential, since the pulp and paper industry in Finland uses as much as 82 % wood-based fuels [13, p. 11].

When CO_2 is released to the atmosphere, it acts the same way regardless of its origin. However, biogenic CO_2 originates from the carbon of living organisms, which in turn have absorbed the carbon from the atmosphere through photosynthesis. By extracting biogenic carbon from the normal growth cycle a possibility to remove CO_2 from the atmosphere arises. This effect is also referred to as *negative emissions* and the concept as *bio-CCS* [14, p. 22] or *biomass energy with carbon capture and storage (BECCS)* [15, p. 4]. According to Kemper the term *BECCS* relates to power production whereas *bio-CCS* is considered to represent a wider context [16]. Thus the term bio-CCS is used in this work on pulp and paper industry.

Bio-CCS is usually associated with the risks of *indirect change in land use (ILUC). ILUC* means that when biofuel raw ingredients are grown on land suitable for food production and the need for the food persists, the cropland for the food is obtained elsewhere such as from forests and grass lands [17]. However, the forest and grass lands store greater amounts of carbon than the biofuel crop. When additional biomass needs to be grown for bio-CCS purposes, the concerns about ILUC may be relevant. This is however *not* the case when implementing bio-CCS in pulp mills, as the aim is to store the biogenic CO_2 that is currently being emitted to the atmosphere.

By producing wood-based products, the forest industry temporarily stores significant amounts of carbon [18, p. 555; 20]. Effective use of raw material is also essential for minimal impact on the climate. This is achievable as the same companies that own pulp mills also often have saw mills and thus timber waste is efficiently used for pulping [19].

In Figure 1, an overview of the biogenic carbon cycle of a wood-based product is presented.
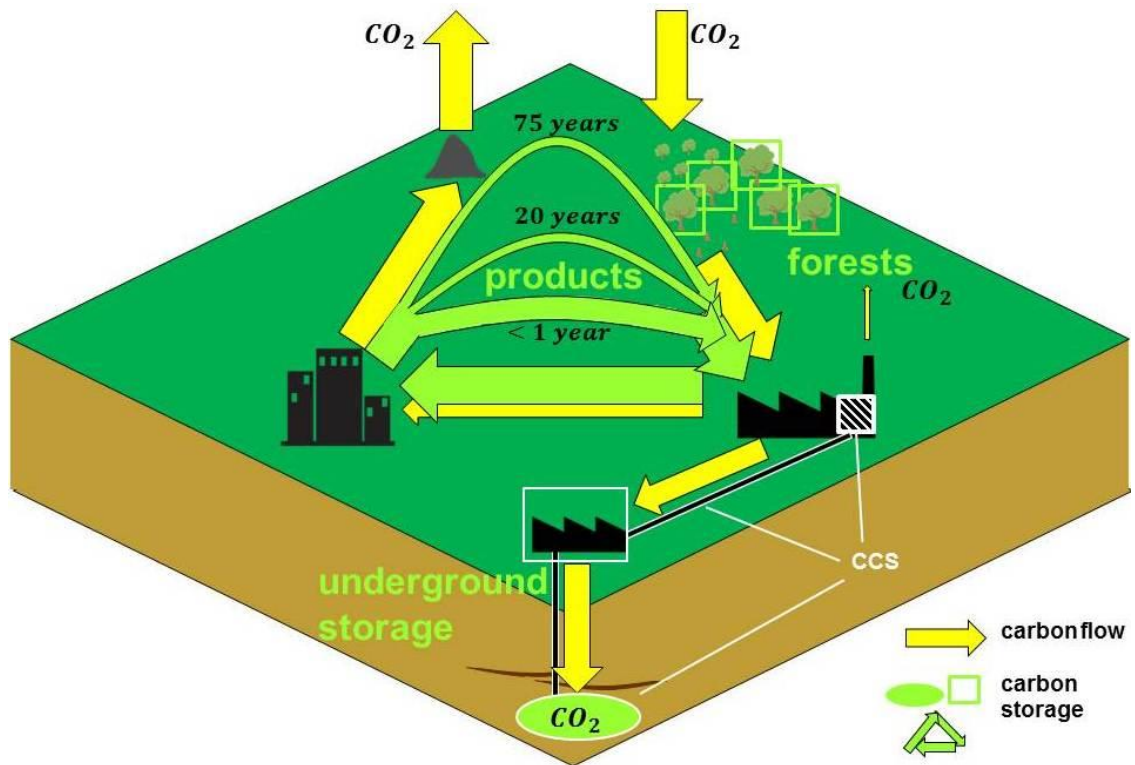
Figure 1. *An overview of the biogenic carbon cycle regarding wood-based products. Wood product average life times [20, p. 2]: biofuels and newspapers < 1 year, furniture 10-30 years and wooden buildings 75 years.*

The pulp and paper industry alters the natural carbon cycle of the forest. The natural cycle consists of growing trees storing CO_2 and dying trees releasing CO_2 until a saturation of stored carbon is reached [18, p. 547]. Producing wood based products adds another temporary storage to the cycle, but some of the CO_2 is emitted at the mill. Recycling products lengthens the carbon storage time. Eventually the carbon is released to the atmosphere, when the product decays or is burnt in a waste treatment plant. Last, CCS implemented in a pulp mill captures biogenic carbon and stores it permanently underground, thus removing CO_2 from the atmosphere and creating *negative emissions*. It is a valid question, which of these storages should be preferred, or all of them. The focus of this thesis is highlighted in the drawing with a white hatched box: is the CO_2 capture from pulp mills *feasible*.

In this thesis it is shown by which means CO_2 can most feasibly be captured from the Finnish pulp mills. To examine the different aspects of technical and economic *feasibility*, the following *research questions* were chosen:

- Which carbon capture technologies are implementable in pulp mills in the near future?
- How much CO_2 could potentially be captured with each technology?
- What is the *break-even price (BeP)* for the emission allowance if biogenic emissions were included?

The break-even price is the price that covers the costs of applying a carbon capture technology, but in this thesis the transportation and storage costs are excluded. To answer the research questions, an extensive literature review on carbon capture technologies was conducted. Three novel carbon capture technologies were modelled and one calculated based on mass and energy balances and then compared with a reference Kraft pulp mill scaled to the production capacity of 1200 ADt/d to assess the costs of each technology. Finally, the previous results and the technologies studied within this thesis were compared under same assumptions regarding the commodity costs, investment parameters and applicable policies. The readiness of each technology was evaluated based on previous research in pulp and paper industry as well as in other industry sectors. The capture potential of each carbon capture option was investigated by examining two of the most significant factors: first, in how many and how large mills could the technology be implemented and second, how much carbon could be captured.

The context for the analyses was the current and expected pulp and paper industry in Finland by 2030. When comparing the technologies, the system boundaries were set to contain only the carbon capture process and the immediately affected units. The transportation and storage costs were only briefly reviewed based on previous research [5, 21].

In this study it has been shown how much the carbon capture should be supported in order to be economically feasible. Technologies including new by-products or other revenue streams may be profitable even without support, but the associated carbon capture potential seems limited. Some of these technologies are still in early stages of development and more pilot and demonstration plants are needed. Such practical studies could be facilitated by cooperation between researchers and the industry.

2. CARBON CAPTURE, UTILIZATION AND STORAGE

Carbon capture, utilization and storage (CCUS) are a means of mitigating the CO_2 emissions from industrial sources. In this thesis CCUS refers to carbon capture and storage (CCS), carbon capture and utilization (CCU) or both. In CCS the CO_2 is first separated, then cleaned and compressed before transporting it to a storage site. In CCU the CO_2 may either be utilized on site or transported to another facility.

According to previous reports [2, p. 7; 21, p. 14; 6, p. 11], *CCS* is a key technology in achieving large scale CO_2 reductions. Utilization of the captured carbon instead of storage would reduce the costs, as the transportation and storage cost may form more than half of the total CO_2 avoidance cost in the Finnish context, as estimated by Teir et al. [5, p. 51]. However, the current utilization potential in Finland is less than one percent of the annual CO_2 emissions [5, p. 14]. New utilization methods, such as synthetic fuel production from CO_2 and H_2, might increase the potential significantly [22]. In the USA, more CO_2 is currently needed for enhanced oil recovery (EOR) [7, p. 42].

CCUS technologies are usually applied to *large point sources*, like power plants and industrial plants, but in the long term other applications should be considered too, as Wilcox states in the first book on carbon capture principles [23, p. 1]. With current technologies the cost for capturing the carbon even in larger scale applications may be as high as 30-85 €/t(CO_2) [5, p. 51]. The large cost range demonstrates the strong dependency on case assumptions. It seems probable that supporting policies like the EU ETS and carbon tax or other sources of revenue are still needed in the near future for wide spread implementation. Some policies, like biofuel support, may be applicable even if support for capturing biogenic CO_2 might not be included in other policies, such as the EU ETS.

2.1 Stages of carbon capture, utilization and storage

The objective of CCUS is to produce a concentrated stream of CO_2 for storage or utilization from flue gas or other gas stream. In principle, it would be possible to store the flue gas as such, but because flue gases usually contain only 3-15 % CO_2 and because of high flow rates, this would be impractical. [5, p. 14] The path of CO_2 from the flue gas to storage or utilization comprises three steps: capture of the CO_2 from a gas stream, transportation to the storage or utilization site and storage or utilization. Additionally, depending on the carbon capture method and destination, different types of treatment

like cleaning and compression may be needed before transportation. [5, p. 15] In the following chapters these steps are presented in brief mainly based on the Special report on carbon capture and storage by the *Intergovernmental Panel for Climate Change (IPCC)* [3]. Later, a detailed literature review on the carbon capture technologies applicable for pulp mills is presented in Chapter 4.

2.1.1 Carbon capture

There are numerous technologies available rather than a single one for carbon capture. A much used grouping of the carbon capture technologies is *pre-combustion*, *post-combustion and oxy-fuel combustion* [3]. Additionally, technologies like *lignin separation* and *fuel switch* are currently used for reducing the carbon emissions in pulp mills, even though they are not considered as carbon capture. For example, replacing fossil fuels with biofuels reduces the fossil CO_2 emissions at a pulp mill or in the surroundings if an extracted biofuel is sold.

Two carbon capture technologies, monoethanolamine (MEA) absorption and physical separation by cooling down the CO_2, suffice to form all the thirteen carbon capture options examined in this thesis. This is because the carbon separation technologies from gas streams are combined with other methods, such as oxy-enrichment and some methods separate carbon before combustion. Moreover, it may be possible to alter the combustion process with oxy-combustion so that the flue gas stream is already pure enough for utilization or storage and less treatment is needed. Usually some 90 % of the CO_2 in the flue gas can be captured, like for instance with MEA absorption [24, p. 89].

There are several reasons for choosing MEA absorption and physical separation as the reference CO_2 capturing methods from gas streams. Firstly, choosing only a couple of CO_2 separation methods from gas streams allows a closer examination of the carbon capture options for the targeted pulp mill units and their combinations. This gives a broader view of the total CCUS possibilities in a pulp mill. The chosen separation method could have been temperature or pressure swing adsorption or even membranes, but the long experience from MEA absorption in natural gas sweetening and enhanced oil recovery (EOR) since the 1970s [25, p. 3] has given it an almost standard position as a reference CO_2 separation method. In the reference studies presented in this thesis regarding carbon capture in pulp and paper mills, MEA absorption was also used for air combustion processes [4, 26]. On the other hand, physical separation is attributed to high separation quality and is more economical when the initial CO_2 concentration is high, as in the case of oxy-fuel combustion [27, p. 3484].

Before transportation the CO_2 stream has to be purified for safety and technical reasons among all and then liquefied. The associated compression work for liquefaction is significant, as the typical pressure is around 100 bar. The case specific costs and required CO_2 quality depend mostly on the transportation method and storage site in question. [6,

pp. 25-26] The compression work and purification are usually included in the carbon capture costs.

2.1.2 Transportation

The CO_2 is transported either with pipelines in a pressurized super-critical phase or with tanker ships in a liquid phase. The corresponding pressures would be around 100 bar and 5-10 bar, respectively, whereas the temperature for ship transport would be around $-50°C$. In general, it would take less time to set up a transportation system with ships than pipelines, but the transportation might be more costly. [6, p. 25; 21, p. 32] In this thesis tanker ship transportation is assumed for the modelled cases, because it could be implemented even in smaller scales and seems more probable in the near future.

In the Finnish context, the location of the plant or mill implementing CCS is essential for economical transportation of CO_2, since there are no suitable storage sites in Finland [28]. According to the final report of the EU GeoCapacity project by Vangkilde-Pedersen et al. the closest suitable formations are located in northern Germany and Poland, Denmark, Norway and the surrounding seas [29, p. 37]. In 2011, the only proximate sites with carbon storage activity were the Norwegian gas fields of Snöhvit and Sleipner [5, p. 33]. As tanker ship transportation seems more likely to take place, mills on the coastal area have an advantage over inland sites.

The transportation costs in the Finnish context are typically higher than assumed in other studies, since the distances are longer compared to central Europe. The probable transportation costs from coastal Finland were estimated by Teir et al. [5, pp. 44-46] to be 7-16 €/t(CO_2) depending on the annual total amount of CO_2 delivered. Additional transportation costs from inland sites to the coast even out to around 4-5 €/t(CO2) for all pipelines from 50 to 350 km in length, when the annual CO_2 flow exceeds 5 Mt(CO_2). In order to achieve lower transportation cost, the emissions of multiple point sources should be combined. As the focus in this thesis is only in the carbon capture part of the CCS chain, the transportation costs were only used in this thesis to estimate the advantage of technologies that require no CO_2 transportation.

2.1.3 Utilization

Utilizing CO_2 in any manner that does not release it afterwards would likely mean great savings in comparison to carbon storage. This is because there would be no storage costs, the transportation costs could decrease significantly and in the case of on-site utilization compression costs might also be reduced. Additional value might be gained from the actual final use of the CO_2. For instance, in 2007 Rubin et al. [30, p. 4446] suggested the value of injected CO_2 for *enhanced oil recovery (EOR)* to be 16-27 $(USA, 2002)/t($CO_2$) which is around the same in €(2002).

The most significant use of CO_2 currently is for EOR, which for example in the USA accounted for 80 % of industrial CO_2 use in 2002 [7, p. 42]. Other uses include carbonated beverages, growth fostering in greenhouses, algal fertilizers [31], inert gas for welding and refrigerants. Pulp mills may use CO_2 for acidity control, treating pulp or in creating filler substances. However, the total industrial usage of CO_2 in Finland is currently less than one percent of the annual emissions. [5, p. 14] Future products, such as synthetic fuel production from CO_2 and H_2 might increase the potential [22]. For the presented reasons, the most interesting utilization possibilities would be on-site use at the pulp mill or possibly transportation for future EOR purposes to the closest oil wells or gas fields.

2.1.4 Storage

Unutilized CO_2 needs to be stored in a safe and permanent way. This means that the risk of CO_2 leaking back to the atmosphere should be acceptably small and eventually fade away. Another challenging feature is the vast amount of CO_2 to be stored – millions of tons every year. Currently the only storage possibility that has been demonstrated in full scale is underground in *geological formations* [6, p. 28].

The safe storage of CO_2 into geological formations has certain requirements. Firstly, the CO_2 is injected in a supercritical phase with a pressure of more than 74 bar. This way the CO_2 acts like steam, pushing into the pores and fractures of the rock. Over time, the CO_2 dissolves into water forming a denser liquid layer under regular groundwater. Eventually – after thousands of years – the CO_2 would form carbonates and be stored permanently. Nevertheless, the storage needs to be controlled for a long time to prevent any leakage. Secondly, the storage needs to be at least 800 meters underground, so that the surrounding pressure exceeds the pressure of injected CO_2. Finally, the formation needs to be naturally well-sealed. Such formations can be found both on- and off-shore. Most practical options include depleted gas and oil reservoirs, saline formations and unminable coal formations. Additional site-specific requirements regarding for instance the quality of stored CO_2 may also apply. [6, pp. 29-30; 21, pp. 36-38]

In the EU GeoCapacity project the total storage potential in Europe was estimated to be around 120-360 Gt(CO_2), of which 25-55 % located in off-shore Norway [29, pp. 20-21]. As CO_2 is already being stored in this region, this seems like the most probable storage site for CO_2 captured in Finland. According to Teir et al. [5, p. 48] the CO_2 storage costs in the Finnish context is estimated to be about 11-12 €/t(CO_2).

Even though storage in geological formations is currently the only economically feasible option, other possibilities are being studied as well. Storage in deep oceans might be possible, but its environmental impacts are unknown and therefore current laws have also prohibited it [21, p. 35]. *Mineralization* or *mineral carbonation* means the storage of CO_2 as calcium or magnesium carbonates. This option provides a secure way for CO_2

storage in Finland and the product carbonate could be used as construction material or landfill without need for control afterwards. However, the process is very energy intensive and costly requiring minerals mining, fine grinding, high pressure and increased temperature as well as chemical treatment. [21, p. 39]

2.2 CCUS projects

According to the Global CCS institute [32], there are currently fourteen operational large scale CCS projects storing 27.4 $Mt(CO_2)$/a altogether. The growth of CC(U)S is rapid; the total annual stored amount of CO_2 has doubled in the last five years. Additionally, eight projects are under construction with around 13 $Mt(CO_2)$/a additional capture capacity and many more being planned.

In most of the projects CO_2 is utilized in EOR and in only three projects geological storages are used. A major step was reached in 2014 as the first power plant with CCS began operation. [32] Despite the rapid growth, the CCUS projects have faced difficulties too. For instance, in 2014 a large oxy-fuel project Schwarze Pumpe was abandoned [33].

2.3 CCUS in the Finnish climate policy and EU ETS

The Finnish government aims to reduce GHG by 80-95 % from the 1990 level [2, p. 9]. This ambitious goal is pursued with various political instruments, such as participating in the EU ETS, biofuel *delivery obligation* [34], *green electricity certificates* [35], involving companies and municipalities with a *voluntary energy efficiency scheme* and supporting renewable energy with *lower taxation, investment support*, *research and development funding* and *feed-in tariff for wind power* [21, p. 95].

EU ETS is one of the most important policies supporting CCUS in Europe. EU ETS regulates the maximal amount of fossil CO_2 and other GHG that may be emitted by largest industrial point sources, such as pulp and paper mills, as well as aviation. The fixed amount of total annual emissions is lowered stepwise in the future to decrease the total GHG emissions in Europe. The included companies are required to purchase the amount of emission allowances that covers their GHG emissions. The emission allowances may also be traded and so the price for emitted CO_2 is regulated indirectly. The EU ETS still needs refinement as the emission allowance price has fluctuated from 30 €/t(CO_2) to zero [8, p. 4; 9, p. 17] and between 6 and 8 €/t(CO_2) in 2015 [10].

The current EU ETS directive [36, p. 54] does *not* include biogenic CO_2 emissions and most of the CO_2 emitted in the pulp and paper industry is thus excluded. An impact assessment of the European Commission [37, pp. 178, 184] for the next phase lasting until 2030 shows negligible financial pressure on pulp and paper industry and only slight incentive for reducing its carbon footprint.

Careful consideration is needed, should biogenic emissions be included in the EU ETS. Firstly, combustion of wood is in principle a carbon neutral loop and the emissions should not be charged. The pulp and paper industry also temporarily stores much carbon in its products. The industries might also move their production into other countries where the EU ETS and other emissions controlling policies do not apply which is known as *carbon leakage*. On the other hand the emitted CO_2 acts the same way regardless of its origin and thus carbon capture should be supported for both fossil and biogenic CO_2.

A possible solution to provide an incentive to capture biogenic CO_2, but not punish from emitting it could be including biogenic CO_2 in the EU ETS while continuing to offer *free allocations*. Manufacturing industries receive a decreasing amount of free allocations to avoid *carbon leakage*, to reward from efficient energy measures and bioenergy use. [8] The emissions of a company receiving free allocations are included in the EU ETS, but a proportion of the allowances are given to the company free of charge. If the company uses the allowances, the net effect is zero, but if the company reduces its emissions, it may trade the allowances freely to make a profit.

Even if biogenic emissions would later on be included in the EU ETS, it would not necessarily mean that financial pressure would be placed on the pulp and paper industry to implement CCUS. Instead, the industries could be rewarded for capturing the CO_2. In the European Commission Impact Assessment report [37, p. 178] it is estimated that also in the 2020-2030 period the pulp and paper industry would receive as much as 90 % of its emission credits as free allocations to avoid carbon leakage. The carbon leakage is a real concern in the industry as paper production is already shifting its balance towards South-America and Asia. However, no evidence of including biogenic emissions into the EU ETS was found in the EU Commission report by 2030. Thus, the implementation of bio-CCS is lacking incentives, at least when it comes to EU ETS.

The financial encouragement for pulp and paper industry to cut emissions based on EU ETS seems minor until 2030. The Finnish government may implement other methods to achieve the strict emission reduction targets. Should this be the case, the issue of carbon leakage still has to be accounted for. The low amount of political support seems to point to direction that the financial benefits should be pursued elsewhere, like in energy efficiency, increased production or new products. Even if the EU ETS would not include biogenic CO_2, other policy instruments like lower taxation [38] or delivery obligations [34] may offer significant support. These policies could affect the profitability of the carbon capture options unequally, and for instance carbon capture options with biofuels production seem to have an advantage. The possible unequal support for different technology options should be worth examining in future policy making as well.

3. FINNISH PULP AND PAPER INDUSTRY

In this thesis the main concern is the pulp and paper industry of Finland and more specifically any production units that include pulping. The type of the reference mill is based on this context and thus *a Kraft pulp and paper mill* was chosen. Also the relevant *capture potential* and possible *political instruments*, like biofuel support, stem from this context. In short, the assumptions in this thesis are based on the context of the Finnish pulp and paper industry.

3.1 Characteristics of the industry

In 2013, pulping was the largest domestic wood consumer in Finland. The total volume of forests in Finland was estimated to be 2357 million m^3. This provided a yearly growth of some 104 million m^3 and additional 11 million m^3 round wood was imported to Finland. The total domestic wood consumption was 73.9 million m^3, of which pulping took the largest share of some 52 %. Sawmills consumed the second most wood, 31 %, and around 9 % was used as fuel. The 40 million m^3, which was not classified as domestic consumption, included addition of total forest volume, natural drain, logging residue and round wood export. [12, p. 31]

Papermaking is a traditional field of industry in Finland. The production units have constantly grown bigger in size leading to ever fewer owners. Despite that, most of the forests in Finland are privately owned.

According to the Finnish Forest Industries Federation, there were 50 operating pulp and paper mills in Finland in August 2015 [39], as shown in Figure 2.

Paper-, paperboard-, chemical pulp- and BCTMP-mills in Finland
Finnish Forest Industries Federation, 17.8.2015

- ● Paper mills (21)
- ● Paperboard mills (12)
- ● Chemical pulp mills (15)
- ● BCTMP mills (2)
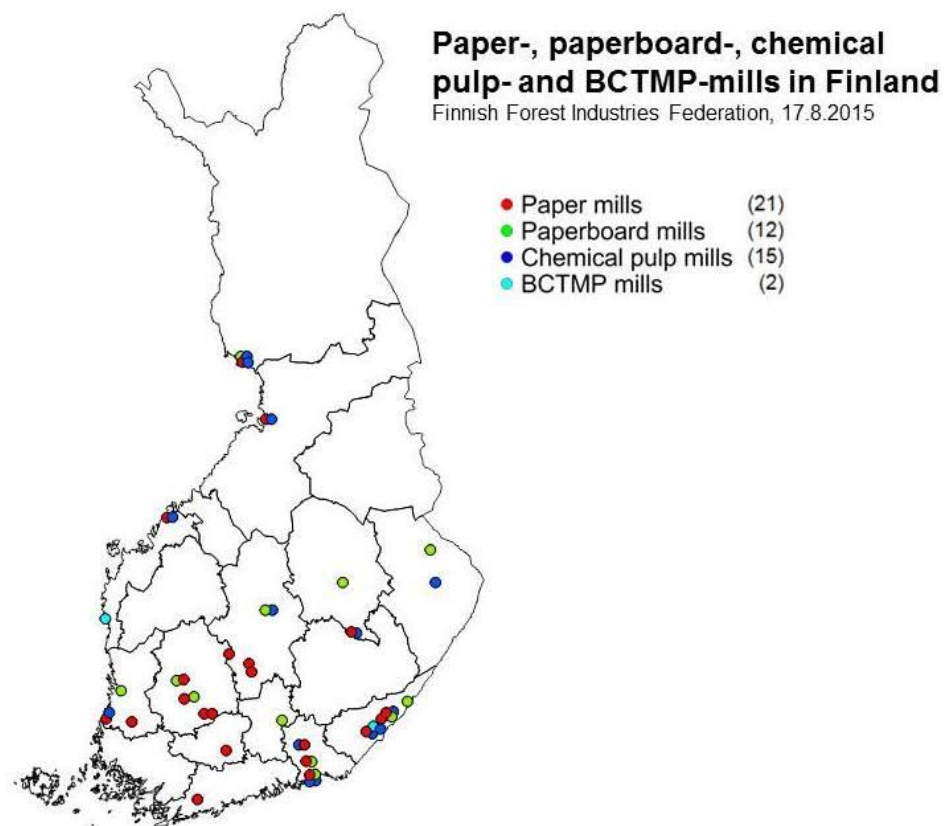
Figure 2. Paper-, paperboard-, chemical pulp and bleached chemi-thermomechanical pulp (BCTMP) mills in Finland. Adapted with permission from the Finnish Forest Industries Federation. [39]

It can be seen, that most of the Finnish pulp and paper mills are located in the southern and central Finland and mainly on inland sites. This may play an important role for the logistics method.

Since the millennium change, global overcapacity and the availability of cheap tropical fibre in the market has forced companies to reform their businesses. Finnish mills have been sold and shut down, but also new market areas and products are investigated. [19] Current trends include new *wood-based materials* as well as *biofuel production* [40]. As the businesses already need to be reformed, the possible implementation of carbon capture technologies might offer significant synergy benefits for these trends.

The wood flows in the Finnish pulp and paper industry are now used to roughly estimate the *total biogenic CO_2 emissions*. Such data was not as easily accessible as the data of *fossil* emissions, which was available due to public emissions trading in the EU ETS. Roughly 50 % of dry wood is carbon [41, 42] and the density lies somewhere between 300 and 550 kg/m^3 [41]. Therefore, some 15 million tons of carbon that flows through the Finnish forest industry every year. If this carbon is burnt stoichiometrically,

55 million tons CO_2 would be formed. Instead, much of this biogenic carbon is stored in products thus creating negative emissions. Pulping handles about half of this carbon flow – equivalent to 28.6 million tons of CO_2. A diagram illustrating the carbon flows in a modern pulp and paper mill is presented below in Figure 3, as published by Onarheim et al. [43].

Figure 3. *Carbon flows in a modern pulp and paper mill. Adapted with permission.* *[43]*

As can be seen, most of the carbon is emitted from the recovery boiler and around 40 % is stored in the pulp. Thus the total potential of bio-CCS is reduced by the same 40 % and the remaining 60 % form the theoretical total **bio-CCS potential of 17 Mt(CO_2)/a** in the Finnish pulp mills. Meanwhile, some **11 Mt(CO_2)/a** is already stored **in the pulp products**. These are only approximations, since some of the carbon exit the mill as waste and by-products, thus decreasing the potential of post combustion carbon capture for instance. In addition to this biogenic CO_2, some fossil CO_2 originating from used fossil fuels is emitted at the mills.

The pulp and paper industry continues to be a significant part of the Finnish economy. Besides being the largest domestic wood consumer, the value of pulp and paper industry is recognizable 2 700 million euros or 1.3 % of the gross domestic product (GDP) of Finland. The value of the whole forest sector in Finland has been around 6 200 million euros in 2014. [12, p. 349]

Pulping and papermaking is of great importance to the Finnish economy, but the uncertain future drives the industry to look for new alternatives. In addition, the industry contributes significantly to the carbon balance by storing biogenic carbon into products. Nevertheless, large mills emit significant amounts of CO_2 that could be captured. In order to study the possible carbon capture technologies, a typical Kraft pulp and paper mill is presented as a reference in the following chapter.

3.2 Combined Kraft pulp and paper mill

A combined Kraft pulp and paper mill produces paper and by-products from wood with the help of chemicals. The production consists of two main processes: *pulping* and *papermaking*. Pulping is the production of *pulp* from raw ingredients, mainly wood. The pulp is a dispersion of separated wood fibres and water [44, p. 55], whereas papermak-

ing is the refining of pulp into dried paper. The pulping is of most interest in this study, because the recovery of its chemicals contains combustion processes characteristic to the pulp and paper industry, which are an important source of CO_2 emissions.

There are four main categories of pulping processes: *chemical, semi-chemical, chemi-mechanical* and *mechanical pulping*. Here, a chemical pulping process called *Kraft pulping* is used as a reference because of its wide spread use in Finland. [39]

The Kraft pulping process can be divided into two parts: the first one is called *fibre line* and the second one *chemical recovery*. The purpose of the fibre line is to process the wood into pulp for paper making. The chemical recovery regenerates the chemicals needed in the digester of the fibre line, but it is also important for manufacturing by-products, saving energy and environmental aspects [44, p. 101]. Figure 4 illustrates an overview of the combined Kraft pulp and paper making process.
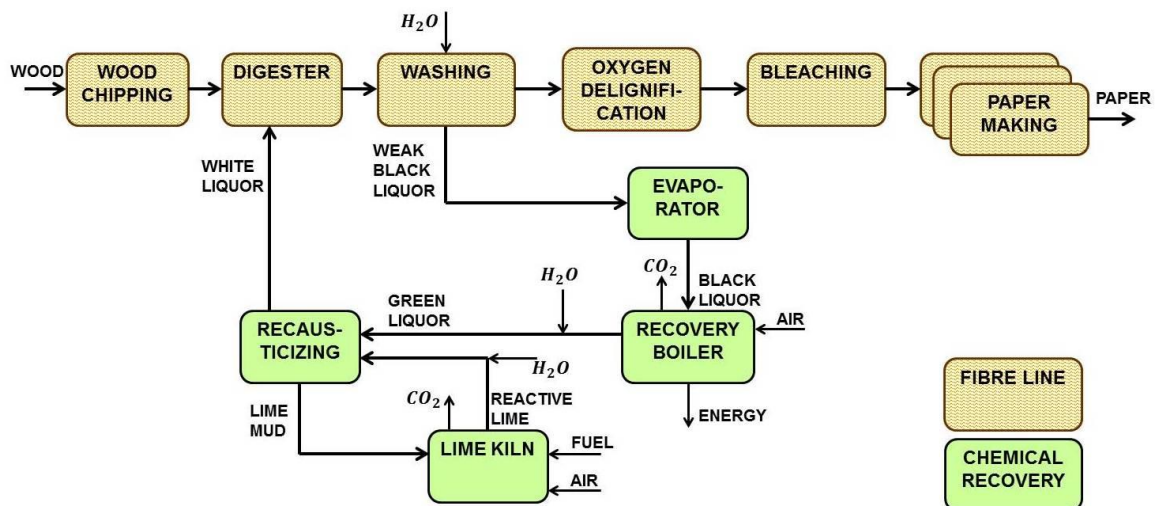
Figure 4. *A combined Kraft pulp and paper making process. [45]*

As can be seen from the diagram above, the fibre line consists of the unit processes of *wood chipping*, cooking in the *digester*, *washing* the pulp, *oxygen delignification* and *bleaching*, after which the pulp continues to paper making. [45]

The chemical recovery is of most interest in this work. It begins from the *digester*, which is the main junction of the fibre line and the chemical recovery loop. In the digester, *wood chips* are cooked in a mixture of chemicals called *white liquor*, consisting mainly sodium hydroxide (NaOH) and sodium sulphide (Na_2S), to separate the fibres into pulp. The pulp is washed with water and the spent chemicals dissolve to form *weak black liquor*, which is concentrated in a series of *evaporators*. After this, the concentrated *strong black liquor* is burnt in a *recovery boiler*. In the recovery boiler the inorganic components are recovered as smelt and dissolved in water to form *green liquor*, essentially sodium sulphide and sodium carbonate (Na_2CO_3). Many combustion reactions take place in the recovery boiler, which motivates its closer examination later in this

study. Next, the green liquor is converted to white liquor in a process called *recausticizing*. The sodium carbonate in the green liquor reacts with calcium hydroxide (Ca(OH)$_2$) to form sodium hydroxide (NaOH) and calcium carbonate (CaCO$_3$). The reaction is

$$Na_2CO_3 + Ca(OH)_2 \rightarrow 2\,NaOH + CaCO_3. \tag{1}$$
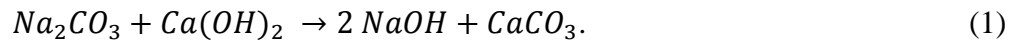
The white liquor continues into the digester unit to be reused in the cooking of wood chips. The calcium carbonate is converted to calcium oxide (CaO) and carbon dioxide (CO$_2$) in a long furnace called the *lime kiln*. This reaction requires a lot of heat and therefore the lime kiln consumes *fuel*. This gives a reason to inspect the lime kiln more closely in the next chapter. After the lime kiln, water is added to the calcium oxide to form the calcium hydroxide needed in the recausticizing. Additionally, the chemical recovery process provides for by-products such as tall oil, turpentine, heat and electricity. [44, p. 101-115]

3.2.1 Recovery boiler

The combustion reactions in the recovery boiler also result in large amounts of biogenic CO$_2$, thus making the recovery boiler the larges point source of CO$_2$ in a pulp mill Therefore, it is attractive for different carbon capture options.

The recovery boiler is a large and complex unit in which many chemical reactions take place. It is the most expensive single piece of equipment in a pulp mill [44, p. 110]. In short, first the concentrated black liquor is sprayed into the furnace. Second, the spray forms char and is then burnt with the help of a controlled amount of air. The molten sodium sulphide and sodium carbonate are collected into the dissolving tank in order to make green liquor. On the second hand, the exhaust gases flow through an evaporator, a super heater and an economizer for optimised heat recovery.

A schematic conventional recovery boiler is presented below in Figure 5.
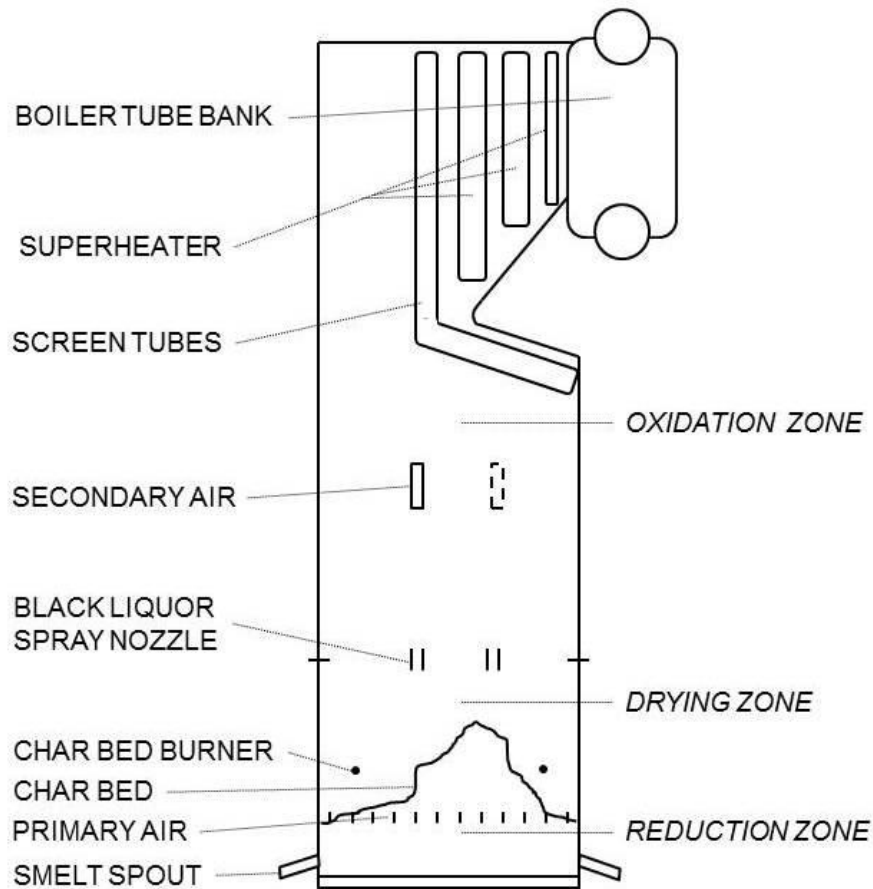
Figure 5. *Schematic figure of a conventional recovery boiler. [44, p. 109]*

The recovery boiler can be divided into three different zones according to the surrounding conditions: *reduction, drying* and *oxidation zone*. Changes in oxygen level as well as varying temperatures cause different chemical reactions to occur. The total reactions are those of *combustion* as well as the *conversion of sodium salts* and *reduction of a supplementary chemical*. The reactions other than combustion are [44, p. 110]:

$$2\ NaOH\ +\ CO_2 \rightarrow Na_2CO_3 + H_2O \quad \text{and} \tag{2}$$

$$Na_2SO_4 + 4\ C\ \leftrightarrow\ Na_2S + 4\ CO. \tag{3}$$
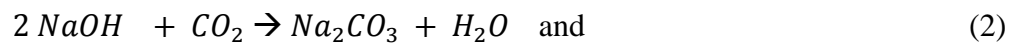
These reactions occur stepwise so, that changes in the later reactions should not affect the changes in the previous reactions.

3.2.2 Lime kiln

As it was said, the main function of the lime kiln is to convert the *calcium carbonate* from the recausticizing back to *calcium oxide* and finally *calcium hydroxide*. This requires heat and high temperatures up to 1200 °C. Conventionally a burner fuelled with

oil or natural gas is used, but many mills use gasified biomass. The combustion of these fuels results in CO_2. The other reaction in the lime kiln, called *calcining*,

$$CaCO_3 + heat \rightarrow CaO + CO_2, \tag{4}$$

also results in CO_2.

These two reactions make the lime kiln another point source of CO_2 and possible for carbon capture technologies. In fact, the calcium loop of a pulp mill resembles greatly the novel concept of *calcium looping combustion* as presented by Blamey et al.*,* especially if the lime kiln is operated as an oxy-fuel process [46, p. 262].

It should be noted, that the CO_2 *from the calcining reaction* originates from wood and is therefore mainly biogenic. Only supplementary lime used to cover losses might be of fossil origin. The specific amount of CO_2 from each origin has been studied by Miner and Upton [47, p. 734]. 196 kg of biogenic CO_2 per ton of pulp product originates from the calcining and around 100 kg from burning fossil fuels leading to an estimate for the total CO_2 emissions of around 300 kg(CO_2)/ADt. The fuel-related emissions range widely from 50 to 300 kg. It should be noted, that the *biogenic* CO_2 emissions of the lime kiln are about an order of magnitude smaller than that of the recovery boiler, as was seen in Figure 4 in Chapter 3.1. The lime kilns are still the most potential targets in pulp mills for capturing *fossil* CO_2.

A conventional lime kiln is a counter-current process, where the lime mud flows in the opposite direction as the combustion gases. The process is presented in Figure 6.
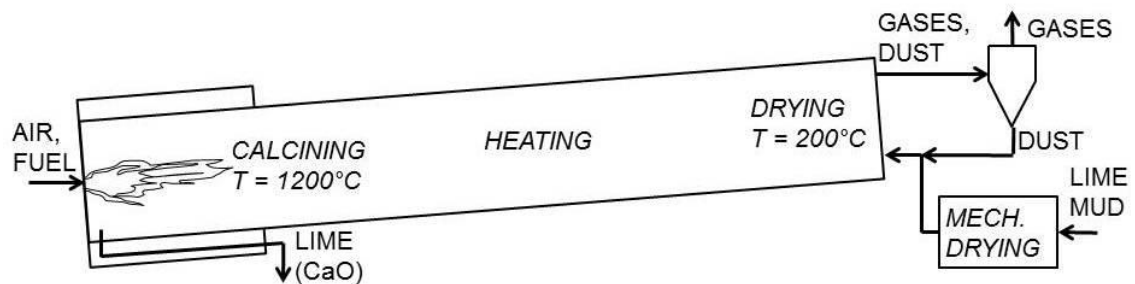
Figure 6. A schematic figure of a conventional lime kiln. [44, p. 117]

Before conveying the calcium carbonate, or *lime mud,* into the lime kiln the lime mud is dried mechanically usually with a vacuum drum filter or disc filter. In the lime kiln the mud is thermally dried further. Then it is heated with the combustion gases in a long rotary furnace, typically more than a hundred meters in length. Heating the lime mud results in the calcining reaction presented above in Equation 4. The produced calcium oxide is cooled, screened and crushed. [48, pp. 13, 161-191]

Additionally, *lime dust* is captured from the exhaust gases with cyclones and fed back to the process to save raw ingredients. Nevertheless, some five percent supplementary lime, possibly of fossil origin, is needed to cover the losses [49, p. 24].

3.2.3 Multi-fuel boiler

Another source of CO_2 emissions at a pulp mill is the *multi-fuel boiler* used heat and electricity production. The fuel used in pulp mills may be fossil, biomass or a combination of fuels, hence the name multi-fuel boiler. In modern Finnish pulp mills wood residues and peat are used, as stated in recent news [50, 51]. Other names used for the boiler are *bark boiler* or auxiliary boiler.

According to boiler providers [52, 53], multi-fuel boilers are used also for instance in agriculture and power production from municipal waste. Because of its less unique nature compared to the lime kiln or the recovery oiler, the multi-fuel boiler is left with little attention in this study. Typical fossil emission reduction methods would include various energy efficiency measures leading to lower fuel consumption and to increase the proportion of biomass used, as explained for instance in the United States Energy Protection Agency report [49, pp. 10, 22].

4. CARBON CAPTURE IN PULP MILLS

As was seen in the previous chapter, pulp mills are significant point sources of mainly biogenic CO_2. This offers a possibility to create negative CO_2 emissions and therefore pulp mills seem potential for implementation of carbon capture technologies. Smaller amounts of fossil carbon flow through the plant originating from external fuels and supplementary chemicals. The combustion processes in the recovery boiler and the lime kiln are of specific interest and therefore all the investigated technologies concern the two units. In this study it is **assumed, that implementing carbon capture does not affect alter the pulp production**. In some technology options, exceptions to this are made regarding possible production increase.

There are several uses for CO_2 in a modern Kraft pulp and paper mill. The pulp and paper industry is also one of the largest CO_2 *consuming* industries in Finland [54, p. 159]. Possible uses of CO_2 in pulp and paper industry include regulating pH and reducing calcium carbonate dissolution [55, p. 5; 56], pulp washing [55, p. 7; 56; 57], soap acidulation [55, p. 9; 56], wastewater treatment [55, p. 10] and treating build-up in screening equipment [58]. More uses are being developed, but the big picture is still, that the total CO_2 utilization potential is only 0.5-1.0 % of the total human-caused CO_2 emission in Finland [54, p. 159; 5, p. 14].

Currently only a few pulp mills globally significantly implement carbon capture technologies. *Lignin separation* is in use in one operational mill in Domtar's Plymouth in North Carolina to produce lignin to the market [59] and another one will be applied in Sunila, Finland [60], which will use the separated lignin to replace fossil fuel in its lime kiln. A demonstration unit of *black liquor gasification to transportation fuels (BLG to DME)* is operational in Piteå, Sweden [61]. *Oxy-enrichment* without carbon capture is also piloted for recovery boilers in Sweden for improved energy efficiency and higher production capacity [55]. Many of technologies assessed in this thesis have not been tested in pulp mills yet. The experience from other fields of industry with similar equipment was however in some cases substantial. Finally, some of assessed technologies have only been studied with mathematical models and preliminary experiments. In addition to lignin separation, BLG to DME and oxy-enrichment, the technologies studied in this work included *air combustion with monoethanolamine (MEA) -absorption, black liquor gasification with combined cycle (BLGCC,) oxy-fuel combustion, chemical looping combustion (CLC) and pre-calcination before lime kiln.*

The technology options studied in this thesis and the replaced parts of the conventional process they replace are listed in Table 1.

Table 1. *Studied carbon capture technologies.*

Target	Technology	Added and replaced
Recovery boiler	1. Lignin separation	Added: Lignin separation
Recovery boiler	2. Air combustion with monoethano-lamine-absorption	Added: MEA
Recovery boiler	3. Oxy-enrichment with monoethano-lamine-absorption	Added: ASU, MEA
Recovery boiler	4. Oxy-fuel combustion with physical separation	Added: ASU, SEPA, new RB; Replaced: RB
Recovery boiler	5. Chemical looping combustion	Added: CLC, new RB; Replaced: RB
Recovery boiler	6. Black liquor gasification with combined cycle	Added: BLG, combined cycle; Replaced: RB
Recovery boiler	7. Black liquor gasification to transportation fuels	Added: BLG, motor fuel production; Replaced: RB
Lime kiln	8. Fuel switch	Added: new fuel; Replaced: fuel
Lime kiln	9. Pre-calcination before lime kiln	Added: pre-calcination
Lime kiln	10. Air combustion with monoethano-lamine-absorption	Added: MEA
Lime kiln	11. Oxy-enrichment with monoethano-lamine-absorption	Added: ASU, MEA
Lime kiln	12. Oxy-fuel combustion with physical separation	Added: ASU, SEPA, new LK; Replaced: LK
Recovery boiler and lime kiln	13. Oxy-fuel combustion in both the recovery boiler and the lime kiln	Added: ASU, SEPA, new LK, new RB; Replaced: LK, RB

The recovery boiler is targeted with seven technology options, the lime kiln with five options and both units are targeted with one option.

The studied technologies are also collected in Figure 7.
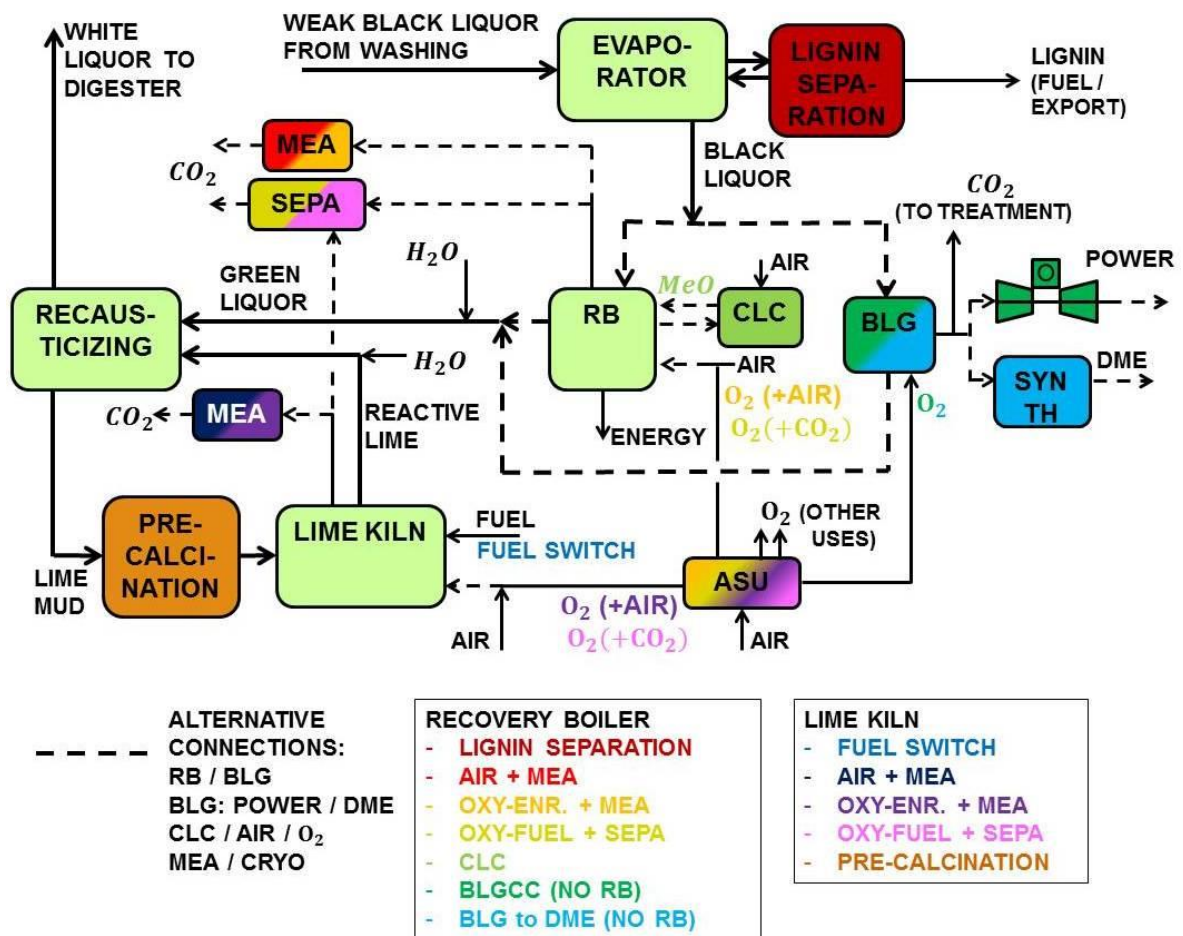
Figure 7. *Chemical recovery of a Kraft process modified with the studied technologies. ASU = air separation unit, MeO = metal oxide, RB = recovery boiler, SEPA = physical separation unit, SYNTH = synthetization to DME.*

In the diagram above, the light green units represent the conventional chemical recovery. Each of the other colors represents one technology and the multi-colored units are needed for multiple technologies. As can be seen, oxy-enrichment and oxy-fuel combustion are studied for both the recovery boiler and the lime kiln. In addition, the possibility of implementing oxy-fuel combustion in both the recovery boiler and the lime kiln was studied.

The recovery boiler options are discussed next in Chapter 4.1, thereafter the lime kiln options in Chapter 4.2 and finally the option regarding both units in Chapter 4.3. This thesis does not consider the application of several technologies with the exception of oxy-fuel combustion in the recovery boiler and the lime kiln.

In these chapters at least the following information is provided for each technology:

- Technical maturity
- Process description
- Requirements for implementing
- Carbon reduction potential
- Costs

Additional information may regard for example possible profits gained from implementation or safety notions found in previous literature.

4.1 Recovery boiler options

The studied carbon capture technologies that concentrate on the recovery boiler are *lignin separation, air combustion with MEA absorption, oxy-enrichment with MEA absorption, oxy-fuel combustion, chemical looping combustion, BLGCC* and *BLG to DME*. The order represents roughly the increasing amount of modifications needed in the recovery boiler. Thus the pre-combustion method of lignin separation is discussed first and BLG-options last as they replace the recovery boiler totally.

4.1.1 Lignin separation

Lignin separation can be used to lower the CO_2 emissions of a pulp mill by separating carbon rich lignin fractions before the recovery boiler. Lignin separation is most economical, when the capacity of the recovery boiler is limiting the maximal pulp production [59]. In the future, lignin use as raw material for instance for carbon fibres may make the process attractive for many more mills.

A number of technology options have been developed for separating, such as precipitation, membranes, ultrafiltration or nanofiltration [62]. Precipitation seems to be the readiest option, since commercial operation with the patented LignoBoost technology, owned by Valmet, is already operational [59, 60]. Therefore, LignoBoost is chosen as the investigated lignin separation technology in this study.

LignoBoost is a parallel operation to evaporation and comprises four steps. An overview of the process is presented in Figure 8.
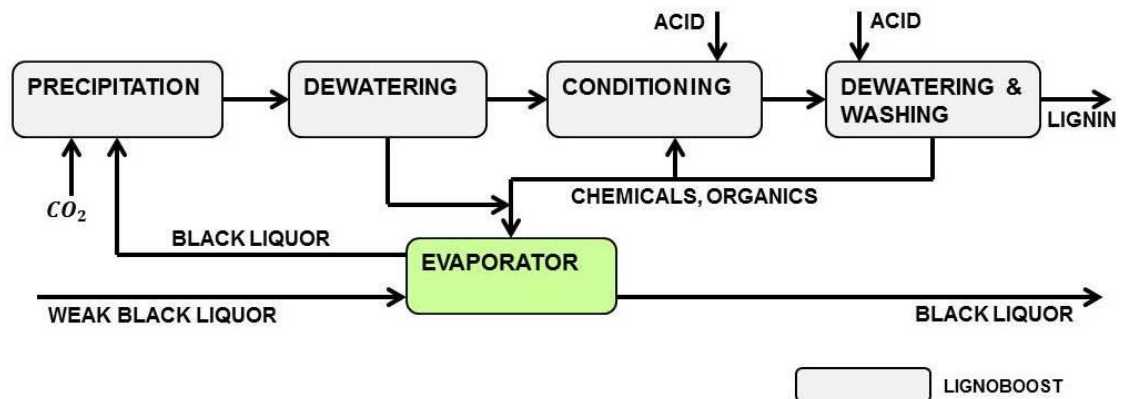
Figure 8. An overview of the LignoBoost process [63].

Black liquor from the evaporator is first *precipitated* by acidifying it with CO_2. The precipitated black liquor is *dewatered* with a filter press. Then the substance is dissolved again into reused washing water from dewatering and washing and acid in a stage called *conditioning*. Finally the treated mass is *dewatered* again *and washed*, resulting in concentrated lignin cakes. [63]

The LignoBoost technology can be applied for both existing and new mills. If the recovery boiler is limiting the production, more pulp can be produced through the implementation of LignoBoost, since the chemical processing capacity of the recovery boiler is increased up to 25 % [59]. However, a loss in electricity production is expected.

The end effect of lignin separation on carbon emissions depends on the end use of the separated lignin. Approximately 30-45 wt-% of the black liquor solids is lignin as presented by Rojas et al. [64, p. 88] and in this work a value of 35 wt-% is used as the Wallmo et al. [65, p. 12] present in lignin extraction context. Currently as much as 25 wt-% of the total lignin can be captured with LignoBoost [59], although in the future up to 50 % is expected [66]. As lignin contains more than 60 wt-% carbon [67, p. 121], a significant carbon flow is redirected to form a by-product of lignin. This flow would otherwise be emitted as CO_2 in the recovery boiler. In addition to usage as biofuel, lignin has numerous other uses, such as road dust control, water treatment and concrete additive [68]. In this thesis, it is **assumed that the sold lignin replaces natural gas in an external plant**.

4.1.2 Air combustion in recovery boiler with MEA absorption

For post combustion carbon capture *MEA absorption* is the conventional and most used option as it has been commercially availability since the 1970s. Therefore, MEA absorption is also chosen in this work for post combustion carbon capture from air combustion and oxy-enrichment flue gases in both the recovery boiler and the lime kiln.

A simplified amine absorption process is presented below in Figure 9, as described by Harun et al. [69, p. 296].
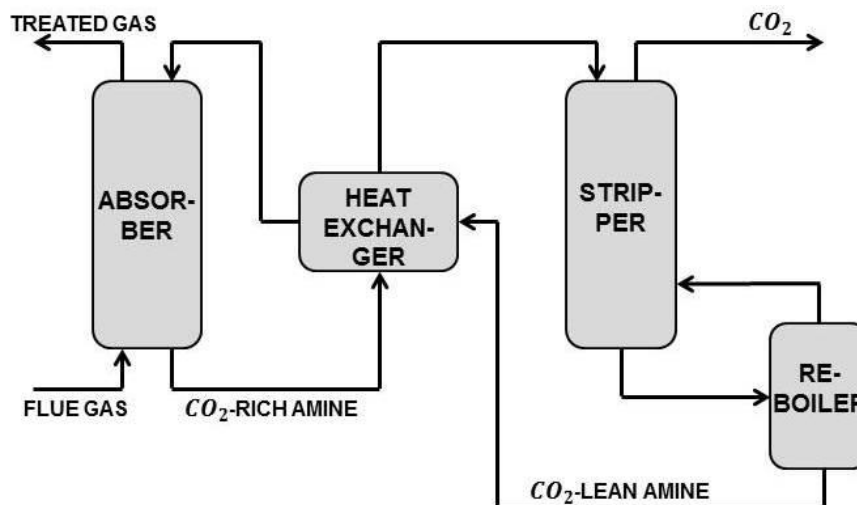
Figure 9. Schematic MEA absorption process. [69, p. 296]

In MEA absorption the flue gas enters the absorber, where a counter-current flow of absorbent captures the CO_2. Clean flue gas exits the absorber and is emitted through the stack. The CO_2-rich absorbent continues through the heat exchanger and is heated with the recycled lean absorbent before entering the stripper. In the stripper steam regenerates the absorbent stripping it of CO_2. The regeneration of the absorbent uses around 3 MJ/kg heat. [70, p. 8] This can be brought to the reboiler as low-pressure (LP) or medium-pressure (MP) steam depending on the process integration as explained by Hector & Bernetsson [26, p. 3027]. The energy consumption is highly dependent on the initial CO_2 level in the flue gas, as studied by Notz et al. [24, p. 106]. After cooling, the regenerated absorbent is recycled via the heat exchanger back into the absorber.

The energy penalty of MEA absorption is significant, but very good CO_2-capture rates and high CO_2 purity can be achieved. No operational full-scale MEA absorption plants were found in pulp and paper industry. The energy consumption together with the investment costs form the basis for the costs of MEA absorption. The energy penalty could be lowered, for instance by increasing the concentration of CO_2 in the original flue gas. Thus an additional investment in oxy-enrichment could lead to lower operating costs.

4.1.3 Oxy-enrichment in recovery boiler with MEA absorption

Enriching combustion air with oxygen is a relatively simple modification to the existing recovery boiler. Oxygen is mixed into the air supply and injected into the furnace resulting in increased oxygen content. Verloop et al. report modifications that increase the oxygen concentration in the combustion air from 21 vol-% to 25 vol-%. [71, p. 2] This leads to less nitrogen and therefore higher CO_2 concentration in the flue gas. Smaller

equipment might suffice leading to savings in investment costs. Additionally, a MEA absorption unit presented in Chapter 4.1.2 would be added for CO_2 capture. In very high initial CO_2 concentrations, like with oxy-fuel combustion, other capture methods may be even more efficient as discussed in the related oxy-fuel chapters.

Oxygen enrichment has technical and economic advantages and disadvantages. To produce the oxygen in large amounts, it is economical to invest in an *air separation unit (ASU)*. If the consumption of oxygen exceeds 20 t/day, the investment is likely to pay off [71, p. 2]. Toftegaard et al. [72, p. 589] citing a large body of evidence suggest that currently for large amounts of oxygen, *cryogenic distillation* is the only feasible technology. Cryogenic distillation is very energy intensive, consuming as much as 60 % of the total energy of carbon capture [72, p. 589]. One should not forget that extra safety measures are needed when dealing with almost pure oxygen. Oxygen enrichment may hold economic benefits as well, such as increased throughput of 10-20 % black liquor in the recovery boiler, reduced amount of flue gas, better combustion control, lower emissions and greater reduction efficiency [71, p. 2].

For recovery boilers, oxygen enrichment is commercially available [71, p. 1; 73; 55, p. 9], but no record of full-scale recovery boilers utilizing it was found. Only a year-long piloting period was mentioned in a company brochure [55, p. 9]. Nevertheless, oxygen enrichment has been long in use in many other fields of industry, such as glass, metal and cement production [71, p. 1].

4.1.4 Oxy-fuel combustion in recovery boiler

Oxy-enrichment was the concept of increasing the oxygen concentration in the air feed, whereas in *oxy-fuel combustion* the air feed is replaced with almost pure oxygen and recycled flue gas. In this thesis it is assumed that a carbon capture unit is needed to reach the required quality of CO_2 for transportation and storage. Toftegaard et al. [72, pp. 584-587] present an oxy-fuel combustion process for solid fuels. Their general, but coal-oriented presentation is modified here for the recovery boiler. For large industrial applications, such as the recovery boiler, oxygen is typically produced with an *air separation unit*. The oxygen is injected into the furnace together with recycled flue gas recovered from flue gas treatment. The reason for mixing oxygen with recycled flue gas, consists mainly of CO_2 and water vapour, is to reduce the combustion temperature and to decrease corrosion. Part of the flue gas stream always continues to additional treatment for carbon capture. The lack of nitrogen compared to air combustion leads to high CO_2 concentrations in the flue gas.

Figure 10 shows the described principles of oxy-fuel combustion in a recovery boiler.
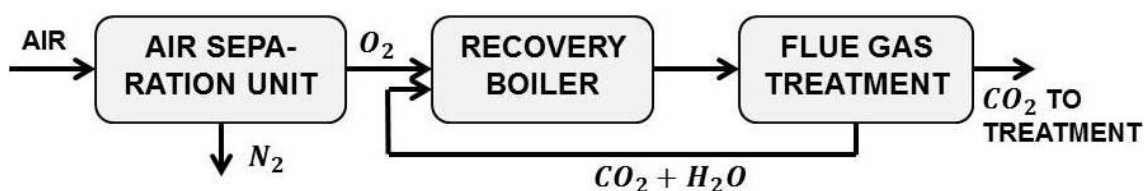
Figure 10. *Principal functioning of oxy-fuel combustion in a recovery boiler. [72, p. 585]*

Flue gas treatment includes compression, cooling and dehydration to condense water and purification [72, p. 588]. In the oxy-fuel options of this thesis the CO_2 is purified by repeatedly cooling it down and pressurising it in a *physical separation* process [74, Appendices 'PFD 5 & 6'] until a pressure of 6.5 bar, a temperature of -51 °C and a CO_2 concentration of 90 wt-% are reached.

Oxy-fuel combustion *in recovery boilers* is still in research phase. Oxy-fuel combustion could be a promising technology for carbon capture in power plants [72, p. 581]. For pulp mills, the implementation is more complicated due to different fuel composition and the original function of the recovery boiler in recovering the cooking chemicals. To maintain the recovery of the cooking chemicals the oxy-fuel modifications should not significantly alter the reactions in the reduction zone, as presented in Chapter 3.2.1. However, more data is needed to validate this assumption [26, p. 3026], possible acquirable from pilot and demo plants. In literature, oxy-fuel combustion is only listed as a "possible" technology in pulp mills [75, p. 14; 76, p. 82]. There is little literature on the subject and no pilots were found. In other industry sectors the experience with oxy-fuel combustion is broader. A 30 MW_{th} pilot power plant with oxy-fuel combustion was developed in Germany [33], in glass making oxy-combustion is applied commercially [77, p. 36] and for cement and metals production oxy-combustion is commercially available [78].

In addition to excellent carbon capture possibilities, oxy-fuel combustion may offer other advantages similar to those presented previously for oxy-enrichment. CO_2 emission reductions of 90-95 % are usually expected of oxy-fuel plants implementing carbon capture [72, p. 588], but the pilot plant in Germany was announced to reduce almost 100 % [33]. Low nitrogen concentration leads to low NO_x emissions, lower flue gas volumes [72, p. 590] and possibly lower flue gas treatment costs. If more of the benefits of oxy-enrichment [71, pp. 2-4] could be extrapolated, then oxy-fuel combustion in recovery boiler could also mean greater throughput, higher reduction efficiency and higher thermal efficiency in the recovery boiler. Air separation units also produce a great amount of nitrogen. It is estimated that for example a 500 MW_{th} oxy-coal power plant would require some 9700 tons of oxygen daily, thus creating a 31 800 ton by-product stream of nitrogen [72, p. 585]. Some of this nitrogen might be utilized on-site [71, p.

5]. The suggested uses are for instance pneumatic transport, pressure transfer and filling of empty containers, also known as blanketing.

Oxy-fuel combustion has also economic disadvantages. Firstly, the current production of oxygen by cryogenic distillation requires plenty of energy, up to 60 % of the total energy consumption of carbon capture [72, p. 589]. It is possible to minimize the energy penalty with optimized process integration. Secondly, the investment costs seem uncertain for the first pulp mill applications, since the recovery boiler needs structural changes [76, p. 82], but there is little knowledge available on these. With the current distillation technology, it is estimated that multiple large air separation units would be needed to provide enough oxygen [72, p. 589]. It is also recommended that a storage for liquid oxygen is added for secure operation of the oxy-fuel combustion unit. The liquid oxygen storage also helps dealing with changes in the recovery boiler process, which the ASU alone might not be able to follow.

Previous research has pointed out some technical issues that need to be addressed. For instance, *air leakage* into the process may cause significant dilution especially with aging equipment [72, p. 587]. As with oxy-enrichment, safety measures need reassessment. Tube wall temperatures in the recovery boiler may rise, leading to higher corrosion rates. Moreover, oxy-fuel combustion might increase the concentrations of corrosive substances. Higher temperatures in the lower furnace are expected to lead to higher particulate loading for the flue gas treatment [72, p. 587].

4.1.5 Chemical-looping combustion in recovery boiler

Like presented in the previous studies [79-84], in *chemical-looping combustion (CLC)* an *oxygen carrier* like metal oxide particles provides oxygen for combustion. This enables only oxygen, not nitrogen or other gases of the air, to enter the furnace. Like with oxy-fuel combustion, CLC produces nearly clean CO_2 without significant energy penalty. An overview of the CLC principle is presented in Figure 11.
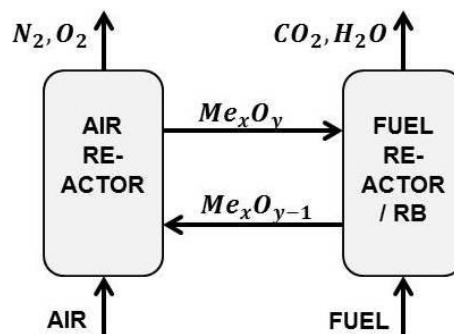
Figure 11. *A simplified chemical-looping combustion (CLC) process [79].*

The chemical loop consists of two reactors: one for air and the other for fuel combustion. First, metal oxide from the air reactor enters the furnace reactor carrying oxygen.

There, the oxygen is released during combustion. The reduced oxygen carrier is then directed back to the air reactor where re-oxidized by the oxygen from air. The CLC process results in oxides of the substances present in the fuel and in oxygen lean air. [79]

CLC for *recovery boilers* is still a very novel concept. No studies focusing on the subject specifically were found, but the idea was suggested before by Hektor et al. [26, p. 3025], stating that the implementation requires further investigation. CLC for power plants using coal or other fossil fuels has however been studied more. Several studies were found [79-84] and pilot plants are emerging, including one 1 MW_{th} plant [80] built lately in Darmstadt, Germany, exceeding any previous pilots in size.

There are many unsolved issues with CLC in recovery boilers. Firstly, if CLC is used for burning coal, the flue gas may need less treatment before transport [79; 81, p. 3102]. When applied to a recovery boiler a different fuel composition is expected due to the cooking chemicals and different nitrogen and sulphur concentrations. Therefore cleaning methods similar to oxy-fuel combustion may still be needed. This poses a possible economic drawback because of energy consumption and other operating costs. CLC has been studied mostly for combustion of gaseous fuels. Therefore the black liquor should probably be gasified before or during the combustion as explained in the previous studies [81, p. 3110; 84, p. 237]. This is a more complex process and requires further investments. Other modifications include the air reactor and a re-designed recovery boiler or black liquor gasification plant. A design where the same oxygen carrier circulates in two fluidized bed reactors is proposed by Lygnfelt et al. [81, p. 3103] for gaseous fuels and designs for solid fuels are presented in a review by Adanez et al. [84, p.237]. The original purpose of the recovery boiler, recovering the cooking chemicals, may be at risk. Due to the unresolved technical issues related to CLC in recovery boilers, the technology is **excluded from further analysis in this thesis**.

4.1.6 Black liquor gasification with carbon capture

Choosing *black liquor gasification (BLG)* usually replaces the conventional Tomlinson recovery boiler and is therefore mainly considered for *new* pulp mills. The resulting *synthetic gas* can either be used for electricity production in a combined cycle gas turbine or for motor fuels production. These processes are referred to as *black liquor gasification with combined cycle (BLGCC)* and *black liquor gasification to transportation fuels, in this work to dimethyl ether, (BLG to DME)*, respectively. In both cases, carbon capture can be included in the process [85, p. 35]. [86, p. 21]

Over the years, different BLG technologies have been studied. These include the SCA-Billerud process, the Manufacturing and Technology Conversion International (MTCI) process, the direct alkali regeneration system (DARS) process, BLG with direct causticization, catalytic hydrothermal BLG and the Chemrec BLG process [87]. Only few of these technologies have reached a commercial stage. In 2007, the International Energy

Agency (IEA) has estimated the Chemrec process to be the readiest technology [88]. This appears still to be correct, since a BLG to DME demonstration facility in Sweden has been operational since May 2012 [61]. Therefore the Chemrec process was chosen as the BLG technology in this study. The BLGCC and BLG to DME processes are presented in Figure 12.
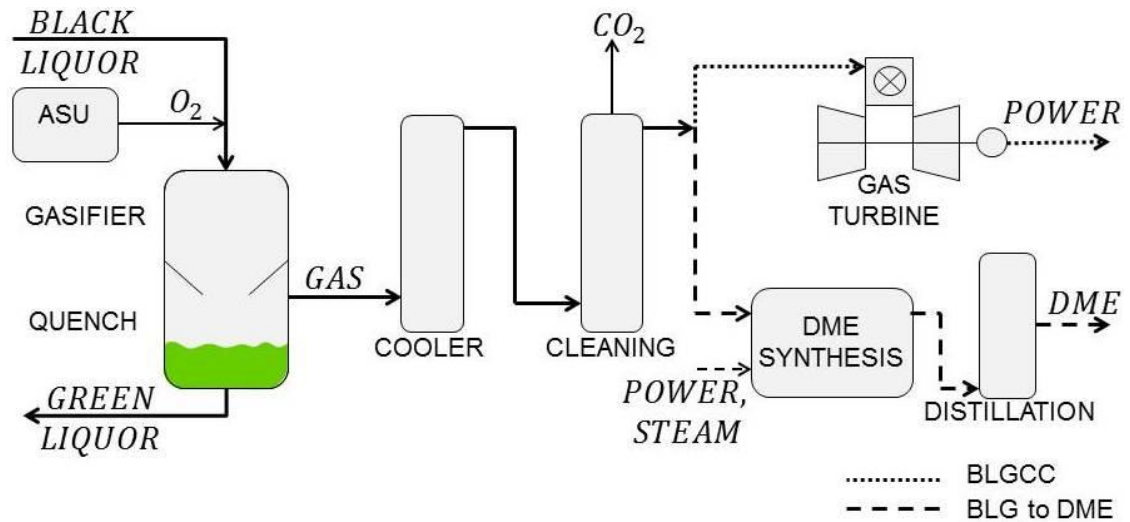
Figure 12. *Principles of BLG to DME and BLGCC. [86, pp. 22, 24; 89, p. 33]*

Black liquor is first evaporated in a pressurised reactor under reducing conditions. An ASU is needed to provide the necessary oxygen. Inorganic smelt and ash are removed from the reactor in a quench zone beneath the gasifier and are dissolved to form green liquor. From the gasifier, the raw synthetic gas is cooled in a counter-current condenser. If carbon capture is applied, the CO_2 is captured in the cleaning with post-combustion absorption [89, pp. 31-33]. The gas is cleaned of hydrogen sulphide before it undergoes *a CO-shift reaction*

$$CO + H_2O \rightarrow CO_2 + H_2 \qquad (5)$$

for better CO_2 recovery and is stripped of CO_2. In the BLGCC case the cleaned gas is burnt in a gas turbine for power production. In the BLG to DME case, the synthetic gas is refined in an energy consuming *transportation fuel synthesis* unit and distilled into transportation fuels, such as *dimethyl ether (DME)* and *methanol*. [86, pp. 21-25, 44; 89 pp. 31-33]

Additional biomass or other fuels relative to a conventional recovery boiler may be needed to compensate the steam demand of the BLG process [87, p. 8008]. BLG may also lead to greater lime kiln and causticizer loads because the chemical recovery is altered [90].

Carbon capture with BLG options shows great promise compared to carbon capture in a conventional recovery boiler in terms of economic feasibility [4, p. 1017]. The capture

potentials of BLG to DME and BLGCC are slightly different. In the BLGCC process all of the produced synthetic gas is stripped of CO_2 and contributes to emission reductions, while in the BLG to DME process fuel replaced by the motor fuel determines the total CO_2 reduction.

The economic performance of a BLG plant with carbon capture is mainly affected by its investment and operating costs, political instruments such as possible biofuel support, electricity price and fuel prices [4, pp. 1022-1023]. As with all emerging technologies with no full-scale plants operating, the investment cost is uncertain. In addition to that, the economics are very sensitive to the level of supporting policies [4, p. 1028].

4.2 Lime kiln options

In the following chapters the carbon capture options for the lime kiln are presented. As with the recovery boiler options, the order of presenting resembles an increasing amount of modifications required. The investigated technologies include *fuel switch, pre-calcination*, *air combustion with MEA absorption, oxy-enrichment with MEA absorption* and *oxy-fuel combustion.*

As opposed to the recovery boiler, the CO_2 from the lime kiln may originate from *both fossil and biogenic* sources. The fossil emissions of the lime kiln originate from the burnt fossil fuels and supplementary lime, which may originate from fossil sources. The main fraction of the carbon in the calcining reaction however originates from the black liquor extracted from wood chips and is therefore biogenic. The current policies favour carbon capture from the lime kiln, since only fossil CO_2 is included in the EU ETS.

It is significant to notice that the carbon flow through the lime kiln is about an order of magnitude smaller than the flow through the recovery boiler. This means that options with large investments that benefit from the economy of size may be less suitable for lime kilns than recovery boilers or both units, as might for example be the case of oxy-fuel combustion.

4.2.1 Fuel switch in lime kiln

Switching from fossil to biogenic or less carbon-intensive fuels is a common method for reducing the carbon footprint of pulp mills. In this thesis its costs and the decrease in carbon footprint are used as a reference for the carbon capture methods.

The change from oil to natural gas or biofuels and its potential has been examined in general by Möllersten et al. [91, pp. 696, 700]. Possible liquid biofuel substitutes have been summarized by Ikonen [92, pp. 16-23], including vegetable oils, methanol, turpentine, terpene residue, distilled tall oil fractions and animal fat derivatives. Earlier, Pusa and Salin [93] have studied the usage of solid fuels like bark, wood residues and peat.

The magnitude of carbon emissions reduction depends on the amount of fossil carbon in the alternative fuel and the amount of fossil fuel being replaced. In this thesis the fuel switch **from natural gas to lignin** was evaluated. Another common alternative fuel would be gasified biomass.

Alternative fuels are already in use in some lime kilns today. According to a questionnaire by Francey et al. [94] and cited by Ikonen [92, p. 13], 16 out of 67 lime kilns in nine countries around the world used other fuels than oil in 2011. Solid fuels can be utilized in lime kilns by two means through either pulverisation or gasification. Gasification has the benefit of separating most of the problematic substances, like aluminium and silicon, in the process so that also bark can be utilized. For pulverisation however, Pusa and Salin recommend to use only bark-free fuels. As long as the fuel is dried to approximately 85-90 wt-% dry content, all fossil fuel oil used in the lime kiln could be replaced. [93] Co-firing is more common than full replacement and according to Ikonen [92, pp. 17-19] typical proportions range from 10-15 % of the total heat consumption of the kiln.

Requirements for alternative fuel use depend on the properties of the new fuel. Some fuels, like bark, need to be gasified prior to combustion. Other fuels, such as bio-oils, may be easier to apply directly because of their similar properties to heavy fuel oil [92 p. 27]. The cost of fuel switch is dependent on the possible investment of new equipment in addition to fuel price. In this thesis it was **assumed that the related investment costs were small** compared to fuel costs.

4.2.2 Pre-calcination before lime kiln

According to Tsupari [95], a concept of indirect *pre-calcination before the lime kiln* is currently under study and the preliminary data used in this thesis is based on his yet unpublished results. The purpose is to create small amounts of highly concentrated CO_2 for on-site utilization, not storage. A fraction of the lime mud is separated before the kiln and heated to around 700-900 °C indirectly, without exposure to ambient air, in order to prevent dilution.

An overview of the process is presented below in Figure 13.
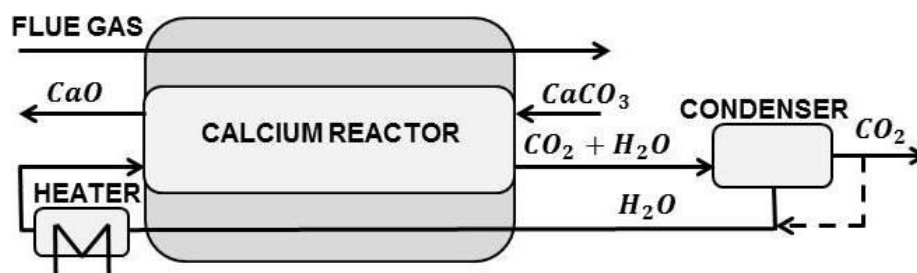
Figure 13.   *Overview of pre-calcination before the lime kiln. [95]*

Flue gas from the lime kiln provides part of the heat for the calcining reaction in the reactor and additional heat is provided with a separate heater. Water evaporates from the lime mud, but additional steam may be used to control the reaction temperature. Once the steam is condensed, a concentrated stream of CO_2 is formed. Possibly some of the CO_2 could be recycled after the condenser back to the calcium reactor via the heater. [95]

The technology is still under development as possible heating options are studied and preliminary experiments are run in a bench scale reactor. Considered heat sources include oxy-fuel combustion, indirect heat transfer from another reactor and electric heaters.

A similar system was proposed for CCS from cement lime kilns in 2008 by Rodríguez et al. [96]. The process includes two interconnected circulating fluidized bed reactors, one for burning the fuel and the other for calcination. The bed was suggested to be fluidized with CO_2 and possibly steam. This gives support to the idea of heating with another reactor. Also their preliminary calculations showed promise, as the cost for avoided CO_2 was as low as 19 \$/t(CO_2). Further analysis from the same research team [97] on the system has shown that around 50 % of the CO_2 emissions of the cement lime kiln could be avoided.

Implementation to a pulp mill lime kiln may differ from implementation to cement lime kilns regarding for instance, the scale of the process. Nevertheless, pre-calcination in pulp mill lime kilns could replace at least some of the CO_2 purchased to the mill making it financially sound.

4.2.3   Air combustion in lime kiln with MEA absorption

The MEA absorption process in general is described and illustrated earlier in Chapter 4.1.2, here only the flue gas source is the lime kiln. Change in the flue gas source results in different flow rates and concentrations. This would lead to different absorption plant sizes and investment costs. For instance, the CO_2 from the *calcining reaction (4)* leads

to higher initial CO_2 concentration in the flue gas and affects the capture break-even price, as shown later in this work in Chapter 6.2.1.

In short, the MEA absorption is commercially available and relatively easy to apply. The current political support is not enough to cover the CCS costs, which partly explains the absence of MEA implementations in pulp mill lime kilns.

4.2.4 Oxy-enrichment in lime kiln with MEA absorption

Oxy-enrichment in the lime kiln with MEA absorption functions according to the same principles as in the recovery boiler as described in Chapter 4.1.3. In brief, the combustion air for the lime kiln is enriched with oxygen and CO_2 is captured from the flue gas with MEA absorption. According to the *Best practise guidebook for pulp and paper industry* from year 2005 by *Focus on Energy* [98, p. 21], the oxy-enrichment part should not require large modifications in the lime kiln for a new pulp mill. However, the technology may not be applicable for existing kilns, or require greater investments [98, p. 21].

According to McCubbin [99], citing Garrido et al. [100], oxy-enrichment has been in use in pulp mill lime kilns since the 1970s. Despite that, the technology has remained relatively rare. In a conference held in 1981, Garrido et al. have mentioned that eleven pulp mills in North-America had adopted the technology. Presently, no up-to-date information on lime kilns utilizing oxy-enrichment was found. Moreover, the combination of oxy-enrichment and carbon capture in lime kilns is still under research, but the cement industry has shown promising initiative [101, p. 97; 102, p. 99; 103, p. 26].

Oxy-enrichment can be implemented to achieve savings by lowering fuel consumption by 7-12 % and to reach emission. A return on investment (ROI) of 35-60 % has been documented [99; 98, p. 21]. Combining oxy-enrichment with MEA absorption could offer a beneficial synergy; the flue gas from oxy-enriched combustion contains higher levels of CO_2 leading to lower MEA requirement in regenerating the absorbent. Any new pulp mill implementing MEA absorption for its lime kiln should also consider oxy-enrichment.

4.2.5 Oxy-fuel combustion in lime kiln

The principle of oxy-fuel combustion in the lime kiln is to replace combustion air feed with almost pure oxygen from an ASU and recycled flue gas. The flue gas is recirculated to lower the combustion temperature from that of pure oxygen combustion.

It is possible that the captured CO_2 from the lime kiln may require less cleaning with oxy-fuel combustion than with other technologies. At least preliminary studies in the *cement* lime kiln pre-calcination point to this direction [104; 105, pp. 209-211; 103, pp.

27-28]. This could lead to savings as the energy consumption of CO_2 treatment would decrease.

Oxy-fuel combustion in pulp mill lime kilns is in research stage, but oxy-fuel combustion has only recently been piloted in cement plants, according to a fresh information paper by the *IEA Greenhouse Gas R&D Programme (IEAGHG)* [104] citing Gimenez et al. [106]. In the pilot the aim was to achieve a high enough concentration of CO_2 for compression and transportation with the oxy-fuel equipment only. Whether the implementation to *pulp mill* lime kilns is possible, is yet unsure. The pilot has shown good performance constant operation to be possible and also the carbon break-even price to be lower than with air combustion with MEA absorption. Eriksson et al. predict similar effects for pulp mill lime kilns with a validated simulation model [105, pp. 211-214]. The pilot project has also shown that oxy-fuel combustion in lime kilns by retro-fitting – modifying existing equipment – may be possible, which was earlier [103, p. 29] considered to be most likely out of the question. Technical challenges were faced, including air leakage into the flue gas stream leading to more dilute CO_2 concentrations. This drawback was attributed to the small pilot plant size and avoidable in larger applications. The pilot has shown that the possibility of shifting the calcining reaction balance due to oxy-fuel combustion can be avoided with higher temperatures in the furnace.

Implementing oxy-fuel combustion in pulp mill lime kilns commercially would require more research. A new design of the lime kiln, an ASU and possibly also a physical separation unit for flue gas treatment could be needed. The operating costs would be greatly affected by this, as the ASU consumes plenty of energy.

4.3 Oxy-fuel combustion in both the recovery boiler and the lime kiln

It may be possible to take advantage of the economy of scale and increase CO_2 capture rate by implementing oxy-fuel combustion in both the recovery boiler and the lime kiln. This would mean that the same air separation unit would provide the oxygen for both combustion processes and that the CO_2 would be treated in the same unit. Otherwise the processes are assumed similar to oxy-fuel combustion in a single unit.

A general process diagram is presented in Figure 14.
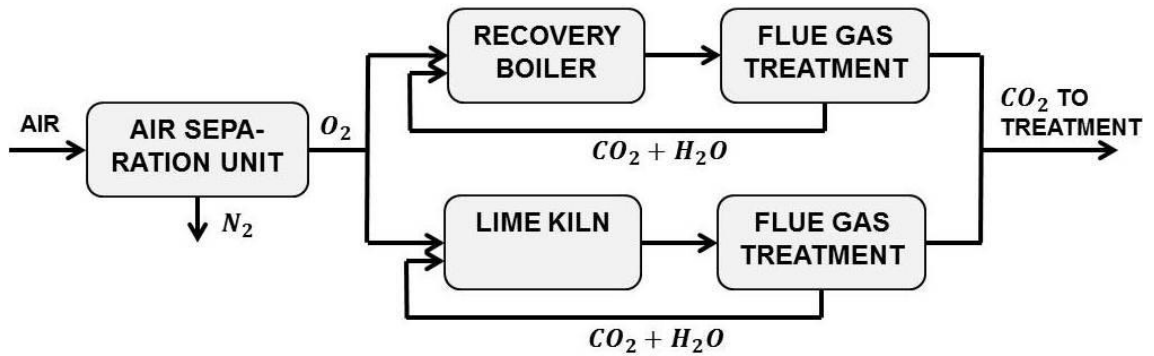
Figure 14.   *Oxy-fuel combustion in both the recovery boiler and the lime kiln.*

As described in Chapters 4.1.4 and 4.2.5, the CO_2 from the recovery boiler would likely need to be separated by physical separation. For the lime kiln, this operation might not be needed if a high enough purity of CO_2 is achieved with the calcining reaction and an optimal fuel choice.

It should be reminded that the amount of CO_2 from the recovery boiler is roughly an order of magnitude greater than that of the lime kiln. Hence the economic feasibility of oxy-fuel combustion in the recovery boiler dominates the economic feasibility of this option. Carbon capture from oxy-fuel combustion lime kiln and recovery boiler is still in an early research phase, as neither of these units has been demonstrated in oxy-fuel combustion in a pulp mill environment.

5. MODELLING OF OXY-FUEL COMBUSTION

The following cases were modelled in this study:

- Air combustion in the recovery boiler and the lime kiln (Case 0, reference)
- Oxy-fuel combustion in the recovery boiler (Case 1)
- Oxy-fuel combustion in the lime kiln (Case 2)
- Oxy-fuel combustion in both the recovery boiler and the lime kiln (Case 3).

All the cases were modelled with Balas software version 3.2.01. The program is developed at VTT for modelling *steady-state* chemical processes focusing on the pulp and paper industry. The model was based on mass and energy balances for which the software provided ready unit operations. [107]

The observations made from this model suitable for system level steady-state operation, such as overall efficiency and flow rates. This sufficed to assess the operating costs and separated CO_2 flows of the oxy-fuel options. The specific design of each unit process was not addressed; only large scale implications like reaction rates or heat losses were specified. The Balas software has previously been used for similar studies investigating energy use and CO_2 emissions in pulp and paper industry environment [108-111].

First the modelled system is presented in general in Chapter 5.1 and then case-specific modifications are given in Chapter 5.2. The outputs of each case are summarized and compared in Chapter 5.3.

5.1 Model overview

The model used in this thesis was built on an **unpublished pulp mill model used in previous studies by Kaijaluoto [112]**. Within this thesis, the existing model was supplemented with a *lime kiln* to form the reference air combustion model, as this part of the mill had not been of interest in the previous studies for which the model had been used. Then, this model was altered for each of the oxy-fuel cases. All of the modifications were done under the consultation of Kaijaluoto.

An overview of all the models used in this thesis is presented below in Figure 15.
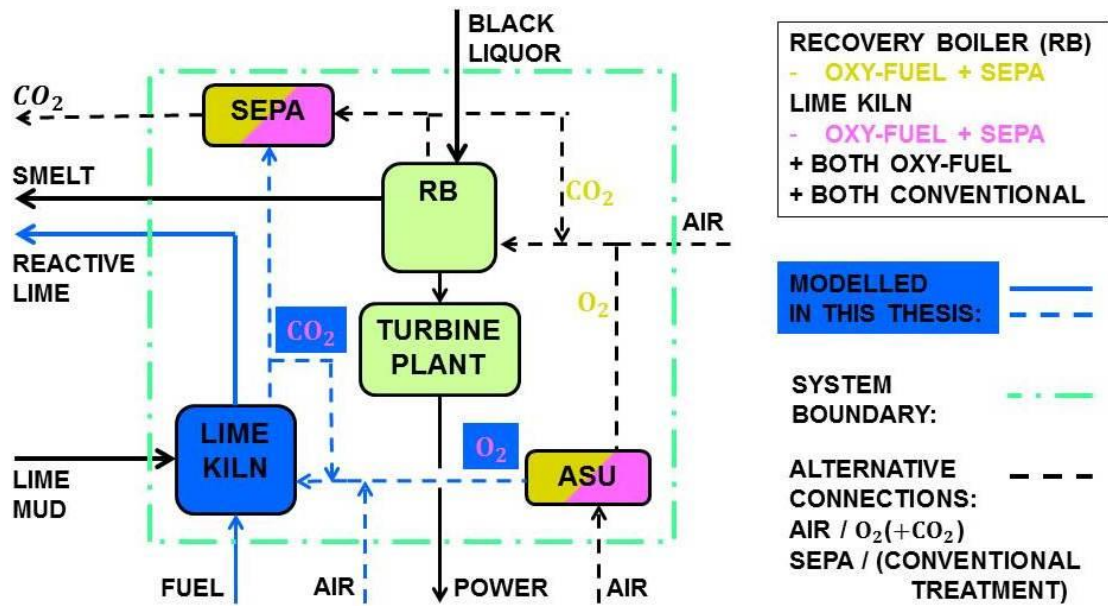
Figure 15.   *Combined overview of all the modelled cases.*

Blue units and flows were modelled within this thesis and the rest was adapted from the previous model. Dashed lines represent connections active in part of the cases and continuous lines the connections active in every case. The *recovery boiler*, the *lime kiln* and the *turbine plant* were active in all of the cases, whereas the *ASU* and the *carbon capture and treatment systems with physical separation unit (SEPA)* and their related connections were connected to the rest of the model only when necessary. The carbon capture and treatment procedure consisted of flue gas cooling, purification, compression, physical separation and CO_2 liquefaction, but for convenience all of this is represented by SEPA. For the air combustion case the physical separation was simply left out as in each of the cases other pollution control methods may still be needed. The system boundary is highlighted with the turquoise box.

The pulping process outside the system boundary was assumed to be unaffected. This means that the black liquor, smelt and lime flows were the same in each case. The black liquor flow **56.9 kg/s with 80 wt-% dry solids** was obtained from the previous VTT model which corresponds a pulp mill production volume of 2376 air-dry tons per day (ADt/d). Each of the modelled units is discussed more closely in the following subchapters with a strong emphasis on the lime kiln, which was modelled in this thesis.

5.1.1   Recovery boiler

The recovery boiler model obtained from the previous work at VTT consisted of the following main *calculation modules*: a *boiler* with typical recovery boiler reactions, one *air preheater* and one *superheater*. Regardless of the investigated case, the recovery

boiler was **assumed to maintain the same boiler feed water and reaction temperatures as well as same chemistry**.

When oxy-fuel combustion was applied the only change was that the oxygen was provided as a mixture of recycled flue gas and separated oxygen from the ASU. The flue gas was lead through an *electrostatic precipitator* and, depending on the case, into the carbon capture process or out of the stack.

The recovery boiler connects to the turbine plant, the ASU and the carbon capture process. In- and outputs that were used from this part of the model in economic calculations were *black liquor flow*, *flue gas flow*, *CO₂ content in flue gas* and *oxygen flow*. Moreover, the steam connections with the turbine plant affect the turbine shaft power.

5.1.2 Lime kiln

The lime kiln was the largest unit modelled within this thesis. According to Gullichsen and Fogelholm [48, p. 178] lime kilns are classified as *counter-current reactors* with *direct contact heat exchange*. Modelling the counter-current nature of the lime kiln with Balas induced certain difficulties. Firstly, the only option to have two material streams, the reacting lime and the combustion air, flowing in opposite directions and exchanging heat and reacting progressively was to discretize the process. Also the combustion of the lime kiln fuel was assumed to take place along the kiln. The model used in the air combustion case (Case 0) is presented as such in Appendix A and a list of main modules and process streams in Appendix B. The model consists of *two burner modules*, *two heat exchangers*, *one reactor*, and a lime mud *dryer*. Additionally the model structure led occasionally to diverging calculations in the solver, which were solved by setting constant values on some variables when *initializing* the model. Initializing here means simulation of the model smaller sections at a time, setting initial iteration values and − when necessary − setting some variables as constants. Eventually these restrictions were removed and a stable model was achieved.

The lime mud is first dried and non-reactive components are removed as waste. Then, the lime is heated in the first counter-current heat exchanger after which the calcium carbonate reacts according to the calcining reaction (4) to calcium oxide and CO₂ in an isothermal reactor. The resulting CO₂ is mixed with the combustion air whereas the calcium oxide preheats the combustion air in the second counter-current heat exchanger and continues to the surrounding chemical loop. The preheated air is lead to the first burner, which uses most of the fuel. The combustion gases are mixed with the CO₂ from the calcining reaction and provide the heat for the endothermic calcining of the calcium carbonate. Some fuel is used in the second burner to provide enough heat for increasing the temperature of the lime mud to the required level for the calcining reaction to take place. Finally the flue gas continues out of the stack in the air combustion case. Heat loss in the lime kiln is modelled with an additional cooler.

The model was verified by comparing some of the key parameters with literature values. These included *combustion gas temperatures*, *lime mud temperatures*, *lime mud dry solids, fuel consumption, heat loss, residual calcium carbonate* and *residual oxygen*. The comparison is presented in Table 2.

Table 2.   *Comparison of process values between the modelled lime kiln and literature as presented by Gullichsen and Fogelholm. [48, pp. 180, 187-188]*

Parameter	Unit	Model	Literature
Combustion gas, T – pre-heated	°C	226	250
Combustion gas, T – inside	°C	226 - 1690	250 - 1700
Combustion gas, T – out	°C	264	181
Lime mud, T – in	°C	74.8	60
Lime mud, T – inside	°C	74.8 - **800**	800 - 1100
Lime mud, T – out	°C	260	350
Lime mud dry solids, feed	wt-%	74	75
Fuel consumption per product	kJ/kg	**6175**	5548
Heat loss per CaO	kJ/kg	**272**	649
Residual $CaCO_3$	wt-%	**8.4**	2.0
Residual O_2	wt-%	2.1	2.0

The process values of the model matched adequately with the values given in literature. With the achieved quality of the model, the lime temperature in the kiln remained lower than in literature and the calcination reaction had a lower conversion rate of calcium carbonate leading to higher residual in the product. To better match the total energy consumption, a smaller heat loss was used to balance the higher fuel consumption presented in the literature. Better correspondence could have been achieved by discretizing the process further, so that heat transfer would be more evenly distributed. This however may have caused more instability problems in the already vulnerable model structure.

The lime kiln in the model was connected to the ASU and the SEPA. The lime mud flow **5.79 kg/s** was given as input based on the previous model. Process values used in follow-up calculations were *flue gas flow*, *flue gas composition, burner heat loads* and for the oxy-combustion cases the *oxygen flow*.

5.1.3 Air separation unit

The air separation unit model complied with the specifications presented by Dillon et al. in an IEAGHG publication [74, Appendix 'PFD 3']. The model consisted of a series of compressors and heat exchangers cooling the gas flow in between the compressors and a separation module. The heat could be used at the turbine plant in heating the boiler feed water if *heat integration* is considered possible, in this work only for new pulp mills. The modelled ASU used 0.77 MJ/kg (214 kWh/t) electricity for oxygen separa-

tion. However there has been improvement since the reference IEAGHG study in 2005 [74] and the most efficient ASUs in 2009 used only 0.58 MJ/kg (160 kWh/t) [113, p. 10] and a development target for 2015 was 0.52 MJ/kg (145 kWh/t). Thus the electricity consumption was afterwards **scaled to 0.55 MJ/kg** (153 kWh/t).

The ASU was connected to the recovery boiler, the lime kiln and, when heat integration was considered possible, to the turbine plant. Process values used in further calculations were *electricity use*, *oxygen flow* and *heat duty* from heat integration.

5.1.4 Physical separation and flue gas treatment

Also the carbon capture process model complied with the specifications presented by Dillon et al. [74, Appendices 'PFD 5 & 6']. The model consisted of two stages: *cooling and compression* and *physical separation and liquefaction*. The first stage consisted of a cleaning unit and a series of compressors and heat exchangers to produce a clean CO_2 flow in a pressure of 30 bar suitable for separation. The combined *compressor shaft power* and the *heat duty* were used in later calculations. In the second stage, CO_2 was separated physically from the compressed flue gas flow and the CO_2 liquefied with further compressors and flash tanks. The resulting temperature was -51 °C and pressure 6.5 bar, which are suitable for tanker ship transport as discussed earlier in Chapter 2.1.2. The CO_2-lean flue gas was used in a turbine to produce electricity.

The process values used in economic calculations were the *net power consumption* and *separated CO_2 flow*. The carbon capture sections were connected to the recovery boiler and the lime kiln. When heat integration was considered possible, a heat circuit connection to the turbine plant was established as well.

5.1.5 Turbine plant

The turbine plant model represented a typical pulp mill turbine plant and was **based on the unpublished pulp mill model from Kaijaluoto [112]**. The turbine plant consisted of five turbines with inlet pressures of 103 bar, 30 bar, 13 bar, 7 bar and 4 bar as well as a feed water tank. The model included three steam connections in between the turbines for recovery boiler feed water heating and heat integration circuits. The heat integration circuits were connected to the ASU and the carbon capture process. For follow-up calculations the *turbine shaft power* and the *combined heat duty* from the integration was used.

5.2 Modelled cases

Of all the thirteen studied technologies, only the oxy-fuel combustion cases were modelled in this study as the data for the other technologies was otherwise available.

The modelled cases and the active main units in each case are listed in Table 3.

Table 3. *Modelled cases and main units. X = unit is active in the case.*

Case	RB	LK	ASU	SEPA	TURBINE
Case 0: Air combustion (reference)	air	air	-	-	X
Case 1: 4. Oxy-fuel combustion in the recovery boiler	oxy	air	X	X	X
Case 2: 12. Oxy-fuel combustion in the lime kiln	air	oxy	X	X	X
Case 3: 13. Oxy-fuel combustion in both the recovery boiler and the lime kiln	oxy	oxy	X	X	X

The four modelled cases were based on the reference case of air combustion, which was then altered to oxy-fuel combustion in both the lime kiln and the recovery boiler. Oxy-fuel combustion in only one of the units was modified by detaching the oxy-fuel related connections from one unit at a time. This way the carbon capture and ASU sections had to be initialized only once, saving much work from dealing with the related model section stability issues. The changes back to air combustion were restricted to the lime kiln, the recovery boiler and the turbine plant.

In the cases where either the recovery boiler or the lime kiln was operating in oxy-fuel combustion, the ASU and carbon capture sections received no information of the change. Therefore the change in power consumption, O_2 and CO_2 flows were calculated afterwards. The energy demand of carbon capture and the emission reduction were calculated based on the CO_2 flow from either the lime kiln or the recovery boiler relative to the energy demand and emission reduction of both units. Similarly the energy demand for oxygen production was allocated to each unit based on their oxygen consumption.

The difference between rebuilt pulp mills and those without rebuild was not accounted for in the model, but in follow-up calculations by for example considering heat integration to be possible only for rebuilt mills. Similarly the ASU and the carbon capture processes were assumed to be the same regardless of the number of units functioning in oxy-fuel combustion. The differences in economic performance between the oxy-fuel options were calculated later based on the logic that *larger investments lead to lower investment costs per produced mass unit*.

5.2.1 Case 0: Air combustion

Case 0 was the *reference case*, where both the recovery boiler and the lime kiln combusted with air and no CO_2 was captured. Later, the energy consumption and the CO_2 emissions from Cases 1-3 were compared to this case.

The parameters affecting the net energy balance of this case consisted of the used *lime kiln fuel* and gained *turbine shaft power*. The black liquor flow was left out of the com-

parison, since it was set to remain the same in all four cases. The CO_2 emissions in this case were the combined emissions from the recovery boiler and the lime kiln and were calculated based on the *flow rate* and *CO_2 content* of the flue gases.

5.2.2   Case 1: Oxy-fuel combustion in the recovery boiler

In Case 1 an *oxy-fuel recovery boiler with carbon capture* was modelled and the lime kiln operated in air combustion. The chemistry in the recovery boiler was set to remain unchanged compared to air combustion; only the combustion air was changed to a mixture of separated oxygen and recycled flue gas. The similar chemistry requirement also determined the amount of flue gas recycled.

The effect of oxy-fuel combustion in the net energy balance of the modelled section of the pulp mill was seen in changed turbine shaft power. The energy balance benefitted from the lack of nitrogen, but the increased concentration of water in the combustion gas mixture decreased the efficiency because of the higher specific heat capacity. The net effect was transferred to the turbine plant along the steam connections. Heat integration with the ASU and the carbon capture provided a possible heat surplus usable at the mill, but the carbon capture heat integration was left out, because of its low temperatures. Emission reductions were achieved by connecting the carbon capture section to the model but still some CO_2 was emitted along with the waste gas.

5.2.3   Case 2: Oxy-fuel combustion in the lime kiln

For Case 2, oxy-fuel combustion in the lime kiln with carbon capture was modelled together with an air combustion recovery boiler. The temperature in the reactor was set to be the same in the *oxy-fuel lime kiln* as in the air combustion lime kiln. The combustion air was replaced with a mixture of oxygen from the ASU and recycled flue gas.

As with the recovery boiler, the energy balance was affected by the lack of nitrogen as well as the increased concentration of water in the combustion air mixture. For the lime kiln however the change in energy demand was seen in decreased fuel consumption. Emission reductions were achieved by separating CO_2 in the carbon capture section.

5.2.4   Case 3: Oxy-fuel combustion in both the recovery boiler and the lime kiln

In Case 3 both the recovery boiler and the lime kiln were modelled in oxy-fuel operation. The parameters were set as described above for Cases 1 and 2. Similarly, the effects of oxy-fuel combustion on energy balance and CO_2 emissions were based on the same principles.

5.3   Model outputs

For the economic calculations the model outputs from the reference mill as well as the data from previous literature were **scaled linearly to a chosen reference mill size of 1200 ADt/d**. Exceptions to this scaling procedure were the investment costs and MEA absorption energy consumption as explained later in Chapter 6. An *electricity balance* was calculated as the sum of power produced in the turbine plant and consumed by the ASU and the carbon capture units. The heat from process integration with the ASU was obtained as a separate flow from the model and its effect only accounted for in the economic calculations. The results from the model are listed in Table 4.

Table 4.   *Summarized model results.*

Unit	Output	Case 0 Air(both)	Case 1 OF-RB	Case 2 OF-LK	Case 3 OF(both)
General	Mill capacity [ADt/d]	2376	2376	2376	2376
	Black liquor flow [kg/s]	56.9	56.9	56.9	56.9
Recovery boiler	Flue gas flow [kg/s]	293.4	92.5	293.4	92.5
	Flue gas CO_2 content [wt-%]	18.4	58.1	18.4	58.1
	Flue gas O_2 content [wt-%]	4.4	4.4	4.4	4.4
	Flue gas N_2 content [wt-%]	65.5	1.9	65.5	1.9
	Flue gas H_2O content [wt-%]	11.8	35.6	11.8	35.6
	O_2 consumption [kg/s]	-	51.9	0.0	51.9
Lime kiln	Flue gas flow [kg/s]	9.9	9.9	4.7	4.7
	Flue gas CO_2 content [wt-%]	26.3	26.3	54.2	54.2
	Flue gas O_2 content [wt-%]	2.1	2.1	2.7	2.7
	Flue gas N_2 content [wt-%]	50.1	50.1	0.9	0.9
	Flue gas H_2O content [wt-%]	21.1	21.1	42.2	42.2
	O_2 consumption [kg/s]	-	-	1.3	1.3
	Burner 1 heat load [MW]	13.4	13.4	13.4	13.4
	Burner 2 heat load [MW]	2.5	2.5	1.9	1.9
Air separation unit	O_2 production [kg/s]	-	51.9	1.3	53.2
	Electricity consumption [MW]	-	40.1	1.0	41.1
	Heat from integration [MW]	-	20.5	0.5	21.0
Turbine plant	Power generation [MW]	121.0	129.7	121.0	129.7
Carbon capture	CO_2 captured [kg/s]	-	50.6	2.4	53.0
	Electricity consumption [MW]	-	20.1	1.0	21.1
Total	Electricity balance [MW]	+121.0	+69.5	+119.0	+67.5
	relative to *Case 0* [MW]	N/A	**-51.5**	**-2.0**	**-53.5**
	Lime kiln fuel (gas) [MW]	-15.9	-15.9	-15.3	-15.3
	relative to *Case 0* [MW]	N/A	**+/- 0.0**	**+0.6**	**+0.6**
	Heat gained [MW]	-	**+20.5**	**+0.5**	**+21.0**
	CO_2 captured [kg/s]	-	**50.6**	**2.4**	**53.0**

As can be seen from the model parameters table, introducing oxy-fuel combustion with carbon capture to the recovery boiler or the lime kiln resulted in higher energy con-

sumption. Although oxy-fuel combustion itself resulted in slight energy savings in the lime kiln and in the recovery boiler, the ASU and the carbon capture unit consumed even more energy than what was saved. It should be noted that savings in the conventional flue gas treatment could be achieved because of smaller flue gas volumes, but this was out of the scope of this thesis.

The scale of the oxy-fuel combustion process in the lime kiln was much smaller than in the recovery boiler. The captured CO_2 flow and the energy consumption of the recovery boiler option were about 20 times and 40 times, respectively, that of the lime kiln option. Thus in Case 3, oxy-fuel combustion in the lime kiln and in the recovery boiler, the lime kiln had only a minor role.

The CO_2 concentration in the flue gas increased with oxy-fuel combustion from 18.4 wt-% to 58.1 wt-% for the recovery boiler and from 26.3 wt-% to 54.2 wt-% for the lime kiln. The initial modelled concentrations seem reasonable – at least an example [48, pp. 102-104] of an air combustion recovery boiler flue gas results in 19.6 wt-% CO_2 and given the calcination reaction in the lime kiln the initial CO_2 concentration should be higher than that of the recovery boiler. The outputs from the modelled cases were used in economic calculations to assess the capture potential and break-even price of these technologies.

6. PROCEDURE FOR THE ECONOMIC CALCU-LATIONS

Part of the thesis was to compare results from literature to this thesis. For each technology option a *break-even price (BeP) for biogenic CO_2 emission allowance, excluding transportation and storage costs,* and a *capture potential by 2030* were calculated. The capture break-even price is defined as the relevant time scale was limited on one hand by growing uncertainties with longer scales and on the other by adequate reaction time for mill owners for project planning and initiation time with short scales. Two cost estimates were calculated:

- a *higher cost estimate* for the carbon capture process itself excluding any policy effects and other possibly not available income and
- a *lower cost estimate* including near future policies and other probable related sources of income.

Some of the studied carbon capture technologies require large modifications at the mill when implemented. These technologies were considered available only if the mill is rebuilt after its assumed technical lifetime of 40 years is exceeded, because then the implementation costs would be lower. To estimate the capture potential in all Finnish pulp mills, the technical age distribution of Finnish pulp mills was investigated. **Mills exceeding their technical life expectancy by 2030 were assumed to be rebuilt.** The statistics of the Finnish Forest Industries Federation [114] give reason to **assume that the total pulp production in Finland remains the same until 2030**. This information combined with production capacities and separated CO_2 fractions were used to estimate the capture potential of each technology.

The studied technology options are listed in Table 1 in Chapter 4. The data of the closely related technology options *2. Air combustion with MEA absorption in the recovery boiler* and *3. Oxy-enrichment with MEA absorption in the recovery boiler* as well as *10. Air combustion with MEA absorption in the lime kiln* and *11. Oxy-enrichment with MEA absorption in the lime kiln* were presented in the same columns, since the capture potential were the same and the break-even prices similar. In the columns of the combined technologies the higher cost estimate represents the technology without oxy-enrichment and the additional beneficial effects of oxy-enrichment were only included in the lower cost estimate. This is because some of the beneficial effects of oxy-enrichment, such as increase in pulp production, may depend on the plant configuration. The technology options *5. Chemical looping combustion in the recovery boiler* was left out, because the data was incomplete due to the immaturity of the technology.

Thirteen different technology options were studied in this thesis and each of them had its unique, but largely similar calculation procedure. An example of the full calculation procedure for the technology option *13. Oxy-fuel combustion for both the recovery boiler and the lime kiln* can be found in the Appendix C, because its procedure included all the most common steps.

6.1 Assumptions

Multiple assumptions were made regarding the reference mill, commodity costs and the effects of the carbon capture technologies. The assumed values of the most important variables are listed below in Table 5.

Table 5. *Summarized economic calculation variables.*

Variable name	Value	Unit	Source / Comment
Sold lignin price	450	€/t	[66, 115]
Liquid biofuel (DME) price	78	€/MWh	[4, pp. 1023-1024] 47-109 €/MWh
Purchased electricity price	45	€/MWh	[116], 5 year average
Lime kiln fuel (natural gas) price	40	€/MWh	[117] 5 year average
Value of heat from process integration	20	€/MWh	[118] produced from biomass
ASU electricity consumption: ref. (2012) vs. model (2005)	0,71	-	[113, p. 10]
Capital recovery factor (CRF)	0,20		[4, p. 1023]; 40 years, i = 20 %
Scale-up factor	0,70		[26, p. 3029]
LignoBoost investment cost	5,00	M€/t(lign)/h	[66] 5 M€/t/h
ASU investment reference cost	280,00	M€	[74, p. 5; 119] 315 M$ * $-€ = 280 M€
ASU reference production volume	124,00	kg(O2)/s	[74, p. 5] 10373 t(O2)/d = 124 kg/s
Oxy-fuel modifications - additional investment	0,65	-	[74, p. 5, 120 p. 59] 55-70%
Air-RB investment reference cost	0,2174	M€/(ADt/d)	[121] 250M€ for 1150 ADt/d
Air-LK investment reference cost	0,0365	M€/(ADt/d)	[122] 7 M€ for [123] 192 ADt/d
Liquid biofuel tax relief (DME from pulp residue)	19,04	€/MWh	[38] 0.1214 €/l; [124] 0.85 kg/L, 27 MJ/kg => 19,04 €/MWh
Investment support	20	%	[125] 25-70% * CRF; [126] max. 30 %
EU ETS: CO2 allowance	25	€/t(CO2)	[37, p. 180]
R&D funding	40	%	[125] 40-65%, [126] max. 40 %
Oxy-enrichment increased pulp production coefficient	0,15	-	[71, p. 2]
LignoBoost production increment	20	%	[66]
Currency exchange ($-€)	0,89	€/$	[127]
Production capacity in all rebuilt mills by 2030	5712	ADt/d	This thesis: mills aged 26 and up
Production capacity in all mills with no rebuild < 2030	15068	ADt/d	This thesis: mills aged 6-25
Mill production capacity (scaled to from 2396 ADt/d)	1200	ADt/d	
Pulp price	700	€/ADt	[128, p. 10] 5 year average
Operating hours per year	8000	h/a	
Lignin lower heating value	24400	MJ/t	[129, p. 57]
Natural gas lower heating value	47141	MJ/t	[130]
CO2 emissions per natural gas mass unit	2,8	kg/kg	[131]
Carbon in lignin	60	wt-%	[132, p. 121]
Fossil CO2 proportion in lime kiln emissions	0,338	-	[47, p. 734] 100 kg / (100+196) kg

The first values in the table are commodity costs and by-product values followed by parameters related to capital costs – especially the investment cost estimates for novel technologies have great uncertainties. In the middle and below, parameters of the political instruments and optional sources of income are listed. Variables regarding the reference mill can be found at the bottom. The public data on LignoBoost was supple-

mented to state of the art level using the in-house expertise at VTT. [66] A detailed description of the most important estimated values and their implications is presented in the following subchapters.

When considering suitable values for the variables listed in Table 5, several aspects had to be considered. The mill was assumed to be located that far away from the surrounding infrastructure with **no possibility of selling district heating**. For all higher cost estimates **the operation of the fibre line was assumed to remain unchanged**. Only for lignin separation and oxy-enrichment options an *increase in production capacity* was assumed in the lower cost estimate. The pulp production would be increased if a recovery boiler or a lime kiln was previously limiting the pulp production. For rebuilt mills a beneficial effect similar to the increased pulp production accounts for a smaller investment cost.

The purchased **electricity price was assumed to depend heavily on the market fluctuations,** and the *electricity transmission fee* and *electricity production tax* were assumed to be small in comparison. Moreover, the transmission fee may or may not concern the mill, because the pulp mills may either be net-producers or net-consumers of electricity. The applicability of the transmission fee depends on whether the pulp mill belongs to an integrated pulp and paper mill or not. The effect of the electricity production tax is small, because the mill is counted as *an industrial electricity consumer, class (II),* and thus subject to an *electricity production tax* of 7.03 €/MWh [133, p. 10] and the effect further decreased by the *tax refund* of up to 85 % for energy intensive industries [133, p. 16] to which pulp mills are categorized. The monthly averages of electricity have ranged from 20 €/MWh to 90 €/MWh during the past 5 years [116].
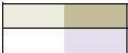
6.2 Break-even price

The break-even prices of the studied technologies were evaluated from different components depending on the technology in question. The results from the literature were recalculated to match the assumptions presented in Table 5.

Table 6 lists various effects that were calculated in this thesis, additional to the results in the literature and the commodity costs based model of used in this thesis.

Table 6. *Calculated additional effects for the studied carbon capture technology options.*
X = Effect calculated in this thesis.

	1. Liginin separation rebuilt	1. Liginin separation no rebuild	2&3. Air&OE-RB+MEA rebuilt	2&3. Air&OE-RB+MEA no rebuild	4. OF-RB+CRYO (CASE-1) rebuilt	6. BLGCC rebuilt	7. BLG to DME rebuilt	8. Fuel switch rebuilt	8. Fuel switch no rebuild	9. Pre-calcination rebuilt	9. Pre-calcination no rebuild	10&11. Air&OE-LK+MEA rebuilt	10&11. Air&OE-LK+MEA no rebuild	12. OF-LK+CRYO (CASE-2) rebuilt	13. OF-BOTH+CRYO (CASE-3) rebuilt
Size-scaled investment cost	X	X	X	X	X	-	-	-	-	-	-	X	X	X	X
Change in electricity costs	X	X	X	X	X	-**	-**	-	-	X	X	X	X	X	X
Change in lime kiln fuel costs	-	-	-	-	-	-	-	-	X	X	X	-	-	X	X
Previous policy effects subtraction	-	-	X	X	-	X	X	-	-	-	-	X	X	-	-
By-product value	X	X	-	-	-	-	X	-	-	-	-	-	-	-	-
EU ETS (fossil CO2 only)	-	-	-	-	-	-	-	X	X	-	-	X	X	X	X
Investment support	X	X	X	X	X	X	X	-	-	-	-	X	X	X	X
R&D funding	-	-	-	-	X	X	X	-	-	X	X	-	-	X	X
Increased pulp production capacity*	X	X	X	X	-	-	-	-	-	-	-	X	X	-	-
Oxy-enrichment investment costs	-	-	X	X	-	-	-	-	-	-	-	X	X	-	-
Biofuel tax support	-	-	-	-	-	-	X	-	-	-	-	-	-	-	-
Heat integration	-	-	-	-	X	-	-	-	-	-	-	-	-	X	X
No need for CO2 transport & storage	X	X	-	-	-	-	-	X	X	-	-	-	-	-	-

*For rebuilt mills as savings from smaller RB

** Included in results from previous literature

Included in both cost estimates

Included in the lower cost estimate

Rebuilt mills

Mills without rebuild

The change in electricity costs refers to the modification of the literature result based on the electricity price assumed in this thesis. The policy effects that were assumed in the results in the literature were subtracted and the assumed policy effects were added. The EU ETS was assumed to include only fossil CO_2 and therefore only a part of the lime kiln emissions was accounted for.

For the technology options modelled and calculated in this thesis the break-even price was evaluated by comparing to the reference. The *break-even price* was defined as *the CO_2 emission allowance price that is enough to cover the costs of applying a technology option, excluding the transportation and storage cost, and assuming that biogenic emissions are included in the EU ETS*. Exceptions to this are the lower estimates for the lime kiln options, where the current allowance price for fossil emission is taken into account as support. The commodity costs, investment costs and financial support from the included policies affected in the net revenue of the pulp mill. The difference in the revenues of a modified pulp mill and the reference mill was divided by the difference in CO_2 emissions.

The break-even price was calculated as

$$BeP \ [€/t(CO_2)] = \frac{Change \ in \ revenue \ [€/a]}{CO_2 \ studied \ mill \ [t/a] - \ CO_2 \ reference \ mill \ [t/a]} \ . \tag{6}$$

For the technologies other than oxy-fuel combustion or fuel switch, the BeP was only modified from previous studies to fulfill the assumptions of this work. The following subchapters explain how the effects listed in Table 6 were calculated.

6.2.1 Higher break-even price estimate

In the *higher cost estimate* for the break-even price it is assumed that no supporting policies exist and only the income from by-products and possible savings in commodity costs are included. Thus the break-even price is formed of the *investment cost* of the technology and the *commodity costs*. Possible by-product values were calculated like the commodity costs.

For the technology options calculated in this thesis, most of the *commodity costs* for the modelled cases, like the electricity price and fuel costs, as well as product values, like the sold biofuel, were calculated with the following principle:

$$Annual \ value \ [€/a] \\ = (New \ amount \ [t \ or \ MWh/a] - \ Reference \ amount \ [t \ or \ MWh/a]) \times Price \ [€/ \ t \ or \ MWh] \ , \tag{7}$$

where the annual value is the annual net effect of purchasing or selling a commodity, new amount is the amount used or produced in the modified mill and the reference amount is the amount used or produced in the reference mill. The costs, product values and policy effects of the technology options adapted from the literature were recalculated as

$$Annual \ value \ [€/a] \\ = Amount \ in \ literature \ [t \ or \ MWh/a] \ \times \frac{New \ price \ [€/ \ t \ or \ MWh]}{Price \ in \ literature \ [€/ \ t \ or \ MWh]} \ , \tag{8}$$

where the amount in literature is the amount of the commodity used according to the literature source in question, new price is the price or sum of the support assumed in Table 5 and price in literature is the commodity price used in the literature source in question.

To even out the fluctuations in the market prices, approximate 5 year averages were used for the pulp, electricity and natural gas prices. The estimated value of the separated lignin was a cautious estimate of 450 €/t, because the values presented earlier [66, 115] range widely from 400 to 900 €/t. This could be explained by the different qualities and uses of lignin either as a fuel or as a material.

The operating costs of MEA absorption were scaled according to the CO_2 concentration in the flue gas. According to Raynal et al. [134, p. 746] 83 % of the operating costs is assumed to be attributed to the regeneration of the absorbent. It was considered adequate to scale the operating costs based on the absorbent regeneration energy alone. The absorbent regeneration energy is dependent on the CO_2 concentration in the flue gas. Figure 16 illustrates the dependency of the regeneration energy from the CO_2 concentration based on the results of Notz et al. [24, pp. 86, 106].
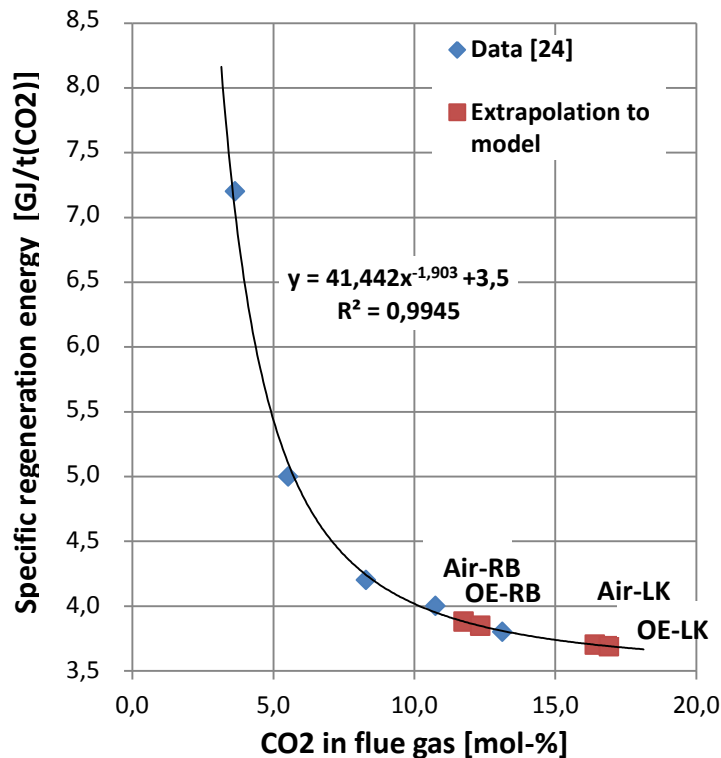
Figure 16.   *MEA absorption regeneration energy for the recovery boiler (RB) and the lime kiln (LK). [24, pp. 86, 106]*

The fitted function was used for estimating the operating costs in the MEA absorption options of this thesis. The CO_2 concentrations in the flue gases of the lime kiln and the recovery boiler were calculated based on the data from the used Balas-model. Finally the operating cost was calculated according to Equation 7.

The *capital recovery factor (CRF)* used to estimate the *annual capital costs [€/a]* is defined as [135]

$$CRF = \frac{i \times (1+i)^n}{(1+i)^{n-1}}$$

(9),

where *i* is the interest rate and *n* the number of annuities. In this thesis the CRF was set to 0.2 as Jönsson et al. have done in their conservative estimate in a similar study [4, p. 1023].

Then the annual capital cost was calculated as

$$Annual\ capital\ cost = CRF \times Investment\ cost \ . \tag{10}$$

The investment costs were modified according to the scale of the plant or technology unit. The scaling factor is defined by equation [26, p. 3029]

$$C = C_0 \times \left(\frac{S}{S_0}\right)^R , \tag{11}$$

where C is the investment cost [€] and S the size or volume [kg/s] in the studied mill, C_0 the investment cost [€] and S_0 the size or volume in a reference plant [kg/s] and R is a *scale-up factor,* in this thesis 0.7 based on a similar study by Hektor and Berntsson [26, p. 3029] as listed in the Table 5. The reference investment costs for oxy-fuel options were estimated by combining data from *oxy-fuel power plants* and *air-combustion lime kiln and recovery boiler investments*. The investment cost of a coal-fired oxy-fuel power plant is assumed to be around 65 % higher than that of a conventional power plant [74, p. 5; 120, p. 59] and the same proportional increase was evaluated to be the investment cost of oxy-fuel combustion modifications to the lime kiln or the recovery boiler.

6.2.2 Lower break-even price estimate

For the second set of calculations the effects of applicable current and expected near future policies were applied. In addition, some uncertain yet probable streams of income were included. This way a more affordable, but still realistic break-even price estimate was formed.

The effect of EU ETS on the pulp and paper industry is expected to be minimal. Minor financial support could be gained as income, when the fossil CO_2 emissions from the lime kiln are reduced. The current trading scheme excludes biogenic CO_2 and this approach was assumed to remain in the near future as well. According to the European Commission [37, p. 178] the amount of free allocations, given to companies free of charge, for the pulp and paper industry is estimated to be as much as 90 % from the required amount of emission allowances between 2021 and 2030. Free allocations are granted to avoid carbon leakage and to reward for already accomplished energy efficiency investments, as discussed in Chapter 2.3. The price of carbon emission allowance was assumed to be the same as in the European Commission Impact Assessment report, at 25 €/t(CO_2) [37, p. 180]. The income from EU ETS on fossil CO_2 affecting the break-even price of biogenic emission allowance was calculated as

$$Income\ from\ EU\ ETS\ [€/a] = CO_{2,fossil}[t(CO_2)/a] \times Allowance\ price\ [€/t(CO_2)] , \tag{12}$$

where the income from EU ETS is the annual granted support, $CO_{2,fossil}$ is the annual amount captured fossil CO_2 and allowance price is the assumed price of emission allowances.

Investment support and *research and development (R&D) funding* are offered to companies as investment grants, competitive loans and loan guarantees. According to Rikama [136] the largest public funding organizations are *the Ministry of Agriculture and Forestry*, *the Ministry of Employment and the Economy*, *the Finnish Funding Agency for Innovation (Tekes)* and *Finnvera.* The amount of the support was estimated based on the information from the Ministry of Employment and Economy [126] and Tekes [125]. Factors regulating the amount of the support are among others the company size, novelty of the project concerning the investment and project partners.

Typically the investment support from Tekes is offered as a highly competitive loan covering 25-70 % of the total project costs. The interest rate is currently 1.00 %. The interest rate would decrease the CRF from 0.2 to 0.045 and result in a support of 3.9-10.9 % of the total cost. For research projects, the Tekes funding varies between 40-65 % of the total project cost.

In 2015 the Ministry of Employment and Economy offered grants of up to 40 % for investments in novel renewable energy or energy efficiency technology innovations and up to 30 % for investments reducing the environmental impacts of energy production in general. In this thesis an investment support of 20 % was used for the mature parts of the technologies and R&D funding of 40 % for the parts of technologies under research. Technology options considered applicable for each support instrument were listed in Table 6. The ASU is part of many of the technology options, but it was considered a mature technology and its investment was assumed to be funded by the investment support, not the R&D funding.

Increased pulp production capacity was attributed to lignin separation and oxy-enrichment options. The increase was based on the assumption, that the recovery boiler or the lime kiln limits the total production. To evaluate the benefits of this increase in pulp production, a five-year average pulp price of 700 €/t was estimated based on the data of RISI [128, p. 10]. Of this value, around 25 % could be *realizable income* [66], as other operating costs increase with the pulp production increase. The benefits from the production increase were calculated as

$$
\begin{aligned}
&\textit{Additional income } [\text{€}/a] = \\
&\textit{Pulp production } [t/a] \times \textit{Production increment } [\%] \times \textit{Pulp price } [\text{€}/t] \times \\
&\textit{Realizable income } [\%],
\end{aligned} \tag{13}
$$

where the additional income is the income from the increased pulp production, pulp production is the pulp production of the reference mill, production increment is the achieved increase in production relative to the reference case, pulp price is the assumed

price for the sold pulp and realizable income is the realizable proportion after taking the increased operating costs into account.

Additional investment costs were attributed to oxy-enrichment with MEA absorption. These options were presented in the same columns with air combustion with MEA absorption. Thus the investment in ASU's and their integration were considered additional *compared to air combustion.* The more valuable increase in pulp production led to lower total break-even prices and therefore the cost estimates including oxy-enrichment were the *lower cost estimates* in spite of the additional investments. The ASU investment for the modelled recovery boiler was scaled with Equation 11.

Biofuel tax support was related only to the BLG to DME option. A production tax is collected by the Finnish government for energy carriers and electricity [137]. The tax for biodiesel oil is 40.63 cent(€)/l, but a tax relief of 12.14 cent(€)/l or 37 % is offered if the fuel is produced sustainably using non-edible biomass [38], which is assumed to be the case for the studied pulp mills. This corresponds to 19.04 €/MWh, as the density of biodiesel is 0.85 kg/l and the lower heating value 27 MJ/kg [124, pp. 3-6]. Thus the annual tax compensation was calculated as

$$Annual\ tax\ support\ [€/a] = Biofuel\ produced\ [MWh/a] \times Tax\ relief\ [€/MWh]\,, \qquad (14)$$

where annual tax support is the annually granted support, biofuel produced is the amount of produced biofuel (DME) and the tax relief is the decrease of taxation per energy unit.

Heat integration with the ASU and the carbon capture unit was considered possible for the lower cost estimates only, because it may not be applicable to all plant configurations. The value of the process heat estimated based on the price of purchased biomass, because an increase in heat consumption would likely lead to increased combustion of biomass. The related savings were calculated for the modelled cases according to Equation 6.

Based on the data from Teir et al. [5, pp. 44-48], the costs for transportation and storage were estimated to be 16 €/t(CO2) and 11.5 €/t(CO2), respectively. These represent the costs of transportation from an inland pulp mill to a storage site in a depleted fossil fuel formation. For the technology options *1. Lignin separation* and *8. Fuel switch* no transportation or storage is required and therefore they have an advantage compared to the other options. The savings were then calculated according to Equation 7.

6.3   Capture potential

The *carbon capture* potential was defined as *the annual amount of CO₂ emissions captured, or fossil emissions avoided in the cases of lignin separation and fuel switch, if the technology is implemented in all applicable mills in Finland*. The factors affecting the

capture potential were the carbon separation rate, applicability in mills without rebuilding the mill and whether the capture method is applied in the lime kiln or the recovery boiler. The carbon separation rate for MEA absorption and physical separation is typically set to around 90 % of the entire CO_2 flow. The process of pre-calcination is different and the carbon separation rate is smaller. In lignin separation and fuel switch, the avoided fossil emission is based on replacing fossil fuels with biofuels by the same energy content. Since some of the technologies require major changes at the mill, these technologies were considered applicable only for pulp mills after major scheduled rebuilding when the *technical life expectancy* of the mill is exceeded. To assess the capture potential it was necessary to find out how many mills exceed their life expectancy by the given time scope, by 2030, and the associated pulp production capacities. The capture potential was calculated as

$Capture\ potential\ [t(CO_2)/a]$

$= Carbon\ flow\ [t(CO_2)/a] \times Capture\ fraction\ [\%]\ \times Applicable\ mills\ [\%]$,   (15)

where the capture potential is the amount of CO_2 captured annually if the technology is implemented in all possible Finnish pulp mills, carbon flow is the annual CO_2 flow in the flue gas, capture fraction is the percentage of CO_2 captured and applicable mills is the relation of the pulp production volume of all the applicable mills and the pulp production volume of the reference mill.

According to Magnusson [138, p. 13] quoting Ahlroth [139] the technical life expectancy of a Kraft pulp mill is around 30-40 years. In Finland no pulp mills over the technical age of 40 years were found and five mills had reached the age of 30 years. In this thesis it was **assumed that the technical life expectancy or its chemical recovery section is 40 years**. The technical ages of the pulp mills were estimated with focus on the chemical recovery section of the mill. The age was determined by the following parameters, here listed in decreasing order of priority:

- the year when the recovery boiler was replaced
- the year when major rebuilding took place around the mill or
- the year when the mill was built.

The technical age distribution of the Finnish pulp mills is presented in Figure 17.
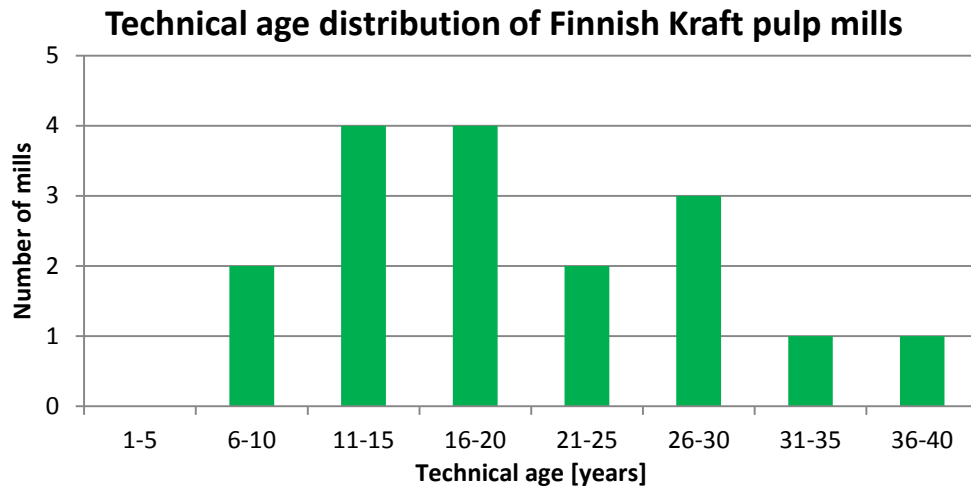
Figure 17.   *Technical age distribution of Finnish Kraft pulp mills. [142-155]*

There were fifteen Kraft pulp mills in total of which five exceeded the technical age of 25 years. Most mills were aged 10-20 years at the time of the study. The age estimates of 7 Stora Enso mills were based only on less credible sources [146, 148-152].

All mills exceeding the life expectancy of 40 years by 2030, or aged 25 or more in 2015, were assumed to be rebuilt. Their **pulp production capacities were assumed to remain the same** after the rebuild as was the total production capacity in Finland. To calculate the percentage of applicable mills in Equation 15, the total production capacity of pulp mills aged up to 25 years was used for technologies applicable in mills without rebuild. The total production capacity of mills over 25 years was used for technologies applicable only in rebuilt mills. The full data of the technical age distribution and the related pulp production capacities can be found in Appendix D.

7. RESULTS

The studied technology options for carbon capture in pulp mills assessed in this thesis were as well as the method for estimating the break-even prices and capture potentials are summarized in Table 7.

Table 7. *Summarized break-even price and capture potential estimation methods.*

Carbon capture technology	Break-even price and capture potential estimation method
1. Lignin separation before the recovery boiler	Previous results recalculated
2. Air combustion in the recovery boiler with MEA-absorption (Air-RB+MEA)	Previous results recalculated
3. Oxy-enrichment in the recovery boiler with MEA-absorption (OE-RB+MEA)	Previous results recalculated
4. Oxy-fuel combustion in the recovery boiler with physical separation (OF-RB+SEPA)	Balas model based calculations
5. Chemical looping combustion in the recovery boiler (CLC-RB)	(excluded from detailed analysis)
6. Black liquor gasification with combined cycle (BLGCC)	Previous results recalculated
7. Black liquor gasification with motor fuels production (BLG to DME)	Previous results recalculated
8. Fuel switch in the lime kiln	Mass and energy balance based calculations
9. Pre-calcination before the lime kiln	Previous results recalculated
10. Air combustion in the lime kiln with MEA-absorption (Air-LK+MEA)	Previous results recalculated
11. Oxy-enrichment in the lime kiln with MEA-absorption (OE-LK+MEA)	Previous results recalculated
12. Oxy-fuel combustion in the lime kiln with physical separation (OF-LK+SEPA)	Balas model based calculations
13. Oxy-fuel combustion in both the recovery boiler and the lime kiln (OF-LK&RB)	Balas model based calculations

The results for the *technical maturity* of the different options are presented in Chapter 7.1. The combined results for *the CO_2 break-even price* and the *capture potential* are presented in Chapter 7.2. Finally a *sensitivity analysis* considering the oxy-fuel options and the fuel switch is presented in Chapter 7.3.

7.1 Technical maturity

The *technical maturity* of a technology was understood as *how likely a technology is to be ready for implementation in the near future*. The time scale was in this thesis set to the year 2030. The technologies were then categorized on a scale from *research* to *pilot* and *commercial*.

The estimates for the technical maturity were based on the number of studies and pilot plants found in the literature and information on commercially available technology. Technologies that were not found to have been piloted yet and where the data was scarce or only described in another context than a pulp mill were classified as being in the *research* end of the scale. Technologies that have been piloted in the pulp mill context or in numerous other contexts were evaluated to be on *pilot* stage or higher. Technologies that were known to already be in industrial use were classified as *commercial*. The results of the technical maturity are presented in Figure 18.

Figure 18.   *Technical maturity of the studied technologies. See the beginning of this chapter for abbreviations.*

The diagram illustrates the wide range of the technical maturity of the studied technologies. This was later reflected in the difficulty to *reasonably* assess the investment costs. The technology considered most immature, *5. Chemical-looping combustion in the recovery boiler*, was left out of the economic calculations because of the lack of reliable data. The technology options *4, 12* and *13,* which were also modelled in this thesis, were found to be rather novel. Technologies estimated to be already or soon ready for implementation included *the MEA absorption options*, *lignin separation* and the *fuel switch*. In addition, *black liquor gasification* and *pre-calcination* technologies would probably be at least ready for large-scale demonstration facilities by 2030, because information on piloting was available.

7.2 Break-even price and capture potential

The maximum capture potential in Finland was determined by the total production capacity in Kraft pulp mills. The total production capacity in mills over the technical age of 25 years was 5712 ADt/d and 15 068 ADt/d in newer mills. The calculations can be found in Appendix D. The amount of captured CO_2 in the scaled reference mill size of 1200 ADt/d was scaled according to the relevant total production capacity in all the possible pulp mills.

The break-even price was evaluated by comparing the technology option in question with the reference mill based on changes in revenue (technology options 4, 8, 12 and 13) or by modifying previous results (technology options 1, 2, 3, 6, 7, 9 and 10) to meet the assumptions used in this thesis, such as commodity costs and investment cost parameters. The evaluated break-even prices excluded transportation and storage costs. The resulting break-even prices and capture potentials are summarized in Table 8.

Table 8. *Break-even prices and capture potentials.*

		Capture potential in rebuilt mills [Mt(CO2)/a]	Capture potential in mills without rebuild [Mt(CO2)/a]	Higher BeP for rebuilt mills [€/t(CO2)]	Lower BeP for rebuilt mills [€/t(CO2)]	Higher BeP for mills with-out rebuild [€/t(CO2)]	Lower BeP for mills with-out rebuild [€/t(CO2)]
Recovery boiler	**1. Lignin separation**	0,40	1,05	-48,50	-197,26	-48,50	-197,26
	2./3. Air/OE & MEA	3,17	8,36	79,25	72,98	79,25	72,98
	4. Oxy-fuel & SEPA	3,50	0,00	81,08	51,00	0,00	0,00
	6. BLGCC	1,69	0,00	144,37	103,56	0,00	0,00
	7. BLG to DME	0,82	0,00	42,07	-98,42	0,00	0,00
Lime kiln	**8. Fuel switch**	0,05	0,13	161,11	108,61	161,11	108,61
	9. Pre-calcination	0,07	0,18	7,30	4,50	7,30	4,50
	10./11. Air/OE & MEA	0,16	0,43	240,01	192,67	240,01	192,67
	12. Oxy-fuel & SEPA	0,17	0,00	139,66	84,79	0,00	0,00
Both units	**13. Oxy-fuel in both**	3,67	0,00	82,34	56,32	0,00	0,00

The results of the break-even prices and capture potentials are also illustrated in two diagrams.

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First, the results for the recovery boiler options as well as oxy-fuel combustion in the recovery boiler and the lime kiln are illustrated in Figure 19.
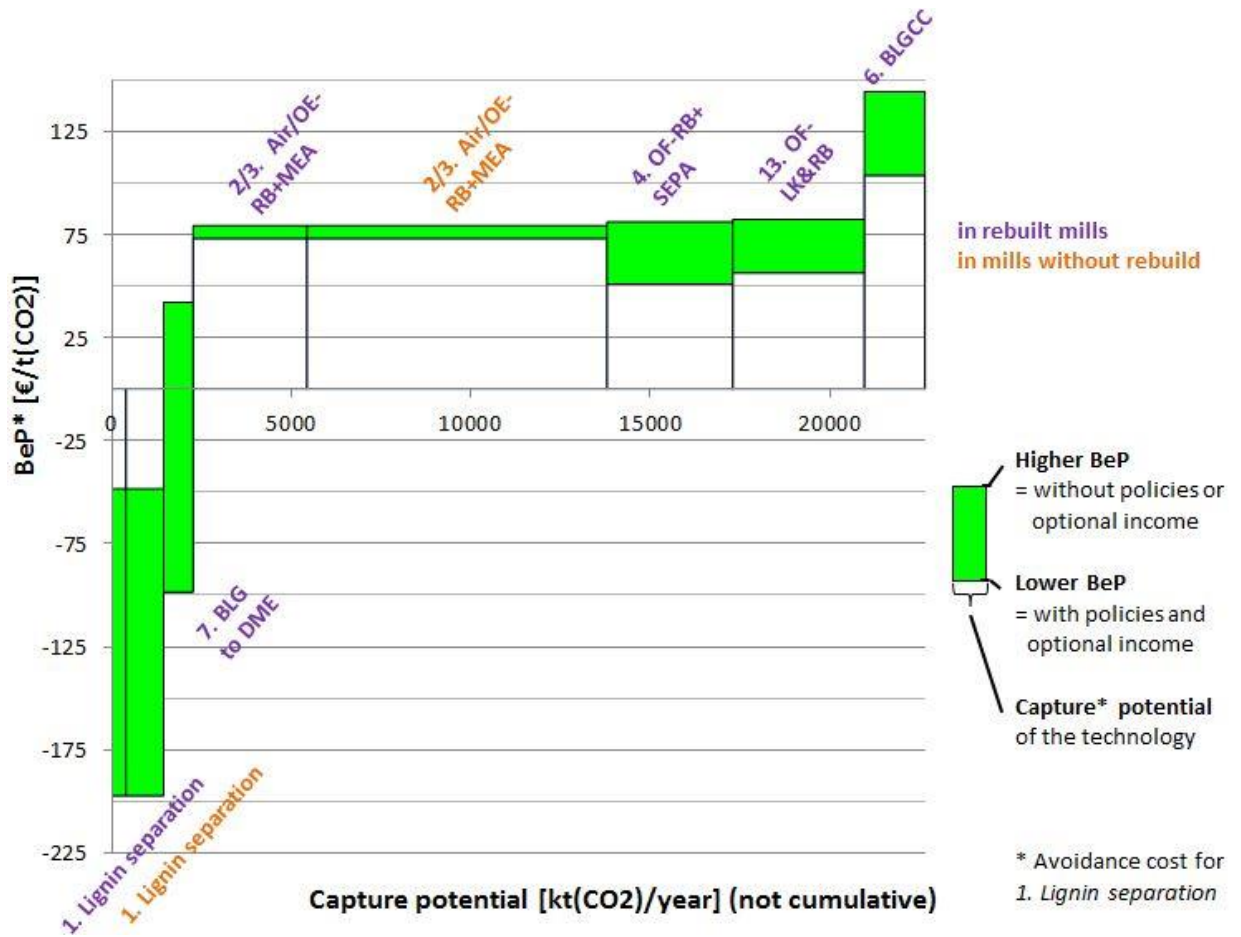
Figure 19. *Break-even prices and capture potentials of the recovery boiler options. Technologies 4 and 13 were modelled in this thesis.*

The corresponding results for the lime kiln options were separated into its own diagram because of the smaller scale of capture potential.

The results for the lime kiln options are illustrated in Figure 20.
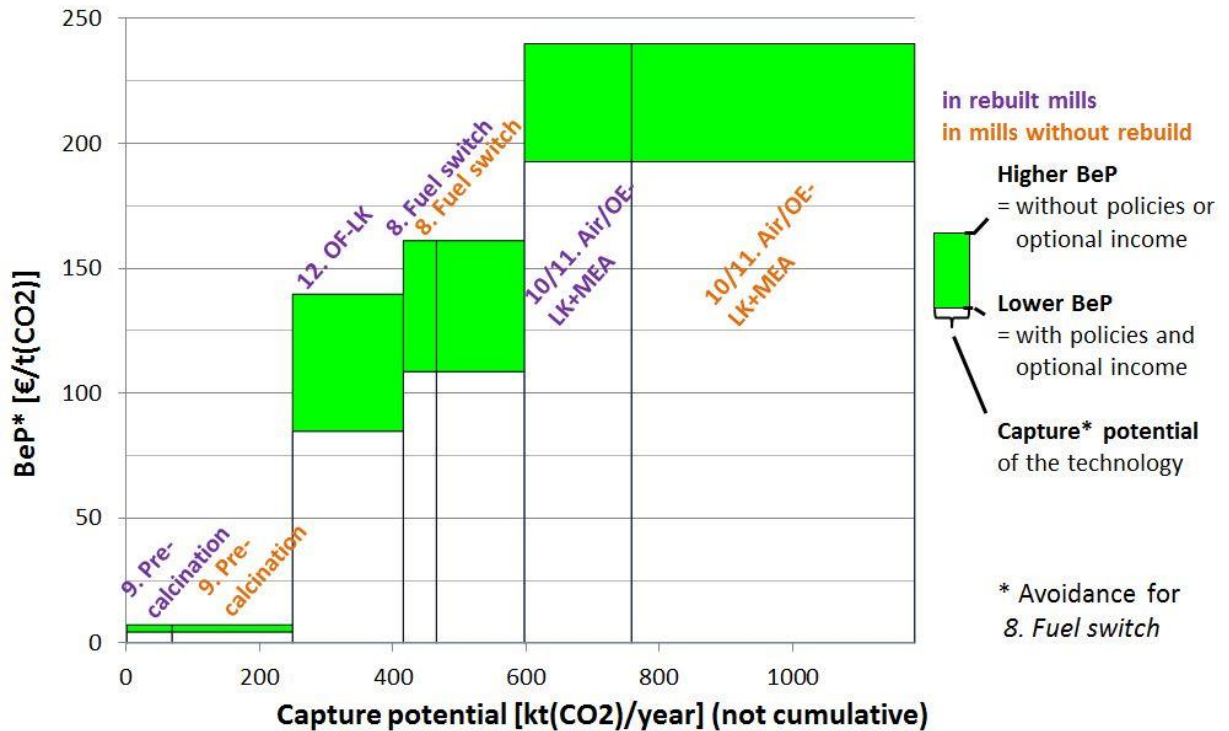
Figure 20. *Break-even prices and capture potentials of the lime kiln options. Technologies 8 and 12 were modelled and calculated in this thesis.*

In the result diagrams, the *capture potential* of a given technology is represented by the width of the column. This is the *total annual amount of CO_2 that is captured (or avoided in the cases of lignin separation and fuel switch) if the technology is applied in all possible Finnish pulp mills.* It should be noted that the capture potentials are not cumulative; implementing pre-calcination would lower the capture potential of MEA absorption, for instance.

The *higher break-even price estimate* excludes any policy effects and possibly unavailable revenue; this is the cost that has to be met by means of supporting policies, increased production or other revenues, if a constrain of unchanged total revenue is applied. For instance, if the EU ETS *including* biogenic CO_2 were the only available revenue in addition to possible savings in commodity costs, the emission allowance price should be greater or equal to the higher break-even price for the technology option to be economically feasible.

The *lower cost estimate* includes all probable revenues and support. The *air combustion options with MEA absorption (2. and 10.)* and their correspondent *oxy-enrichment options (3. and 11.)* were presented in the same columns, where the benefits of oxy-enrichment were included in the lower cost estimate.

The *higher break-even prices* of the technologies with large capture potentials were around 80 €/t(CO_2). Within all the studied technology options the range was much broader: from −49 €/t(CO_2) of the *lignin separation* to around 240 €/t(CO_2) of MEA absorption for the lime kiln. The *lower cost estimates* also ranged widely, from −200 €/t(CO_2) to a bit under 200 €/t(CO_2). MEA absorption was economically least feasible due to the high energy penalty and the large specific investment costs resulting from the small scale of the lime kiln relative to the recovery boiler. Also in general the lime kiln options were more costly due to the higher specific investment costs.

The recovery boiler options including a by-product, lignin or a liquid biofuel, were more economically feasible and could even become profitable. The possible profitability can be seen in the negative break-even prices of the lignin separation and BLG to DME options. The possible profitability of the BLG to DME option depends on the supporting policies, but currently the investment and biofuel tax supports are enough to cover the energy penalty and the investment costs. The probable profitability of lignin separation is also dependent on the investment support, but more importantly dependent on the lignin and pulp prices, which currently result in negative break-even price even without supporting policies. The large difference in the higher and lower cost estimates of lignin separation was due to the possibility of increased pulp production. For the other options the main differences were due to investment grants and the EU ETS excluding biogenic CO_2.

In order to evaluate the costs of the complete CCS chain, the transportation and storage costs of around 27.5 €/t(CO_2), as presented in Chapter 6.2.2, should be added to the break-even prices in Figures 19 and 20. The *lignin separation* and the *fuel switch* require no CO_2 transportation and storage and to demonstrate this advantage, the transportation and storage costs, which other technologies *would* require, were subtracted from the lower cost estimates of lignin separation and fuel switch.

The MEA absorption options had the highest capture potentials totaling around 12 Mt(CO_2)/a of which around 11.5 Mt/a was from the recovery boilers. This corresponds to about 70 % of the theoretical bio-CCS potential in the Finnish pulp mills as presented in Chapter 3. The remaining 30 % consists of the waste gas that is not captured; a capture fraction of 100 % would be less economically feasible, liquid wastes, other emission sources at the mill like the multi fuel boiler, by-products, the small share of mechanical pulping where these capture methods cannot be applied and error margin in the calculations. The large capture potential of MEA absorption was mainly due to the readiness of the technology and thus it was considered also available for pulp mills without rebuilding the existing plant.

The lime kiln options had the lowest capture potentials, as would be expected based on the volume of the carbon flow through the kiln. Oxy-fuel combustion in the recovery boiler had a decent potential of 3.5 Mt(CO_2)/a, although significantly limited by its

applicability only in rebuilt mills. The lignin separation and BLG to DME options had quite high capture potentials as well, 1.5 Mt(CO_2)/a and 0.8 Mt(CO_2)/a, respectively.

7.3 Sensitivity analysis

The purpose of this thesis was to assess the feasibility of carbon capture technologies in Finnish Kraft pulp mills. The studied aspects of feasibility were technical maturity, capture potential and break-even price. The related uncertainties were numerous ranging from global economy to politics and individual process parameters. The time scale was as long as 15 years. Some of the most important factors affecting the results are listed by the aspect of feasibility they affect in Table 9.

Table 9.   *Uncertainty factors related to feasibility of carbon capture in pulp mills.*

Aspect of feasibility	Uncertainty factor
Technical maturity	Unpublished research
	Non-discovered flaws in emerging technologies
	Adaptability of cement lime kiln carbon capture technologies
Capture potential	Modelled CO_2 emissions used in estimates
	Changes in the total pulp production capacity in Finland
	Technical limitations in adapting a technology in an existing mill
Break-even price	Changes in investment costs
	Changes in volumes of supporting policies
	Applicability for supporting policies
	New policies
	Changes in commodity costs
	Technological development
	Amount of possible production increment
	Effects on conventional flue gas treatment
	Installation costs for existing pulp mills

One of the most important uncertainties regarding the technical maturity was the unpublished research done in companies; technologies might get surprising evidence from unrevealed pilot studies. Technical challenges may also occur when implementing technologies from other fields of industry.

The CO_2 capture potential would be affected by the changes in the total pulp production of Finland. New mills may be built or existing mills shut down or developed for higher capacities. There may also be non-discovered technical incompatibilities that prevent some of the technologies from being used at certain mills. Also the CO_2 emissions per produced ton of pulp may vary from the reference mill.

Perhaps the most complex group of factors influencing the break-even price of CO_2 was all the policies affecting the CCS chain. In addition to the changes in the volumes of

financial support, regulations may indirectly lead to beneficial or adverse effects. For instance the *green certificate* for electricity production was not accounted for in this study. A great increase in the demand for certified green electricity might lead to a more affordable case for BLGCC. Future policies would seem to be in favor of biomass use for energy production, as according to the parliamentary Energy and Climate Roadmap 2050 [2, p. 32] the use of biomass for heat, electricity and liquid biofuels would be *tripled* from 15.3 TWh in 2012 to targeted 37-53 TWh by 2030. Because of the uncertainties in supporting policies, the calculations included both a cost estimate with and without policies.

Further uncertainty factors influencing the break-even price were the investment costs, commodity costs and technical development. These factors were taken into account in a *numerical sensitivity analysis*. The accuracy of the investment cost estimate differed much from one technology option to another. For established technologies like the MEA absorption the investment was easier to estimate, but for some technologies even demonstration facilities were lacking and the investment cost had to be estimated by other less accurate means. For example the investment cost of an oxy-fuel recovery boiler was estimated by using the data from an investment in an air combustion recovery boiler [121] and the relative increment in investment costs from oxy-fuel power plants compared to conventional power plants [74, p. 5; 120 p. 59]. Market deviations may affect the break-even price through changes in the interest rate and commodity costs. Another significant factor was the technical development, which may lead for instance to lower electricity consumption. The single most important electricity consumer in these technologies was the ASU and therefore its development was considered most significant.

The uncertainties affecting the break-even price were examined more closely with *a numerical sensitivity analysis*. The analysis was performed for the technology options modelled and calculated in this thesis, including:

4. Oxy-fuel combustion in the recovery boiler,
8. Fuel switch in the lime kiln,
12. Oxy-fuel combustion in the lime kiln and
13. Oxy-fuel combustion in both the recovery boiler and the lime kiln.

For the reasons presented above, the *numerical sensitivity analysis* focused on the major break-even price parameters other than policy instruments, which were accounted for in the higher break-even price estimates in Figures 19 and 20.

The parameters included in the numerical sensitivity analysis were:

- Electricity price,
- Lime kiln fuel prices (natural gas and lignin),
- Capital recovery factor (CRF) and
- ASU electricity consumption.

It should be noted that many commonly examined variables like wood price were excluded, as the operation of the fibre line was assumed to remain unchanged in all of the higher cost estimates and most of the lower cost estimates. According to Teir et al. [5, p. 42] many evaluations operate with an uncertainty of CCS cost estimates in the order of +/- 30%. Thus a deviation as large as **+/- 30 %** was considered appropriate. The deviations were calculated **based on the higher cost estimate.** The effects of these deviations for the chosen technologies are presented in Figures 21-24.

The sensitivity analysis of oxy-fuel combustion in the recovery boiler is presented in Figure 21.
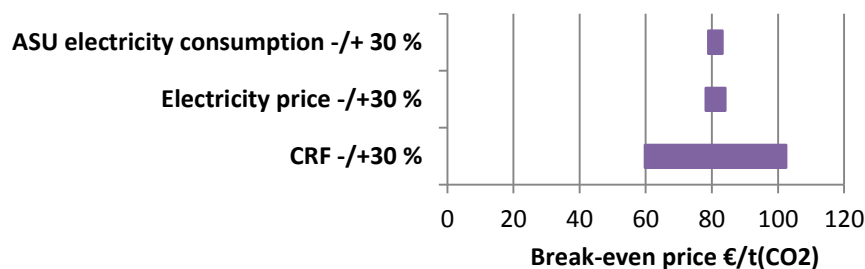
Figure 21.   *Sensitivity analysis of option 4. Oxy-fuel combustion in the recovery boiler.*

As Figure 21 illustrates, the variations in the CRF affected the break-even price the most in the case of *oxy-fuel combustion in the recovery boiler*. This is explained by the large investment cost of oxy-fuel systems and the ASU. The changes in ASU electricity consumption and electricity price were small in comparison.

The sensitivity analysis of option *8. Fuel switch in the lime kiln* is presented in Figure 22.
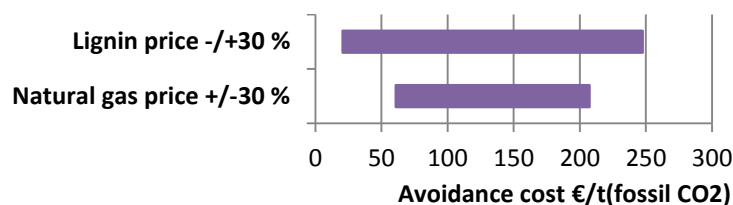
Figure 22.   *Sensitivity analysis of option 8. Fuel switch in the lime kiln.*

The economics of the fuel switch option were assumed to depend mainly on the fuel prices and the associated investment to be negligible. This assumption is reflected in the

results, as the cost of avoiding fossil CO_2 with *fuel switch* proved sensitive to the fuel prices. The sensitivity results from the small difference in the assumed costs of lignin and natural gas prices. Changes in the price of lignin resulted in larger changes in the avoidance cost, because the assumed price was higher than the price of natural gas and the same proportional uncertainty of +/- 30 % was used for both.

The sensitivity analysis of the second oxy-fuel combustion option, applied in the lime kiln, is presented in Figure 23.
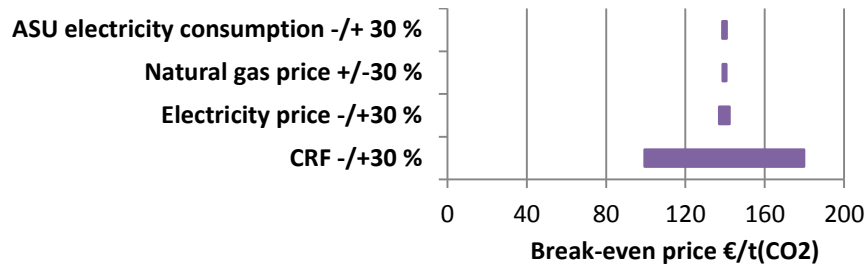
Figure 23.   *Sensitivity analysis of option 12. Oxy-fuel combustion in the lime kiln.*

The option of *oxy-fuel combustion in the lime kiln* was also sensitive to the CRF, as illustrated in Figure 23. Similar to the oxy-fuel recovery boiler, large investment costs were the reason for this. The largest difference to oxy-fuel combustion in the recovery boiler was the *absolute* change in the break-even price, which for the lime kiln ranged from 100 to 180 €/t(CO_2) and for the recovery boiler from 60 to 100 €/t(CO_2). Small changes in the break-even price were observed related to the natural gas price, because the oxy-fuel combustion in the lime kiln resulted in small fuel savings.

Figure 24 presents the sensitivity of oxy-fuel combustion in the lime kiln and the recovery boiler.
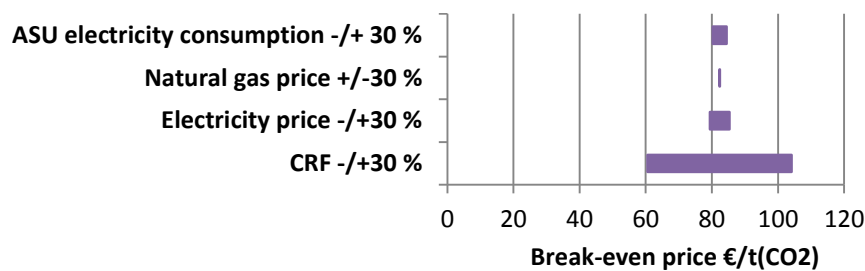
Figure 24.   *Sensitivity analysis of option 13. Oxy-fuel combustion in both the recovery boiler and the lime kiln.*

In technology option *13. Oxy-fuel combustion in both units*, the recovery boiler dominated the investment and thus the results are similar to the results of oxy-fuel combustion in only the recovery boiler. The main difference was that the natural gas price did not affect oxy-fuel combustion in the recovery boiler.

8. DISCUSSION

In this thesis the economic and technical feasibilities of thirteen carbon capture technologies for pulp mills were assessed. The assessment covered the technical maturity, break-even price and capture potential of the studied technologies.

CCS is motivated by the mitigation of the climate change, but for this purpose only large volumes of captured carbon are of significance. By implementing CCS in the Finnish pulp mills it is possible to capture up to 12 $Mt(CO_2)$/a. Small scale implementation of CCS may still facilitate the large scale implementation. Many of the CCS concepts lack the practical experience and further help could be offered as project funding or investment grants.

In addition to possible CCS implementation, the pulp and paper industry already creates a temporary carbon sink by binding biogenic carbon into wood-based products. Cheap tropical fibre has put pressure on the competition in the global market and larger units are built to gain better economic feasibility. The uncertainty in the future of traditional paper products is reflected by the recent fluctuations in the total paper production capacity [156]. The question remains, how to best retain or increase the amount of carbon based products as old products exit the cycle. Two rising trends were found: new materials and liquid biofuels.

Lignin separation was found to be a feasible way avoiding fossil carbon emissions with a capture potential of 1.45 $Mt(CO_2)$/a in Finland and a negative break-even price of −49 €/t(CO_2) to −198 €/t(CO_2) depending mainly on the possible pulp production increase and available investment support. The technology has also been applied at commercial scale already [59, 60]. The separated lignin can either be used as fuel or raw material for carbon fibers for instance. If used as fuel, the environmental effect would come from replacing fossil fuels. If the lignin is used as raw material, the carbon would be stored in the product for its lifetime. Such concepts would be beneficial for the pulp and paper industry, because lignin based products are likely more valuable the lignin as fuel. Lignin based products would also benefit the environment, because the carbon would be stored longer and more so if the material is recycled. Possibilities for applying lignin are only emerging, thus limiting the interest for quick implementation of this option. The implementation requires quite large changes and therefore, it may be most profitable to wait until next major rebuilding at the mill [66].

Another promising concept is BLG to DME. The concept combines carbon capture with liquid biofuels production. Liquid biofuels are expected to have a high demand, as soci-

eties are craving for alternative fuels. The maturity of the technology is improving as it was successfully demonstrated in Piteå, Sweden [61], but large scale implementation is pending. While not necessarily being as profitable as lignin separation in the current market situation, the investment support and lower taxation for sustainably produced liquid biofuels are enough to make BLG to DME economically feasible. The ambitious goals of the Finnish government in increasing biomass use in the energy sector [2, p. 32] may result in more support in the future. Implementing BLG to DME requires major changes at the pulp mill and thus its capture potential is limited to mills that will be re-built in the near future. The capture potential of BLG to DME was 0.82 $Mt(CO_2)$/a with break-even prices varying from 42 $€/t(CO_2)$ to -98 $€/t(CO_2)$.

Small scale carbon capture options may prove to be profitable for producing CO_2 for utilization at the pulp mill or its close surroundings and more easily applicable than large-scale technologies. Pre-calcination before the lime kiln for instance had an esti-mated break-even price of 4.5 to 7.3 $€/t(CO_2)$, with a capture potential of 0.25 $Mt(CO_2$/a), however, the technology is yet to be demonstrated in practice. The break-even price and the investment cost are much smaller than with large-scale tech-nologies, such as MEA-absorption, and thus the effort to implement the technology is significantly lower as well.

The lime kiln options in general had much smaller potentials due to the smaller associ-ated carbon flows. Some technologies can possibly be adapted from the cement indus-try, but if the size is reduced greatly, the specific investment costs would rise, possibly even exceeding the benefits from the increased pulp production. The high specific in-vestment costs of some lime kiln options, such as oxy-fuel combustion, could be low-ered by combining them with other technologies including same unit processes; larger processes using an ASU would be oxy-fuel combustion in the recovery boiler or the BLG options.

The implementation of large scale post-combustion carbon capture in the Finnish pulp and paper industry is possible, but seems unlikely, because the following barriers re-main:

- high break-even prices,
- EU ETS does not include biogenic CO_2 and
- transportation of CO_2 from most Finnish pulp mills to storage outside of Finland lacks economically feasible concepts.

MEA absorption and the modelled oxy-fuel combustion options hold large capture po-tentials up to 12 $Mt(CO_2)$/a and 3.7 $Mt(CO_2)$/a, respectively. The associated break-even prices are high, around 80 $€/t(CO_2)$, without support and excluding transportation and storage. The current investment support for environment protecting technology may decrease the break-even price to around 50 $€/t(CO_2)$ for the oxy-fuel combustion, as

shown in Figure 19, or even lower with possible pulp production increment, but technical issues like air leakage remain to be solved.

Implementing CCS in pulp mills is bio-CCS as mainly biogenic CO_2 is captured, and can result in negative emissions or the reduction of the amount of CO_2 in the atmosphere. A considerable drawback for any *bio-CCS* activity is that the expected progress of EU ETS does not seem to include biogenic CO_2 by 2030, thus making fossil carbon capture more economically feasible. The only studied technology options benefiting from the current EU ETS are the lime kiln options, since approximately a third of the CO_2 emissions of the lime kiln are of fossil origin. However, most of the lime kiln options were rather far from being economically feasible. Political support would be needed for the technology options that are closer to being economically feasible, such as oxy-fuel combustion in the recovery boiler. The support could be redirected by for instance including the biogenic emissions in the EU ETS. Including biogenic emissions in the EU ETS without adjusting the amount of free allocations could increase the financial pressure on the pulp mills. Therefore the amount of free allocations should rise from 90 % to around 98 % to avoid carbon leakage. Still, the break-even price should be at least around 50 €/t(CO_2), excluding transportation and storage costs.

For any large scale CCS activities in Finland the transportation and storage issues remain a significant challenge. *Mineral carbonization* could provide for a local storage possibility, if the economic feasibility is improved. Broader utilization of CO_2 might relieve the pressure from finding more economically feasible storage solutions, but the related utilization potentials are usually quite small. As the experience from the carbon storage facilities amounts, the criteria for the quality of stored CO_2 and the storage site might be revised too. Less strict storage site criteria might make closer storage sites available, as some of the possible geological formations are currently excluded due to their less secure characteristics.

Large scale CCS is expensive, which gives reason to examine the whole carbon cycle of a wood product, as presented in Figure 1. An *underground storage* is only one of the three presented carbon sinks, the other two being *forests* and *wood based products*. The wood based products discussed so far have included new wood based materials and biofuels, but these options regarded mainly wood *residues*. In addition, wooden structures, buildings and furniture should be promoted to replace other more energy intensive materials, like as plastic and metals. The costs of CCS should also be compared to that of growing more forests and avoiding deforestation. As presented in the IPCC Assessment Report [18, p. 553] based on the data of Sathaye and Andrasko [157], forestation related mitigation potential costing as little as 2.7 $(2007)/t($CO_2$) is estimated to amount globally to more than 90 000 Mt(CO_2) between 2000 and 2050. Complicating factors might include supporting policies, local legislation, long time scales, surveillance and land ownership in foreign countries.

9. CONCLUSIONS

In this thesis the feasibility of carbon capture in Finnish pulp mills was studied. Thirteen carbon capture technologies were assessed based on their *technical maturity*, *capture potential* and *break-even price (BeP)*. Three main objectives were set:

- to identify the carbon capture technologies that are implementable in pulp mills in the near future,
- to assess the CO_2 capture potential of each technology and
- to estimate the break-even price for the emission allowance if biogenic emissions are included.

The technical maturity was defined by whether it is possible to implement the technology by 2030. The capture potential gives the potential amount of captured CO_2 if the technology option in question is implemented in all possible Finnish pulp mills. The break-even price is the price of emission allowance including biogenic emissions that results in unchanged economic performance of the pulp mill when implementing a carbon capture technology, *excluding transportation and storage costs*. The compiled data from modelling and literature of all the technologies was recalculated to match the same assumptions of this thesis. The focus was on carbon capture and the full CCS chain was reviewed only briefly. In the wider context the purpose was to find out whether carbon capture in the pulp and paper industry can realistically help mitigate the climate change.

Two sets of calculations were performed for the break-even price: a higher estimate without supporting policies and other possible income, for example from pulp production increase or heat integration and a lower estimate. The higher and lower break-even prices ranged from −49 €/t(CO_2) to 240 €/t(CO_2) and from −198 €/t(CO_2) to 193 €/t(CO_2), respectively. The lowest break-even prices were attributed to lignin separation with the capture potential of 1.45 Mt(CO_2)/a and the technology is commercially available. Another technology with a negative lower break-even price of −98 €/t(CO_2) was black liquor gasification to transportation fuels (BLG to DME) with a capture potential of 0.82 Mt(CO_2)/a, but the technology is still on demonstration level. The higher break-even prices for technologies with capture potentials over 3 Mt(CO_2)/a, such as MEA absorption and oxy-fuel combustion in the recovery boiler, were around 80 €/t(CO_2) and the lower break-even prices ranging from 51 to 73 €/t(CO_2). The higher and lower break-even prices for the lime kiln options were between 140-240 €/t(CO_2) and 85-193 €/t(CO_2), respectively, with the exception of pre-calcination with break-even prices of 7.3 and 4.5 €/t(CO_2), but the technology is novel. The capture potential of every single lime kiln option was less than 0.6 Mt(CO_2)/a, which led to a disadvantage

in the break-even prices of most lime kiln options compared to the recovery boiler options due to the economy of scale.

A sensitivity analysis was provided for the technologies modelled and calculated in this thesis. For the oxy-fuel combustion cases deviations of +/- 30 % in the annual capital costs resulted in the largest changes in the break-even price, from 60 €/t(CO_2) to 100 €/t(CO_2) for the recovery boiler for instance. The maturity of the studied technologies ranged widely, and until practical experience is gained, significant uncertainties remain in the estimates of the investment costs. For the fuel switch option, deviations of +/- 30 % in the lignin price resulted in break-even prices from 20 €/t(CO_2) to 250 €/t(CO_2). Lignin utilization as raw material may rise the demand and thus the price of lignin in the future.

The results of this thesis showed that with the current and expected political support cost efficient carbon capture options should include a tradable by-product or another revenue stream, such as increased pulp production. The most profitable options of lignin separation (BeP: −49…−198 €/t(CO_2)) and BLG to DME (BeP: 42…−98 €/t(CO_2)) included such a by-product. However, the associated capture potentials were limited due to highest possible lignin separation fraction and the remaining carbon in the DME. Supporting policies affected the profitability significantly, as demonstrated for instance by the difference of 140 €/t(CO_2) between the higher and lower break-even price of BLG to DME. Investments in large, novel technologies as well as liquid biofuels production benefitted most from the assumed policies.

Based on the results of this thesis it can be concluded that there is some cost efficient capture potential in the Finnish pulp mills, but increasing the amount carbon based products may be of even greater significance. Currently, up to 11 Mt(CO_2)/a is stored in pulp products for the product lifetime. Large scale CCS implementation in the Finnish pulp and paper industry could potentially abate up to 12 Mt(CO_2)/a, but currently there is no incentive to capture biogenic CO_2. An adequate incentive could be provided by the inclusion of biogenic in the EU ETS and the allowance price rising at least to around 50 €/t(CO_2), excluding transportation and storage costs. The examination of the broader context might also reveal more feasible climate change mitigation options, such as promoting wood use as material, sharing sustainable forestry practices and investing in forestation.

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APPENDIX A: AIR COMBUSTION LIME KILN MODEL
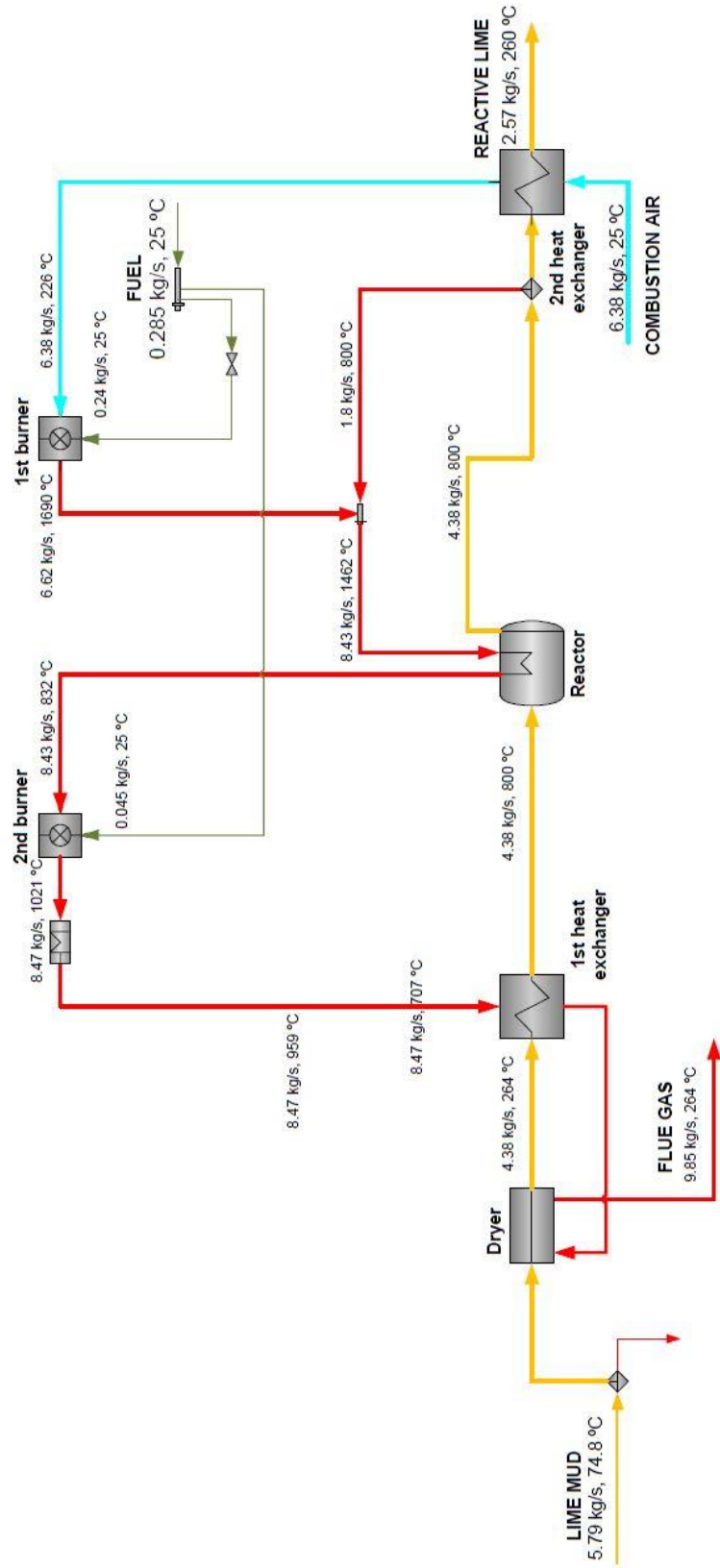
APPENDIX B: MAIN MODULES AND PROCESS STREAMS IN AIR COMBUSTION LIME KILN MODEL

Modules

1st burner
Module type: burner
Fuel load = 13.37 MW

2nd burner
Module type: burner
Fuel load = 2.50 MW

Heat loss
Module type: Heater/cooled, define thermal duty
Thermal duty = - 0.70 MW

Dryer
Module type: Air flotation dryer
Heat loss* = - 0.0 MW
* Heat losses of the entire lime kiln were modelled with the separate *Heat loss* unit above.

1st heat exchanger
Module type: Counter-current heat exchanger with no phase change
Thermal duty = 2.77 MW

Reactor
Module type: Isothermic reactor
Temperature = 800 °C
Thermal duty = 6.91 MW
Conversion rate of $CaCO_3$ = 95 wt-%

2nd heat exchanger
Module type: Counter-current heat exchanger with no phase change
Thermal duty = 1.33 MW

Process streams

Stream class	Name	Flow [kg/s]	T [°C]
Combustion gases	Combustion air (in)	6.38	25
	Combustion air (heated)	6.38	226
	Exhaust gas (1st burner)	6.62	1690
	CO_2 from reactor	1.8	800
	Exhaust gas (mixed)	8.43	1462
	Exhaust gas (reactor)	8.43	832
	Exhaust gas (2nd burner)	8.47	1021
	Exhaust gas (after heat loss)	8.47	959
	Exhaust gas (1st heat exchanger)	8.47	707
	Exhaust gas (dryer – out)	9.85	264
Fuel	Fuel (in)	0.285	25
	Fuel (1st burner)	0.24	25
	Fuel (2nd burner)	0.045	25
Lime mud	Lime mud (in) H_2O 23.88 wt-% $Ca(OH)_2$ 0.51 wt-% $CaCO_3$ 74.62 wt-% CaO 0.38 wt-%	5.79	74.8
	Lime mud (to 1st heat exchanger) H_2O 0 wt-% $Ca(OH)_2$ 0.67 wt-% $CaCO_3$ 98.82 wt-% CaO 0.51 wt-%	4.38	264
	Lime mud (to reactor) H_2O 0.0 wt-% $Ca(OH)_2$ 0.67 wt-% $CaCO_3$ 98.69 wt-% CaO 0.51 wt-%	4.38	800
	Lime mud (to 2nd heat exchanger) H_2O 0.0 wt-% $Ca(OH)_2$ 1.15 wt-% $CaCO_3$ 8.41 wt-% CaO 90.44 wt-%	4.38	800
	Lime mud (from 2nd heat exchanger - out) H_2O 0.0 wt-% $Ca(OH)_2$ 1.15 wt-% $CaCO_3$ 8.41 wt-% CaO 90.44 wt-%	2.57	260

APPENDIX C: EXAMPLE OF THE FULL ECONOMIC CALCULATION PROCEDURE

A) Original data

Parameter	Formula	Value	Unit	Comment / Source
Total oxygen flow	=: O2-tot	53,2	kg/s	Balas-model, CASE-3
Oxygen for lime kiln	=: O2-LK	1,3	kg/s	not in use in this case
Oxygen for recovery boiler	=: O2-RB = O2-tot - O2-LK	51,9	kg/s	
ASU electricity use	=: ASU-el * ASU_eff	29,4	MW	Balas-model, CASE-1: 41,1 MW
ASU heat integration	=: ASU-heat * ASU_eff	15,0	MW	Balas-model, CASE-3
ASU el. use for O2 of the RB	= ASU-el * (O2-RB/O2-tot)	28,7	MW	Based on the O2 share of the RB
ASU heat prod. from O2 of the RB	= ASU-heat * (O2-RB/O2-tot)	14,6	MW	Based on the O2 share of the RB
ASU el. use for O2 of the LK	= ASU-el * (O2-LK/O2-tot)	0,7	MW	Based on the O2 share of the RB
ASU heat prod. from O2 of the LK	= ASU-heat * (O2-LK/O2-tot)	0,4	MW	Based on the O2 share of the RB
Total flue gas flow	=: FG-tot	97,2	kg/s	Balas-model, CASE-3
Total flue gas CO2 content	=: FG-tot-CO2%	57,9	%	Balas-model, CASE-3
Total CO2	=: FG-tot-CO2 = FG-tot * (FG-tot-CO2%/100)	56,2	kg/s	
RB flue gas flow	=: RB-FG	92,5	kg/s	Balas-model, CASE-3
RB flue gas CO2 content	=: RB-FG-CO2%	58,1	%	Balas-model, CASE-3
RB CO2	=: RB-FG-CO2 = RB-FG * (RB-FG-CO2%/100)	53,7	kg/s	
LK CO2	=: LK-FG-CO2 = FG-tot-CO2 - RB-FG-CO2	2,5	kg/s	
CCS el. use	=: CCS-el = 14,29 + 9,153 - 2,425	21,0	MW	Balas-model, CASE-3
CCS heat integration	=: CCS-heat = 0;	0,0	MW	Balas-model, CASE-3; not available (too cool)
CCS el. use, RB share	= CCS-el * (RB-FG-CO2 / FG-tot-CO2)	20,1	MW	Based on the CO2 share of the RB
CCS el. use, LK share	= CCS-el * (LK-FG-CO2 / FG-tot-CO2)	1,0	MW	Based on the CO2 share of the LK
CCS heat integration, RB share	= CCS-heat * (RB-FG-CO2 / FG-tot-CO2)	0,0	MW	Based on the CO2 share of the RB
CCS CO2 avoided	=: CCS-CO2	52,97	kg/s	Based on the CO2 share of the RB
CCS CO2 avoided, LK share	=: LK-CCS-CO2 = CCS-CO2 * (LK-FG-CO2 / FG-tot-CO2)	2,40	kg/s	Based on the CO2 share of the RB
CCS CO2 avoided, RB share	=: RB-CCS-CO2 = CCS-CO2 * (RB-FG-CO2 / FG-tot-CO2)	50,6	kg/s	Based on the CO2 share of the RB
Turbine power	=: turb-el	129,7	MW	Balas-model, CASE-3
Change in turbine power	= turb-el - turb_el(CASE-0)	8,7	MW	Compared to Reference CASE-0
Kiln fuel used	= LK-fuel = 13,37 + 1,87	15,2	MW	Balas-model, CASE-3
Change in kiln fuel used	= LK-fuel - (LK_B1_load + LK_B2_load)_CASE-0	-0,6	MW	Compared to Reference CASE-0
Model mill capacity	=: model_cap	99,0	ADt/h	Balas-model, CASE-3
Model mill scale factor	=: mill_scale = mill_capacity / (model_cap * 24 h)	0,5	-	Compared to selected mill capacity (in 'Variables')

B) Original data with assumptions from this study

Balas model data already based on the assumptions of this study

C) Potential in new mills

Parameter	Formula	Value	Unit	Comment / Source
Carbon reduction with modifications	= CCS-CO2 * mill_scale * 3600 * yearly_h * 0,001 * (1/10^6)	0,770	Mt(CO2)/a	mill size from 'Variables'
Potential in rebuilt mills	= (rebuild_cap/mill_capacity) * Carbon reduction with modifications	3,67	Mt(CO2)/a	in mills rebuilt by 2030

D) Potential in mills without rebuild - not applicable for this technology

E) Investment cost

Parameter	Formula	Value	Unit	Comment / Source
ASU investment cost	= ASU_invest * ((O2-tot * mill_scale)/ASU_prod)^Scale-up	95,99	10^6 €	
Cost of oxy-fuel modifications for a new RB	= OF-RB_invest * mill_capacity	167,41	10^6 €	
Cost of oxy-fuel modifications for a new LK	= OF-LK_invest * mill_capacity	16,41	10^6 €	
Total investment cost	= ASU investment cost + Oxy-fuel RB investment cost	279,82	10^6 €	

F) Stripped operation cost & profit - rebuilt mills

Parameter	Formula	Value	Unit	Comment / Source
Annual capital cost	= Total investment cost * CRF	55,96	10^6 €/a	
Electricity loss	= (-Change in turbine power + ASU-el + CCS-el) * el_pr * yearly_h * (1/10^6) * mill_scale	7,58	10^6 €/a	
Lime kiln fuel savings	= Change in kiln fuel used * LK_fuel_pr * yearly_h * (1/10^6) * mill_scale	-0,10	10^6 €/a	
Stripped cost		63,44	10^6 €/a	
Stripped avoidance cost	= Stripped cost /Carbon reduction with modifications	82,34	€/t(CO2)	

G) Stripped operation cost & profit - mills without rebuild - not applicable for this technology

H) Costs with near future policies and potential profits - rebuilt mills

Parameter	Formula	Value	Unit	Comment / Source
Stripped cost for rebuilt mills		63,44	10^6 €/a	
Investment grant	= (Inv_support/100) * ASU investment cost * CRF * (-1)	-3,84	10^6 €/a	
Lower biofuel taxation				
EU ETS	= Carbon reduction with modifications (CASE-2) * ETS_CO2_pr * Fossil_LK * (1-(ETS_free/100)) * (-1)	-0,30	10^6 €/a	Around 1/3 of LK CO2 emissions are of fossil origin
R&D funding	= Cost of oxy-fuel modifications for a new LK and RB * (R&D/100) * CRF * (-1)	-14,71		
heat integration	= ASU-heat * mill_scale * yearly_h * heat_pr * (1/10^6) * (-1)	-1,21		
Supported cost		43,39	10^6 €/a	
Supported avoidance cost	= Supported cost /Carbon reduction with modifications	56,32	€/t(CO2)	
Support	= Supported avoidance cost - Stripped avoidance cost	-26,02	€/t(CO2)	

I) Costs with near future policies and potential profits - mills without rebuild - not applicable for this technology

APPENDIX D: TECHNICAL AGE DISTRIBUTION OF FINNISH KRAFT PULP MILLS

Pulp mill	Founded	Upgraded	Age	[ADt/d]*	Source
Enocell Oy, Uimaharju	1967	1993	22	1260	[140, 141]
Metsä Fibre Oy, Joutseno	1909	1998	17	1890	[142]
Metsä Fibre Oy, Kemi	1893	1985	30	1616	[143]
Metsä Fibre Oy, Rauma	1996		19	1781	[144]
Metsä Fibre Oy, Äänekoski	1985		30	1452	[145]
Stora Enso Oyj, Heinola		1990	25	726	[146, 147]
Stora Enso Oyj, Kaukopää, Imatra		1996	19	2260	[147, 148]
Stora Enso Oyj, Tainionkoski, Imatra		1996	19	493	[147, 148]
Stora Enso Oyj, Oulu	1937	1984	31	986	[147, 149]
Stora Enso Oyj, Sunila		2000	15	1014	[147, 150]
Stora Enso Oyj, Veitsiluoto		1977	38	1027	[147, 151]
Stora Enso Publication Papers Oy Ltd, Varkaus (2 mills)		1985	30	630	[147, 152]
UPM-Kymmene Oyj, Kaukas, Lappeenranta (2 mills)		2004	11	2027	[153]
UPM-Kymmene Oyj, Kymi, Kuusankoski (2 mills)		2008	7	1452	[154]
UPM-Kymmene Oyj, Pietarsaari		2004	11	2164	[155]
TOTAL number of Kraft pulp mills / capacity			15	20781	
AVERAGE age / capacity			21,6	1385	
Capacity in mills aged 26 years and up				5712	
Capacity in mills aged 6-25 years				15068	

* Production capacity/year divided by 365
Unproven data