Replacing Coal Used in Steelmaking with Biocarbon from Forest Industry Side Streams

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Cover image: Raahe works area and its environment (SSAB 2019a)

Preface

The FOR&MET R&D project (Added Value from Forest Industry for Metals Producing and Processing Integrates) was one of the projects selected to participate in TEKES Challenge Finland initiative. FOR&MET stage 1 (2016) and stage 2 (2017-2019) focused on creating an industrial symbiosis where side streams and waste from the forest industry are used as reductants in metal production and processing as direct substitutes for fossil carbon.

The new large-volume sources of carbonized biochar include sulphur-free lignin from lignocellulosic bioethanol production from sawmill residues (sawdust), wood-based residues (bark) and black pellets from integrated pulp mill operations, which can economically be used to replace powdered injection coal in the blast furnace ironmaking process. In addition, the properties of lignin were evaluated for its possible usage as a component in metallurgical coke. The experimental proof of concept was supported by advanced thermodynamic process modelling of the blast furnace and further assessed with rigorous multiphase analysis of integrated pyrolysis and reduction processes.

With such an approach, the process requirements of ironmaking were set as the measures for the physical, chemical and metallurgical properties of the biomass-based reducing agents. Critical factors such as particle size, water uptake, rheological properties and reactivity of the injection coal as well as the chemical composition of non-process elements were evaluated, while special emphasis was placed on the availability and economic feasibility of the selected biocarbon sources for eventual pulverized coal injection (PCI) usage. The environmental impacts of their application was studied with application-specific life cycle assessment (LCA-methodology), covering GHG emissions and fossil fuel depletion, among others.

The use of forest industry residues as fossil carbon substitutes in metal production and processing will reduce the climate impact of both industries. Underutilized or dumped as a left-over discard, these residues would increase the production-related CO_2 emissions of the forest sector. If, instead, they are used as biocarbon in the steel industry, they provide a green carbon source for steel mills.

Biocarbon-based reductants thus support the development of sustainable forest industry, providing a marketable product for reducing fossil CO_2 emissions. The metal production industry will also directly benefit from decreased CO_2 emission trade costs. In the FOR&MET project the focus has been to elucidate possibilities for market-based synergy between these two major branches of the Finnish export industry, creating mutual competitiveness based on the ample biomass resources of Finland and their sustainable use (Koukkari & Karlsson 2017).

The industrial consortium involved in the FOR&MET Stage 2 R&D project consisted of SSAB Europe Oy (Raahe works), Finnpulp Oy, St1 Biofuels Oy (merged with St1 Oy Jan 1st, 2019) and Valmet Technologies Oy. The consortium companies also committed to supporting the proposed public research project with their own related R&D projects focussed on the same raw materials and/or technologies.

The research institutes responsible for carrying out the research were VTT Technical Research Centre of Finland Ltd and the University of Oulu.

The FOR&MET project's total budget was EUR 686,000 (2016-2019). VTT's contribution was EUR 597,000 and University of Oulu EUR 89,000. Business Finland's share of financing was 61% of the total budget.

Steering group members of FOR&MET project stage 2 were: Jarkko Piirto (Business Finland), Timo Paananen (SSAB Raahe), Olli Mattila (SSAB Raahe), Patrick Pitkänen (St1 Biofuels), Jukka Hietanen (St1 Biofuels), Timo Piilonen (Finnpulp), Jukka Mäkinen (Valmet), Tuomo Hilli (Valmet), Prof. Timo Fabritius (University of Oulu), Hannu Suopajärvi (University of Oulu), Prof. Pertti Koukkari (VTT), Markku Karlsson, and Juha Hakala (VTT; as secretary).

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Espoo, March, 12th, 2019

Juha Hakala, Petteri Kangas, Karri Penttilä, Matias Alarotu, Martin Björnström & Pertti Koukkari

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List of symbols

adt	air-dried tonne
Ba	pulp mill integrated biochar production scenario for bark
basic density	wood dry mass divided by green solid volume
BOS	basic oxygen steelmaking
BP	stand-alone biochar production scenario for black pellets
b-r	bio-reducer
CAPEX	capital expenditures
CDA	carbon direct avoidance
cent	Euro cent (1/100 of Euro [€])
CHP	combined heat and power
DM	dry matter
EUA	European emission allowance
EU ETS	European Union emissions trading system
FT	Fischer-Tropsch
FUPU	future pulp mill
GHG	greenhouse gas
HHV	higher heating value
h-m	hot metal
HP	high pressure steam
LCA	life cycle assessment
LCI	life cycle inventory
LCIA	impact assessment

LHV	lower heating value
LP	low pressure steam
L1, L2	stand-alone biochar production scenarios for lignin
L3	pulp and paper mill integrated biochar production scenario for lignin
NGC	non-condensable gases
OPEX	operating expenses
PC	pulverized coal
PCI	pulverized coal injection
PSD	particle size distribution
RLE	roast-leach-electrowinning process
SAF	submerged arc furnace
SCU	smart carbon usage
SE	steam explosion
thm	tonnes of hot metal
tonne	thousand kilogram
VAT	value-added tax
VMR	Van Marion retort
vol-%	volume percent
wt%	weight percent

1. Introduction

The metal production and processing industry is currently largely dependent on the use of fossil carbon. Coal and coke are needed for both smelting reduction in the production of pristine metal from ores and as an energy source in the subsequent processing stages, in which also heavy mineral oils are frequently used. The iron and steel producing industries represent one of the biggest industrial carbon dioxide emitters, accounting for 4–7% of global (Arens 2010) and a similar range of European (Pardo & Moya 2013) emissions.

The recycling of metals and the recovery of metal values from industrial waste using conventional pyrometallurgical methods are similarly dependent on reductant carbon. The development of such techniques, while showing potential in recovering, for example, technical metals from extensive sources of former production residues is hampered by the necessity of using fossil carbon sources and by the future prospect of economic sanctions for the subsequent release of carbon dioxide into the atmosphere.

The possibility to reduce fossil CO_2 emissions by using biomass instead of fossil carbon sources provides a viable route to mitigate CO_2 emissions in both iron and steelmaking and in the smelting processes of non-ferrous metals production. In Finland, ample sources of biomass are available. Particularly those that can be recovered from the growing side streams of the pulp industry and biorefinery could find a value-adding end-use as a renewable raw material supply for the metal production and processing industry.

1.1 Background

Much effort over many decades has gone into improving the resource efficiency of steel production. Individual processes have been optimized and coupled with the recovery of process gases and waste heat. The by-products of steel manufacturing have found uses as feedstock for other industries, such as slag as building material and in the production of cement. Recycling of scrap steel has increased considerably. The consequence of these multiple incremental improvements is that the global steel industry has achieved very high resource efficiency and producers are reaching the thermodynamic limits of the conventional production technologies (European Commission 2018).

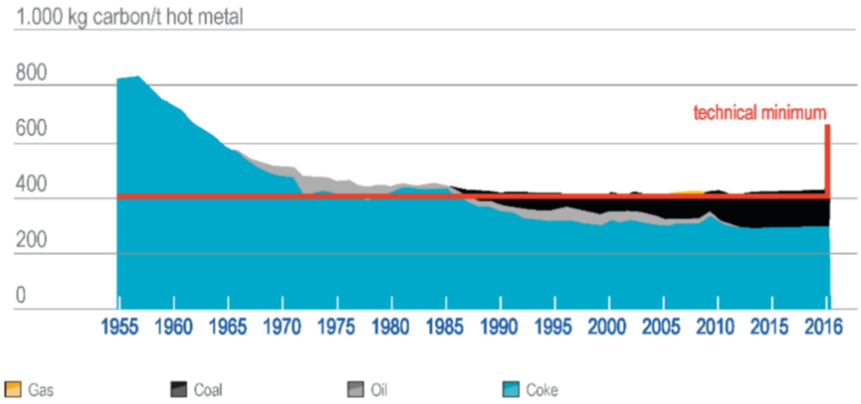


Figure 1. Consumption of reducing agents in the blast furnace (European Commission 2018).

The possible technological pathways for making radical reductions in CO_2 emissions from steel-making include: (i) using the carbon in some possibly unrelated applications of value or (ii) avoiding the use of fossil carbon in the process altogether (ibid.). The first pathway is termed Smart Carbon Usage (SCU); the second is termed Carbon Direct Avoidance (CDA). In the former, one may integrate a chemicals manufacturing process in connection with, for example, blast furnace top gas (ibid.), while the latter includes a variety of technologies where the use of fossil carbon in the steelmaking process can be avoided or replaced by a renewable reducer material or respective energy source.

The anticipated time line as given by the report of the European Commission is schematically represented in *Figure 2*. While the long-term time line anticipates the usage of hydrogen (and even electrolytic) reduction, it is obvious that in the foreseeable future biocoal or from biomass received synthesis gas will be the viable choices as reductants.

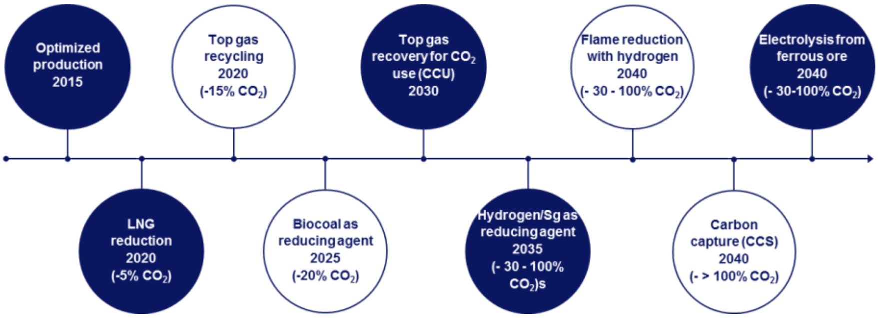


Figure 2. Possible means for reduction of CO_2 impact in the iron and steel industry (European Commission 2018; Suopajärvi et al. 2018; EUROFER 2013).

In Finland, SSAB Raahe with its two blast furnaces is the biggest steel producing site, with an annual production of over 2 million tonnes of standard, premium and special steels. The plant has a coking unit, two blast furnaces, steel melting shop, power plant and hot-rolling mill. The Outokumpu Tornio works, one of the best integrated steel mills in the world, consists of a ferrochrome smelter and steel melting shop with adjacent hot and cold rolling plants. Outokumpu produces ca 1.7 million tonnes/a stainless steel and 520,000 tonnes/a ferrochrome as an intermediate product. Ovako Steel in Imatra manufactures 250,000 tonnes/a engineering steel for customers in the bearing, transportation and manufacturing industries.

Suopajärvi et al. (2013) have estimated the consumption of carbonaceous material in the respective processes as follows: coke in blast furnace ca. 400 kg/tonne (product), powdered coal injection in blast furnace ca 200 kg/tonne (product), coke in FeCr production ca 500 kg/tonne (product), coke in melting furnace ca 10-15 kg/tonne (product). In addition, the sintering of ferrochrome pellets at Tornio would consume ca 50 kg/tonne FeCr-product of coke or coke dust. The respective estimates of CO_2 release figures have been collected in *Table 1*.

Table 1. Estimates of the carbon impact of Finnish steel production (Suopajärvi et al. 2013).

Unit process	**Production [tonnes/a]**	**Carbon or coke [kg/tonne]**	**Carbon content [tonnes/a]**	**CO_2 estimate * [tonnes/a]**
SSAB PCI	2 800 000	200	560 000	1 300 000
SSAB Coke	2 800 000	400	1 120 000	3 483 000
Outokumpu Fe-Cr	520 000	550	286 000	889 000
Outokumpu Arc furnace	1 700 000	15	25 500	79 000
Ovako Steel	250 000	10	2 500	8 000
Total **)	4 750 000		1 994 000	5 760 000

**) based on Statistics Finland estimates of CO_2 emission factors for coal and coke*
***) total steel production of SSAB Raahe, Outokumpu Tornio Works and Ovako Steel.*

The use of biomass-derived fuels and reductants in the iron and steel making industry provides a sustainable option for reducing net CO_2 emissions at a lower capital cost and technological risk than other breakthrough technologies under development. Given that most CO_2 emissions in steel production occur during the reduction of iron ore to hot metal through the use of coal and coke, a key focus of this work has been to partially substitute these with renewable carbon (charcoal or biochar) from sustainable sources such as plantations of biomass species, side streams of forest product industries or forest wastes.

Raw biomass is unsuitable for applications in iron and steel making and it has to be converted into charcoals (chars) through a pyrolysis process before use. The biomass derived chars and hydrocarbon fuels have great potential in lowering the net CO_2 emissions of integrated (blast furnace-BOS route) steel plants. Life cycle assessment can be used to quantify the potential reduction in net CO_2 emissions and may cover the whole life cycle, including plantation, harvesting, transport, pyrolysis and use of chars and bio-oil products.

The ample sources of biomass side streams from different branches of the forest industry can be processed for such uses by drying, torrefaction and pyrolysis. The full potential of biomass-based fuels in metallurgical applications has been a topic of extensive academic study with respect to general economic and environmental perspectives and application-oriented technical issues (Mousa et al. 2016; Wiklund et al. 2016; Suopajärvi & Fabritius 2017; Ng et al. 2018). A recent review of the previous research concerning the usage of bio-based fuels and reducing agents in steelmaking was given by Suopajärvi et al. (2018). Their study focused on the application of biomass in the most commonly used integrated steel production routes. The process requirements were evaluated for specified units while defining the limitations in the physical, chemical and metallurgical properties of alternative reducing agents, setting the conditions that biomass-based reducing agents should meet.

The chemical properties of the solid biocarbon produced via pyrolysis of raw biomass are strongly affected by both the applied pyrolysis technology and the processing conditions. By controlling these conditions, the properties of biochar can be tailored for different applications, such as coke breeze for sintering, coal blend for coke production, injection coal for the blast furnace, and recarburizing in steelmaking (Ng et al. 2018), resulting in optimal performance and greater value-in-use of the char.

Preliminary economic analysis supports the replacement of pulverized coal injection with biochar. However, the economic prerequisites may vary greatly in different parts of the world. Whereas, for example, in Canada or Southern Europe the key factors influencing the economics are the net cost of producing charcoal from biomass, the selection of pyrolysis technology, the value of by-products, and the value-in-use of the charcoal, in Finland and Scandinavia the cost of the biomass itself, even as a side stream from bulk forest sector operations, is often the salient factor. Renewable carbon containing material from the forest sector is nevertheless an asset that can be used both as bio-reducing agents in the short term and, in the longer term, in novel processes with nearly complete coverage by renewables for energy and reducing (Ng et al. 2018, Suopajärvi et al. 2018).

1.2 Previous FOR&MET research

Previous research, conducted particularly in Finland, Sweden and Norway, has provided a firm basis for further progress in implementing the bio-reducing methodology based on local forest resources in metallurgical practice. The viable primary sources are various residues of forest industries, including harvesting, saw and pulp mills as well as the new biorefineries. In the Challenge Finland stage 1 facilitation study (FOR&MET-project) this basis was used for techno-economical assessment of large-scale domestic biochar production and their utilization as a bio-reducing material (Koukkari & Karlsson 2017).

The problem–solution orientation in the FOR&MET stage 1 study also brought up several new options for bio-reducer commercialization not revealed in previously published studies. These include both technically new solutions, such as optional uses of bulk lignin as separated from the lignocellulosic fermentation process and from the Kraft pulping process (recently made commercially available by Ligno-boost; Illi 2016), black pellets from bark, as well as new business-to-business openings including sawdust and residues from integrated sawmill operations (Kyytsönen 2016).

Three such promising business-to-business cases were identified as follows:

1. ST1 bioethanol production in Kajaani will form ca. 100 000 dry tonnes of lignin side product in the near future if a new St1 Cellunolix® plant is built and co-located with the current St1 demonstration plant. A large portion of such side stream can be pyrolysed to biochar and will provide a potential pulverized coal

injection (later referred as PCI) component in the SSAB Raahe blast furnaces. Transportation of the material would require 2-3 trains weekly, depending on whether the pyrolysis unit will be located at the bioethanol production site or at the steel plant.

2. Surplus bark and sawdust from sawmills in the Gulf of Bothnia area will provide an ample source of biomass within a radius of ca 300 km from the metal industries in Raahe and Kokkola. In the case of this raw material source, the bio-based charcoal would be produced on-site at the metallurgical production units.
3. Torrefied bark-based pellet production in the Kuopio area by the planned new Finnpulp Oy biorefinery. The 2-3 trainloads a week produced from this biorefinery would provide the necessary raw material for 20 000 tonnes/a biochar-coal to be used in Raahe or Kokkola.

The FOR&MET stage 1 problem–solution insight was conceptually developed as a marketable business-to-business chain following the business-to-business cases above, and proof of concept was also shown by performing small-scale laboratory pyrolysis and biochar characterization experiments (Koukkari & Karlsson 2017).

Based on this this analysis, the focus of stage 2 was placed on particular case studies where two sources of the developing forest industry in Finland (hydrolysis lignin and bark-based black pellets) were used to evaluate their potential as substitutes for PCI in blast furnaces. The technological data was subjected to techno-economic analysis, including preliminary operational and capital cost (Opex-Capex) estimations for the industrial process-product chain to be developed.

The industrial consortium involved in the Stage 2 R&D project consisted of SSAB Europe Oy (Raahe works), Finnpulp Oy, ST1 Biofuels Oy and Valmet Technologies Oy. Raahe works of SSAB Europe Oy is a leading producer of carbon steel operating two blast furnaces with an overall capacity of over two million tonnes of steel per year. Finnpulp Oy plans to invest in a novel large-scale pulp mill/biorefinery in the vicinity of Kuopio. The planned mill will produce ca. 100 000 tonnes/a of softwood bark suitable for biochar production. St1 Oy is a Finnish energy company producing and selling transportation biofuels. The company develops technology and enabling know-how for the economically viable and environmentally sustainable production of biofuels. In its plant in Kajaani around 80 000 m3 of wet sawdust is processed to bioethanol, leaving some 15 000 dry tonnes lignin per year as a side product. Valmet Technologies Oy is a technology provider committed to developing novel cleantech processes on a large industrial scale. The companies are also supporting public research with respective R&D projects targeting the utilization of these raw materials and related technologies.

2. Description and limitations

2.1 Market information on coal, biofuels and EUA prices, and their dependencies

Release of carbon dioxide from industrial activities is generally valued by two methods, either by a carbon tax or by an emissions trading approach where companies are required to purchase specific permits for their CO_2 emissions. In Europe, the price of CO_2 emissions is set by the European Union Emissions Trading System (EU ETS), while the national level operates by carbon taxation.

In the Finnish carbon tax system, for example, the prices of coal and natural gas used for electricity production purposes are exempt from excise tax, making them significantly cheaper than if used for heat generation purposes only. Combined heat and power plants are subject to 50% taxation. Also, according to Finnish law on the excise taxation of electricity and certain fuels, if coal is used as a raw material, auxiliary material, or as in imminent primary use in goods manufacturing, it is exempt from excise taxes and security of supply payments (FINLEX 1996). It is thus assumed that coking and PCI coal at the steel/iron plant are similarly exempt. Biofuels, such as forest chips and biochar for example, are also assumed to be free of excise taxation at the site of use. Indirect taxation, however, takes place in the production of biochar through electricity and fossil fuel consumption (e.g. transport fuels).

The price of coal and coke in international trade depends on source and type and is often controlled by long-term delivery agreements. For example, in Australia in November 2017, the price of hard coal for coking was estimated at 180% and PCI coal at 120% of the price of thermal coal (IEA 2017). Strong price fluctuations are also more likely compared to thermal coal.

Figure 3 shows the price development of coal at a coastal harbour in Finland with and without excise tax, and *Figure 4* the historical price trend for forest chips at the user's site in Finland. The price level remained low for several years, but in 2018 the trend has been rapidly rising. The price development of emissions trading is visualized in *Figure 5*, based on the situation in late September 2018. An EU emission allowance (EUA) all-time price peak of over 25 €/tonne occurred after this on September 10th 2018 (Sandbag 2018). Since then (evaluated early March 2019) the price has been fluctuating between 16 and 25 €/tonne.

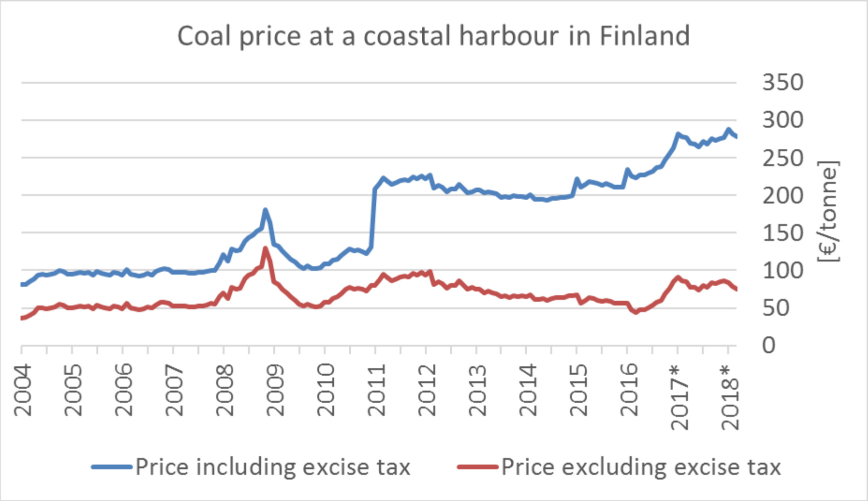


Figure 3. Historical prices of coal at a coastal harbour in Finland; VAT excluded (Statistics Finland 2018a).

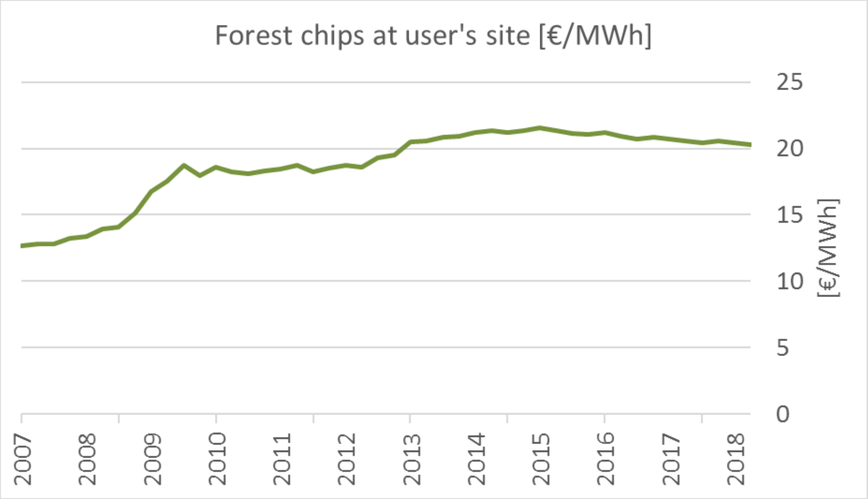


Figure 4. Price history of forest chips; VAT excluded (Statistics Finland 2018b).

The competitiveness of biofuels against fossil fuels in energy production can be compared by the fuel burning costs, i.e. biofuel price compared with fossil fuel price and related emission trading cost. In this work, we focus on coal. Statistics Finland (2018c) specifies a CO_2 emission factor for coal of 93.2 tonnes CO_2/TJ and a lower heating value of 24.9 GJ/tonne. The cost of burning coal, including emissions trading, is illustrated in *Table 2*. Coloured cells indicate the cost-effectiveness of burning biofuels, assuming a biofuel market price of ca. 20 €/MWh, according to *Figure 4*. The cells in the bottom right-hand corner (green font and background) indicate that biofuel is more cost effective than burning coal.

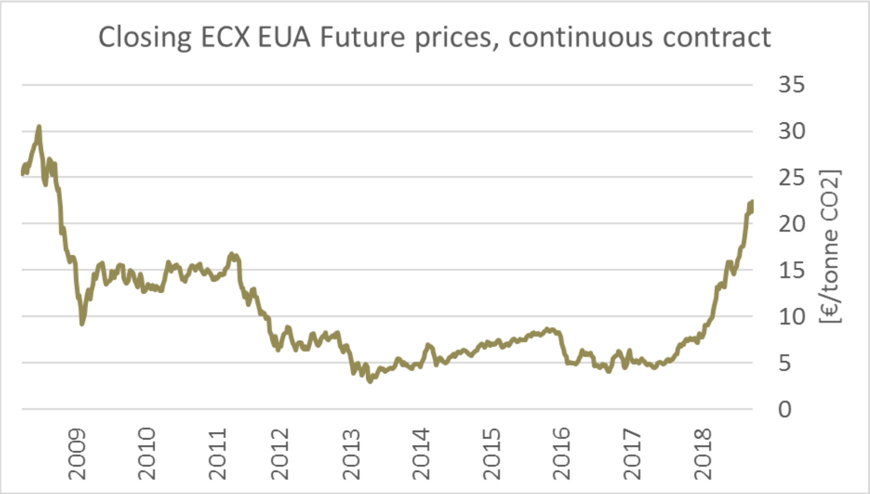


Figure 5. European Union Emissions Trading System carbon market price. Closing ECX EUA Futures prices, Continuous Contract. Data source Sandbag (2018). Based on statistics updated on 24 September 2018.

Table 2. Cost of burning coal, including emissions trading. Coloured cells indicate the viability of replacing coal with biofuels, blue meaning viable. Estimated biofuel market price 20 €/MWh.

Cost of burning coal, including emission trading EUA costs (based on the lower heating value LHV)											
Coal price	Emission trading EUA price [€/tonne CO_2]										
[€/MWh]	0	5	10	15	20	25	30	35	40	45	50
	Coal and EUA [€/MWh]										
0	0	2	3	5	7	8	10	12	13	15	17
2	2	4	5	7	9	10	12	14	15	17	19
4	4	6	7	9	11	12	14	16	17	19	21
6	6	8	9	11	13	14	16	18	19	21	23
8	8	10	11	13	15	16	18	20	21	23	25
10	10	12	13	15	17	18	20	22	23	25	27
12	12	14	15	17	19	20	22	24	25	27	29
14	14	16	17	19	21	22	24	26	27	29	31
16	16	18	19	21	23	24	26	28	29	31	33
18	18	20	21	23	25	26	28	30	31	33	35
20	20	22	23	25	27	28	30	32	33	35	37
22	22	24	25	27	29	30	32	34	35	37	39
Cost – Less than 15 €/MWh				**Cost – Between 19,5 and 15 €/MWh**				**Cost – More than 19,5 €/MWh**			

A further focus of the present study is on the utilization of biochar in metallurgical applications, especially in replacing pulverized coal injected into the blast furnace (PCI coal). For this purpose, tonnage-based coal prices are applied, since also reduction chemistry takes place in the blast furnace and the heating values given for energy production purposes should not be applied as such. The average carbon content of PCI coal is estimated at 80 wt% (ash free, dry). We also assume a typical ash content of 10 wt% (dry) and 10 wt% moisture in PCI coal as received. On this basis, the simulation calculations (Balas® http://balas.vtt.fi/) result in 2.375 tonnes CO_2 per tonne of PCI coal as received. This value is then further applied in our calculations. For comparison, our value is relatively close to the Statistics Finland (2018c) value of 2.32 tonnes CO_2 per tonne of coal.

Table 3 takes into account both the PCI coal price and the emission trading prices. A EUA price all-time peak of over 25 €/tonne of CO_2 took place on September 10th 2018 (Sandbag 2018). Since then (evaluated early March 2019) the price has been fluctuating between 16 and 25 €/tonne. Also, the PCI coal price may well reach the 125 €/tonne level. In 2017 the coal price at the coast of Finland was close to 100 €/tonne (*Figure 3*), PCI coal being ca. 120% of the thermal coal price (IEA 2017), exclusive of possible handling charges from the harbour to the site. By selecting these values, the cost of burning PCI coal, including emission trading costs, is 184 €/tonne, as *Table 3* illustrates.

Table 3. Tonnage-based cost of PCI coal, including emission trading.

Cost of burning PCI coal, including emission trading EUA costs											
Coal price	Emission trading EUA price [€/tonne CO_2]										
€/tonne	0	5	10	15	20	25	30	35	40	45	50
	Coal and EUA [€/tonne]										
25	25	37	49	61	73	84	96	108	120	132	144
50	50	62	74	86	98	109	121	133	145	157	169
75	75	87	99	111	123	134	146	158	170	182	194
100	100	112	124	136	148	159	171	183	195	207	219
125	125	137	149	161	173	184	196	208	220	232	244
150	150	162	174	186	198	209	221	233	245	257	269
175	175	187	199	211	223	234	246	258	270	282	294
200	200	212	224	236	248	259	271	283	295	307	319
225	225	237	249	261	273	284	296	308	320	332	344
250	275	262	274	286	298	309	321	333	345	357	369
275	275	287	299	311	323	334	346	358	370	382	394
300	300	312	324	336	348	359	371	383	395	407	419
						2,375 tonne CO_2/tonne PCI coal					

The production costs of biochar, targeted to replace PCI coal in the blast furnace, were then calculated for a number of scenarios, as presented in detail in *Chapter 7*. Accordingly, tables similar to *Table 2*, but based on PCI tonnages were generated for each calculated scenario, indicating the cost-comparison for replacing PCI coal with biochar in each case.

2.2 Metal industry and reduction processes

2.2.1 Biomass-based reducing agents in metal processing

Biomass can, in principle, be used as bio-based fuel for various purposes at metal processing sites. However, due to its low heating value, the benefits of such an approach are typically modest and become negated, for example, by the required preprocessing and transport costs (Suopajärvi 2013; Wiklund et al. 2016). In contrast, the use of biomass as a reducing agent in roasting or smelting supports the manufacture of value-added metalliferous products. For this purpose, however, the reduction potential of the biomass must be enhanced by extensive pretreatment to increase its natural carbon / oxygen ratio. In *Table 4* some key characteristics of typical biomass components are presented.

Table 4. Characteristics of biomass, biochar, lignin and bituminous coal (Suopajärvi 2013; Tomani 2010; Källi 2016).

Biomass	**Moisture [wt%]**	**Volatiles DM* [wt%]**	**Ash DM* [wt%]**	**C DM* [wt%]**	**H DM* [wt%]**	**N DM* [wt%]**	**O DM* [wt%]**	**HHV [MJ/kg]**
Chopped wood	7.94	82.17	1.77	47.29	6.20	0.42	45.19	18.79
Torrefied: 250°C	2.66	78.16	1.49	54.91	5.87	0.31	40.96	21.21
Lignin**	varied	30-40	0.5-0.8	66.2	6.2	0.2	27.5	25.0***
Biochar: Lignin 500°C	1.3	14	3.0	89.4	2.8	0.3	4.5	33.8
Biochar: Pine bark 500°C	1.7	21	3.5	83.7	3.0	0.5	9.3	20.8
Bituminous coal	1.57	36.6	6.52	77.63	6.3	2.0	9.16	27.72

**DM= dry matter; **Kraft softwood lignin, elemental analysis contains organic sulphur (ca 3%) which is released during lignin pyrolysis; ***HHV of dry lignin.*

There are various means by which biomass-based products can replace fossil carbon in metal production and processing. For example, in the integrated steel plant, biomass products can be used in: (i) coke-making for production of bio-coke; (ii) sintering process for production of bio-sinter, (iii) pelletizing/briquetting for production of bio-composites and/or bio-briquettes, (iv) partial replacement of fuel injected into the blast furnace and (v) bio-recarburization of steel in the ladle furnace. Of these, replacement of coke (or parts of it) would be the most voluminous. Recently, intensive work has been conducted on inserting biomass to the coal blend during coke-making. The resulting bio-coke could be effective at lowering the gasification temperature in the blast furnace and thus minimizing total carbon consumption. It has been estimated that a 20% substitution of biomass for coke in some ironmaking blast furnaces would reduce CO_2 emissions by 15% (de Castro et al. 2013). Even though such figures are not likely to be achieved in most cases, even modest advances would have a considerable impact due to the sheer scale of the global steelmaking industry.

This option is, however, hampered by the detrimental effect of biomass on the thermoplastic properties of metallurgical coke (Montiano et al. 2014). No solution has yet been found for maintaining the mechanical strength necessary for the coke bed in vertical shaft furnaces when even small amounts of biomass, such as sawdust or charcoal, have been added to the coking furnace mix (Mousa &al 2016). The thermogravimetric and differential thermal analysis (TGA and DTA) of coal-biomass blends clearly show three different devolatilization ranges for biomass (denoting decomposition of cellulose, hemicellulose and lignin, respectively), whereas fossil coal pyrolysis shows but one single decomposition range (Vuthaluru 2004; Montiano ibid.).

This has been explained by the different thermal stability of the ether bonds (380-420 kJ/mole) typical of cellulosic material compared to the aromatic C=C bonds (1000 kJ/mole) of coking coal. The wide devolatilization range is assumed to lead eventually to greater mass loss and weakened coke thermoplasticity and mechanical strength even with small (5 wt%) biomaterial additions to the coking blend. However, it is noteworthy from other sources that the same thermal analysis performed for softwood (pine or spruce) Kraft lignin shows little or no secondary devolatilization ranges (e.g. Brodin et al. 2010; Brebu 2010) (*Figure 6*).

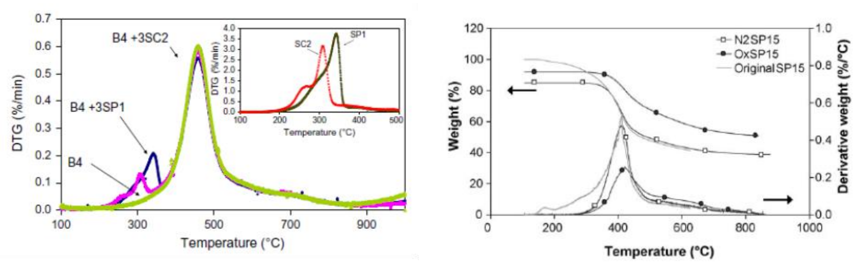


Figure 6. Left: Differential thermal analysis (DTA) of biomass-coking coal blends (B4+3SC2; B4+3SP1) compared with coking coal (B4). Right: Respective thermal analysis of softwood lignins (Montiano et al. 2014; Brodin et al. 2010).

The replacement of pulverized injection coal (PCI) represents the technically more straightforward option, as presently it can be partly composed of nut coke or charcoal (Mousa et al. 2016). The injection of charcoal (to replace tuyere oil or coal injection) will lead to similar environmental benefits to use of bio-coke. It has also been suggested that use of biochar will increase reactivity and support lowering slag and ash formation in smelting operations (Strakhov 2009; Lumadue 2012; Suopajärvi 2015). According to one of the few existing mathematical models for the multiphase blast furnace system, developed by de Castro et al. (2013), the use of charcoal injection could increase blast furnace productivity by 25% (see also the results of Ng 2018). Recently, Ng and co-workers performed an analysis of replacing coal injection with solid biocarbon produced by torrefaction, hydrothermal treatment and pyrolysis (up to 900 C). Further evaluation was performed with a conceptual study including heat and mass balances combined with thermodynamic equilibrium modelling. Their conclusion was that up to a 20% decrease in greenhouse gas emissions could be thus achieved; however, the value-in-use of biocarbon would then highly depend on its carbon-to-oxygen ratio (Ng et al. 2018).

Smelting reduction is also a commonly used pyrometallurgical technique in making ferroalloys in which the reductant carbon can be replaced by biochar (see e.g. Van Wesenbeeck et al. 2016). An emerging option in circular economy is treatment of residues from both ferrous and non-ferrous metals production (Worrell & Reuter 2014). In general, pyrometallurgy has been applied to convert the residues into an inert slag, yet using a reductant in addition to thermal energy. However, the high-temperature treatment also enables the recovery of zinc, copper, lead, silver and other valuable metals that would otherwise be lost with the discard. For such recovery processes, thermal energy can be provided by various fuels or electricity while the reductant is provided as fossil carbon. Thus, for both ferroalloy production and recovery of metal values from waste, use of biochar is an equally viable option. Of the existing metal production in Finland, the formation of zinc and lead containing jarosite waste (180 000 tonnes/a, Boliden Kokkola), which is currently stored in the plant vicinity, is a typical example of such potential residue treatment.

Table 5. Studies related to use of biocarbon as a reducing agent in the iron and steel industry (h-m refers to hot metal and b-r to bio-reducer).

Target	**Bio-reducer**	**Processing of biomass**	**Process integration**	**Charge** [kg/tonne h-m]	**Cost** [€/tonne b-r]	**Investment** [M€]	**Method**	**Other**	**Reference**
Injection coal	Torrefied wood	Torrefaction	Blast furnace top gas to biomass drying	50		22	Optimization		(Wiklund 2016)
Injection coal	Biochar	Slow pyrolysis	Blast furnace top gas to biomass drying; utilization of pyrolysis oil	47		33	Optimization	51 kg pyrolysis oil / tonne h-m	(Wiklund 2016)
Injection coal	Biochar	Fast pyrolysis	Blast furnace top gas to biomass drying; utilization of pyrolysis oil	47		61	Optimization	143 kg pyrolysis oil / tonne h-m	(Wiklund 2016)
Injection coal	Torrefied wood	Torrefaction		120	140-180	13 (50 MW)	Modelling	33-59 €/tonne CO_2	(Suopajärvi 2015)
Injection coal & coke	Biochar	Pyrolysis	Utilization of pyrolysis oil / Stand-alone process	200	360-490	10 (50 MW)	Modelling	22-53 €/tonne CO_2	(Suopajärvi 2015)
Injection coal	Pyrolysis oil	Pyrolysis		< 100	n.a.		Modelling		(Suopajärvi 2015)
Injection coal	Syngas	Gasification		n.a.	n.a.		Modelling		(Suopajärvi 2015)
Injection coal	Bio-methane	Anaerobic digestion		90-150	690-830	38 (50 MW)	Modelling	112-150 €/tonne CO_2	(Suopajärvi 2015)
Injection coal	Biochar	Pyrolysis	Combustion of pyrolysis oil within process	44-64			Optimization		(Helle 2014)
Injection coal	Wood pellets	Pelletizing		20-30	n.a.		Pilot trial	Voestalpine Linz, 1 k-tonnes	(Bürgler et al. 2011)
	Torrefied wood				113-183		Review		(Suopajärvi 2018)
	Biochar				223-513		Review		(Suopajärvi 2018)
	Bio-gas				722-974		Review		(Suopajärvi 2018)
	Bio-hydrogen				890-2002		Review		(Suopajärvi 2018)

2.2.2 Focus on blast furnace operations in steelmaking

Historically, iron was produced in small furnaces using charcoal and iron ore. The combustion air was blown into the hearth with bellows. The product from these furnaces was a solid block of malleable iron containing variable amounts of slag, which was removed by heating and hammering. The block was cut into smaller pieces and forged into tools and weapons or into commercial blooms. In the early eighteenth century, coke replaced charcoal as the fuel and reductant and enabled the expansion of blast furnaces as large-scale production units of cheap, high-quality metal for steelmaking (Yang et. al. 2014).

Industrial blast furnaces use iron ore as the iron-bearing raw material, coke and pulverized coal as the reducing agents and heat source, and lime or limestone as the slag-forming fluxing agents. The main objective of blast furnace ironmaking is to produce hot metal with consistent quality for the subsequent blast oxygen furnace (BOF) steelmaking process. Typically, the specification of steel works requires a hot metal with 0.3–0.7% Si, 0.2–0.4% Mn, and 0.06–0.13% P, and as high a temperature as possible (1480–1520 °C at the tapping). A modern large blast furnace has a hearth diameter of 14-15 m, and a height of 35 m with an internal volume of ~4500 m3. A single large blast furnace such as this can produce 10,000 tonnes of hot metal per day.

The blast furnace is a continuously operating shaft furnace based on the counter-current flow principle. At the top, coke and burden (sinter, pellets, lump ore, and flux) are charged in alternating layers. Charge materials descend under the influence of gravity. In the lower part of the furnace, hot blast from heated stoves is injected through tuyeres. In front of each tuyere, the hot blast reacts with the coke, forming carbon monoxide. The carbon monoxide ascends in the furnace and reduces the iron oxides in the ferrous ores. At the bottom of the hearth, the molten metal is collected. Besides hot metal, slag is formed that floats on top of the hot metal bath due to its lower density. The liquid hot metal (1500 °C) and slag (1550 °C) are tapped regularly. The output of the blast furnace process consists of carbon-saturated iron (hot metal), slag, top gas, and flue dust. The blast furnace is charged semi-continuously with iron-bearing burden, slag formers, and coke. The iron burden consists of sinter, pellets, and/or lumpy ore in various proportions. Slag formers are usually limestone, BOF slag, and quartzite in small amounts to balance the slag rate and chemistry. In an ideal case, sinter and/or pellets are self-fluxing. Coke serves as the basic energy source and as a carbon reductant. Other sources of reducing agents include pulverized coal, hydrocarbons, waste plastics, or biomass, which are injected through the tuyeres. Preheated blast (1200–1300 °C) air or oxygen-enriched air brings an important amount of heat to the process.

Figure 7 shows a schematic configuration of the blast furnace, and in *Figure 8* model diagrams of previously published blast furnace models are shown.

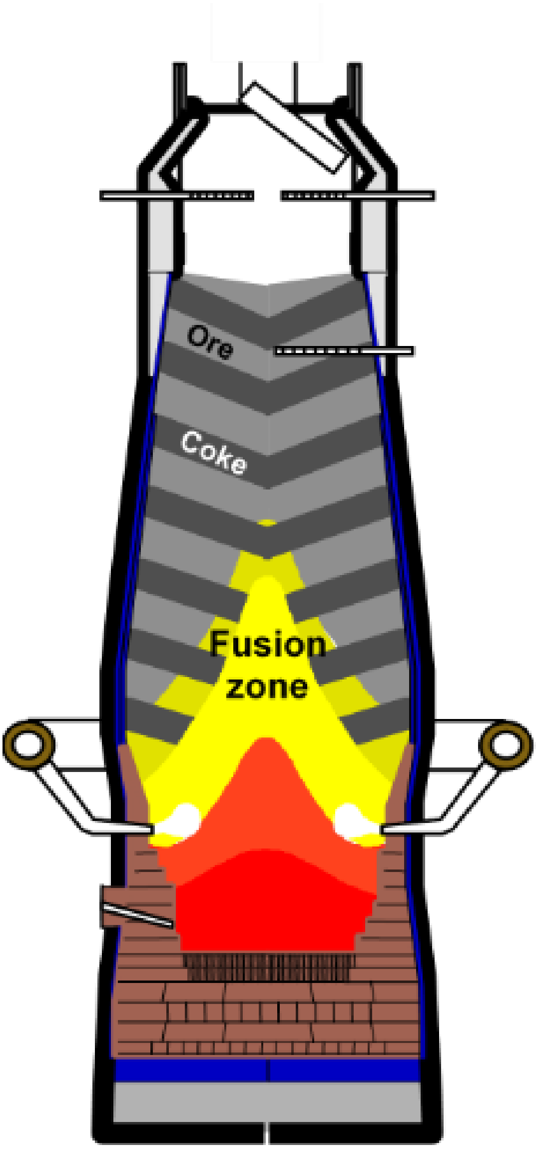
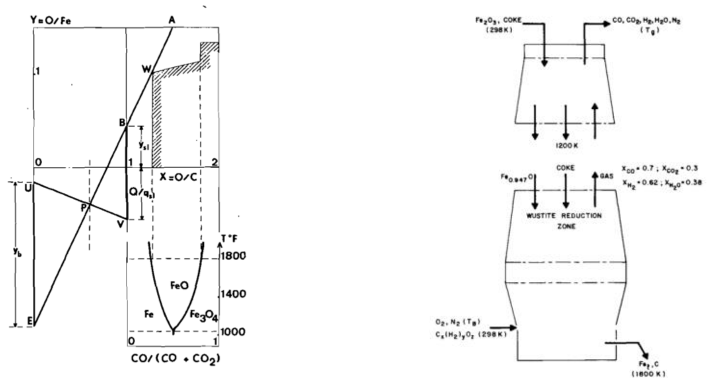
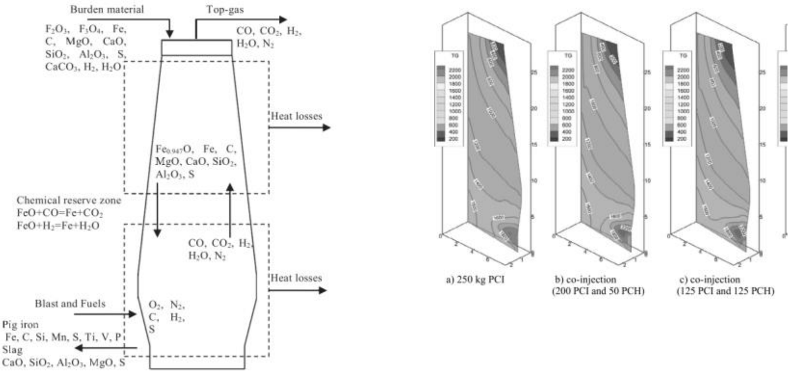


Figure 7. Schematic of a blast furnace.



A. Construction of the operating line under conditions of ideal heat exchange (point P) and ideal oxygen enchance (Point W).

B. Inputs and outputs of the top and bottom segments of a conceptual divided blast furnace when $C_x(H_2)_yO_z$ (a general tuyére injectant) is being injected through the tuyéres.



C. Schematic presentation of the blast furnace process

D. Predictions of inner temperature distributions for 250 kg/thm injection cases: PCI: verized charcoal

Figure 8. Previously published schematic diagrams of blast furnace models: Basic 1D-model structure originally presented by Rist (1967) (A) and utilized by Davenport and Peacey (1979) (B) and by Suopajärvi & Fabritius (2012) (C); and a 3D-model of a blast furnace by de Castro et al. (2011) (D).

2.3 Biomass thermochemical conversion processes

The main thermochemical biomass conversion processes are combustion, torrefaction, slow pyrolysis, fast pyrolysis and gasification. The products are respectively, torrefied biomass, biochar, condensable gases (bio-oil; extracted from pyrolysis gas by cooling), non-condensable gases (NCG) and syngas. The amounts of various conversion products and the percentage of each fraction depend on the process in question, as well as on process conditions (Kangas 2015). Torrefaction and pyrolysis processes are endothermic, thus requiring external energy to sustain the reactions. For wet biomass, a hydrothermal carbonization route is also possible (e.g. Ng et al. 2018). In addition, biotechnological conversion routes exist, such as enzymatic hydrolysis for producing sugars and possible further conversion of these by fermentation.

Possible end uses for the conversion products are numerous, depending of the conversion rate in question:

- Torrefaction improves the energy density and removes excess oxygen from the biomass. Torrefied biomass could be used e.g. to replace fossil fuels in power production and other combustion applications.
- The steam explosion method (Masonite process) may be applied to produce fuel pellets from all types of lignocellulosic material. Biomass chips or splinters are hydrolysed with steam under pressure followed by mechanical defibration (steam explosion). The method produces very durable pellets with good moisture resistance. The heating value uplift is moderate and changes in elemental composition minor.
- Hydrothermal carbonization is typically performed for wet biomass (or sludge) at pressurized autoclaves at temperatures between 200-300 °C. The treatment removes water-soluble inorganic compounds from the solid mass. The product has a relatively low carbon-to-oxygen ratio and heating values similar to those of torrefied biomass.
- Slow pyrolysis maximizes the yield of biochar, which can be used in different applications directly or after further processing, e.g. to replace coal in combustion, as a bio-reducer in metal production, as a soil conditioner, or in water treatment, to mention a few. Hot pyrolysis gases (condensable and non-condensable) are also formed, which can then be used as fuel to replace fossil-based combustibles in different applications. The condensable fraction can be collected to form bio-oil.
- Fast pyrolysis maximizes the yield of bio-oil (from condensable gases). Some NCG is also formed, as well as smaller amounts of biochar. Biochar properties differ somewhat from those obtained by slow pyrolysis, affecting its end use possibilities, e.g. as a bio-reducer.
- In gasification, the biomass hydro-carbons are fully de-volatilized to syngas in the presence of a sufficient amount of oxygen. Main components are CO, H_2, CH_4, CO_2, and lesser amounts of other gases. The syngas can be used in combustion to replace fossil-based fuels in different applications or as an intermediate product for generating hydrogen, alcohols, FT (Fischer-Tropsch) gasoline, FT diesel, olefins, oxo chemicals, ammonia and synthetic natural gas (SNG) (Bain 2004).
- Bio-oil (condensable gases from slow and fast pyrolysis) can be processed further, e.g. to liquid biofuels, to replace, e.g., fossil-based transportation fuels. The

tar-containing fraction of the condensable gases can also be used in binder applications.

- NCG (Non-condensable gases; from slow and fast pyrolysis) compounds differ somewhat from syngas (from gasification) due to lack of oxygen in the reactions. These pyrolytic gases contain H_2, CO, lesser amounts of CO_2, and light hydrocarbons such as CH_4 and C_2H_6. End uses are similar to syngas.
- Sugars can be obtained by breaking down the larger molecules into smaller ones by enzymatic hydrolysis. With the aid of microbes it is then possible to ferment the sugars further to alcohols (ethanol). Chemicals can also be obtained via biotechnology.

Figure 9 illustrates the product streams of thermal conversion as a function of temperature. In the drying phase (100-150°C), moisture is evaporated from the biomass. In the torrefaction phase (150-300°C), the most volatile material is fractionated and the oxygen content of the biomass is reduced. In the pyrolysis phase (300-600°C), the volatiles of the organic biomass fully enter the vapour phase, leaving solid biochar. Gasification then takes place at above 600°C in the presence of sufficient oxygen for carbon monoxide generation and hydrogen and carbon dioxide are formed. All reactive matter is subsequently oxidized producing mainly water and carbon dioxide. Other oxidation products may also be formed, such as nitrogen oxides. These flue gases also contain, for example, volatile alkaline constituents that may further appear as fly ash and sticky condensates when cooling the flue gas (Kangas 2015).

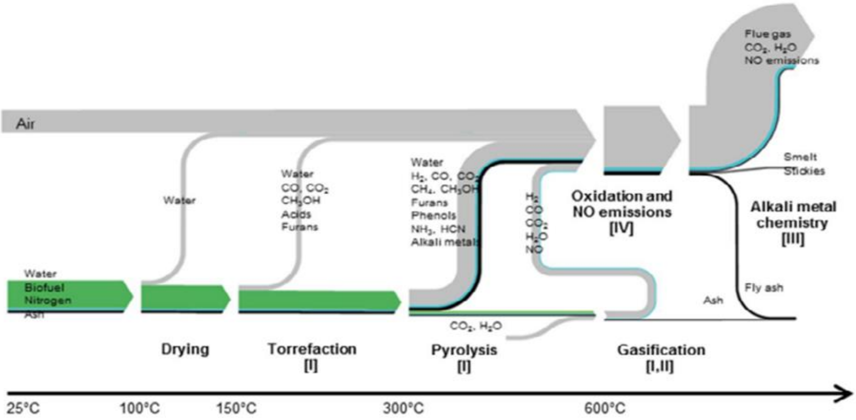


Figure 9. Thermal conversion of biomass as a function of raising temperature (Kangas 2015).

Table 6 provides further information on different conversion technologies, their operating temperatures, residence times, major products, and stages of development. Hydrothermal carbonization is also included, as this treatment is particularly suitable for wet biomass, the slightly carbonized solid residue of which is called HTC biomass.

Table 6. Comparison of thermo-chemical conversion technologies by Suopajärvi et al. (2018).

Conversion technology	Operating conditions	Major products	Development stage	Note
Torrefaction	200-200°C for 15-30 min without the presence of air	Torrefied biomass: slightly carbonized solid residue	Several demonstration plants	
Hydrothermal carbonization	Mixed with saturated water-steam at 150-250°C from a few mins to a few hours	HTC biomass: slightly carbonized solid residue	Several demonstration plants	Suitable for wet biomass. Water soluble inorganic elements are removed with water
Slow pyrolysis	Slow heating to 300-600°C and kept for 15-30 min to several hours without the presence of air	Biochar / Charcoal: highly carbonized solid residues	In use since early human civilization	Process heat often supplied by partial combustion of biomass or volatile products. Particles larger than a few cm
Fast pyrolysis	High heating rate to 400-600°C for a few seconds without the presence or air	Bio-oil: mixture of organic compounds and water; including oxygenates and phenolic compounds	A few commercial plants	Process heat by excess heat from a power station or partial combustion of char. Particles need to be milled less than a few mm
Gasification	600-1400°C with oxidizing agent	Syngas: mixture of H_2, CO and CO_2 with small amounts of CH_4	Several demonstration and pilot plants (coal gasification: commercial plants)	Typically 30-50% of oxygen, or air required for stoic. reactions with steam or CO_2
Combustion	Complete combustion with excess air	High-temperature heat	Commercial plants worldwide	Heat utilized in steam and power generation

Regarding the manufacture of biocarbon to replace PCI coal with the aforementioned side streams of the forest industry as the raw material, slow pyrolysis appears to be the most viable option. This technology gives a high carbon-to-oxygen ratio comparable to that of conventional injection coal and also allows relatively high yields (Suopajärvi 2018). Therefore, the focus of the case studies of this work was on slow pyrolysis (see *Subsection 2.6.2* for a more detailed description of slow pyrolysis).

2.4 Selected biomass sources – properties and availability

In line with the scope of the project, novel high-volume side streams from the forest industry were selected to serve as a basis for the case studies. Sulphur-free lignin from the recently commercialized lignocellulosic bioethanol production using sawmill residues (sawdust) as raw material was selected as a potential source for making substantial amounts of biocarbon. As another option, excess wood-based residues from integrated pulp mill operations, particularly softwood bark and bark-based black pellets, were evaluated.

2.4.1 Hydrolysis lignin

The St1 Biofuels Cellunolix® bioethanol plant in Kajaani produces ethanol from sawdust. The ethanol is used to make high-blend ethanol fuel for flex-fuel vehicles and is also added as a bio-component to petrol. The side-product of the fermentation process is hydrolysis lignin. As St1 Oy plans to increase its ethanol production from lignocellulosic side streams and to expand its operations to new plant sites, the amounts of hydrolysis lignin will increase in the foreseeable future.

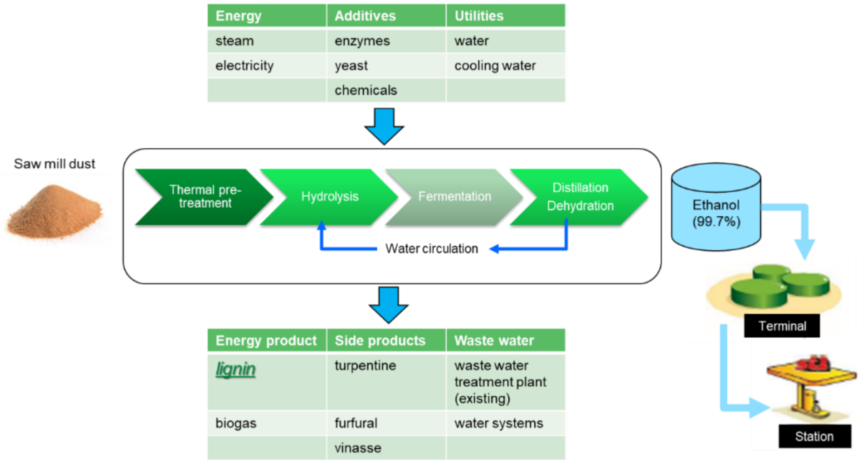


Figure 10. St1 Cellunolix® process, optimized for softwood by Yamamoto (2018).

Further utilization of lignin is an important target for St1, and the company is pursuing several projects alongside the FOR&MET project examining options such as pelletizing the lignin to replace coal, producing bio-crude or biochar from lignin through pyrolysis, and utilizing lignin in bio-chemicals and bio-composites production (Yamamoto 2018).

The annual production capacity of dry lignin at the Kajaani plant is reported to be ca. 15 000 dry tonnes (Yamamoto 2018). Currently, the hydrolysis lignin is utilized at a nearby boiler to produce steam in Kajaani. The dry matter content varies in the range of 45-50%. The lignin content is between 70-80 wt% and residual cellulose 20-30 wt% of the dry matter, while the ash and sulphur contents are low. Some amounts of sugar, acids and degradation products are also present. The lignin has low reactivity (Yamamoto 2018).

In this work, the focus is on producing biochar from hydrolysis lignin by slow pyrolysis. St1 Oy has announced plans to increase Cellunolix® ethanol production by building new plants in other locations, and the available amount of lignin will increase accordingly. In the scenarios chosen in this study we use an annual production level of 98 200 dry tonnes assuming that a larger plant is built to the current demonstration plant location in Kajaani. Samples of hydrolysis lignin were provided by St1 for the FOR&MET project for analysis and pyrolysis experiments carried out at the University of Oulu. Detailed analyses of the lignin raw material are presented in Toloue Farrokh et al. (2019).

2.4.2 Bark

Bark can be obtained from, for example, pulp mill or sawmill debarking lines. It is typically used as boiler fuel at pulp mills. In modern pulp mills, excess bark may be converted by gasification to syngas for combustion, for example, in the lime kiln. Bark can also be further processed into fuel pellets, for example using the steam explosion (SE) technique, to produce compressed 'black pellets' (*Subsection 2.4.3*). The pellets can be used, for example, in coal-fired power plants as biofuel, or converted to biochar by slow pyrolysis.

Instead of using black pellets as an interim step, bark can also be directly converted to biochar by slow pyrolysis integrated within the pulp mill. The resulting hot pyrolysis gases can then be utilized to replace other fuels, for example in the lime kiln. In either case, biochar or black pellets are a marketable side product for the biorefining plant. Both options were selected as a basis for the biochar production scenarios in our work.

The following tables show the characteristic properties of wood-based components and their bark. Typical components of Norway spruce are presented in *Table 7*, elemental analyses of different bark fuels (and wood fuel in general) are given in *Table 8*, and moisture contents in *Table 9*.

Typical lower heating values (LHV) on a dry basis per bark species used as a fuel (collected from different sources) are: pine bark 20 MJ/kg, spruce bark 18.6 MJ/kg, and birch bark 22.7 MJ/kg. These values are reported in Alakangas et al. (2016).

While pyrolysis experiments were performed for black pellets produced by steam explosion, bark samples were not pyrolysed during the project. In the case study assessments it was assumed that the biochar samples produced from SE black pellets in the experiments of the University of Oulu would be similar to the biochar received directly from bark. University of Oulu analysed the black pellet lower heating value on a dry basis to be 19.65 MJ/kg (higher heating value reported in Toloue Farrokh et al. (2019). This value was applied also to bark as the raw material (*Table 13*) in the subsequent scenario calculations. Respectively, the dry composition (elemental analysis) and ash content of bark-based biochar were assumed to be the same as those of biocarbon made from black pellets. Bark dry content was fixed at 40 wt%.

Table 7. Norway spruce components; values in brackets are deviations (Alakangas et al. 2016).

Component	Share, dry basis [wt%]
Cellulose:	26.6 (1.2)
Hemicellulose:	9.2 (1.1)
Lignin:	11.8 (0.9)
Extractives:	32.1 (3.8)

Table 8. Elemental analysis (wt%, dry basis) of wood fuel in general and different bark fuels from different sources (Alakangas et al. 2016).

Wood fuels	**C**	**H**	**N**	**O**	**S**	**Cl**
Wood, in general	48-50	6.0-6.5	0.5-2.3	38-42	0.05	<0.01
Pine bark *)	52.5-54.5	5.7-5.9	0.4-0.3	37.7-39.7	0-0.03	0-0.0085
Spruce bark *)	49.9-50.6	5.9	0.4-0.5	41.4-40.2	0-0.03	0-0.0279
Birch bark	56.6	6.8	0.8	34.2		

*) *Values from two different sources*

Table 9. Moisture contents of different bark fuels from different sources (Alakangas et al. 2016).

Wood fuels	**moisture [wt%]**
PULPWOOD BARK	
Softwood, dry transport	
- dry debarking	40-50
- wet debarking, uncompressed	60-70
- wet debarking, compressed	55-62
Softwood, wet transport or storage in water	
- uncompressed	70-85
- compressed	55-62
Birch	
- wet debarking, uncompressed	65-70
- wet debarking, compressed	55-62
- dry debarking	40-50
ROUNDWOOD BARK	
Softwood	
- dry treatment	40-50
- wet treatment	60-80
Birch	35-50

2.4.3 Black pellets

Pellet fuels can be defined as biofuels made from compressed organic matter or from different biomass, such as virgin wood, industrial woody side streams and waste, agricultural residues, and energy crops. Wood pellets, often called white pellets, are the most common type of pellet fuel and can be made, for example, from sawdust and from industrial woody side streams or co-products. The pellets are extremely dense and can be produced with a low moisture content (below 10%) enabling a very high combustion efficiency.

Heat treated wood pellets are referred to as black pellets due to their appearance after thermal treatment. Black pellets can be used, for example, as a substitute for coal in pulverized coal power plants. Advantages of black pellets include opportunity for outdoor storage, efficient transportation, good grindability, low dust formation and improved calorific value. Black pellets are generally converted from biomass by torrefaction or by steam explosion (SE). Torrefaction conversion is explained in *Subsection 2.3.* The torrefied biomass is further densified into pellets. In the SE method, biomass, or wood fibres, are exposed to saturated steam. Process parameters such as steam temperature and pressure and reaction time determine how much of the hemicelluloses degrade, and what fraction of the feedstock gets converted into volatiles and biochemical compounds. This is followed by a rapid release of pressure, i.e. steam explosion, upon which expansion of water in the cell walls causes the wood fibres to break down into very small particles. The steam exposure is controlled to minimize cellulose and lignin degradation while partially degrading the hemicelluloses. Lignin is caused to form small beads on the surface of the wood fibres. Finally, the material is pressed by a pellet press, whereupon the lignin beads form a coating on the wood fibres resulting in hard, highly water-resistant pellets that produce almost no fines (Strauss et al. 2018).

Valmet, a manufacturer of SE black pellets, claims that they are safer, obtain higher dry and wet durability, and hold more energy per tonne than conventional white pellets, and are thus more cost-effective. Even handing and loading/unloading SE black pellets in the rain is possible, which is not an option with white pellets. No chemical additives (e.g. binders for pellet product) are needed. The process can use low-grade and low-cost raw material such as bark from Kraft pulping or from sawmills, and efficient, modern Kraft pulp mills may not require all of their bark for internal use (Valmet 2015; 2017).

Valmet delivers a complete black pellet production facilities, from biomass infeed to black pellet outfeed. An example plant is visualized in *Figure 11*. Valmet also offers revamps of existing white pellet plants. The plant can be integrated with a packaged combined heat and power (CHP) system that provides steam for drying purposes and for steam explosion, as well as power for the plant, and often surplus power to sell to the grid (Valmet 2017).

In the Valmet process, the biomass material enters the system with a particle size equivalent to normal or micro-sized wood chips, and comes out almost as a powder. There is no need for further treatment between steam explosion and pelletizing. Torrefied black pellets, instead, would require additional densification in between. Biomass requires drying before feeding it from a buffer bin with plug screw into a pressurized reactor. The material is discharged from the reactor in a continuous infeed/outfeed loop. A blow valve and blow line deliver the material to the pelletizing step. The fuel for the CHP plant could be low-grade fuel, such as bark from debarking or other residues on the market, instead of burning the product, high-energy content black pellets, at the site (Valmet 2017).

In the present work, the focus has been on producing biochar from SE black pellets by slow pyrolysis. Samples of SE black pellets, converted from bark, were provided by Valmet for the project for further analysis and pyrolysis experiments, carried out at the University of Oulu. Analyses of the black pellet raw material can be found in Toloue Farrokh et al. (2019).

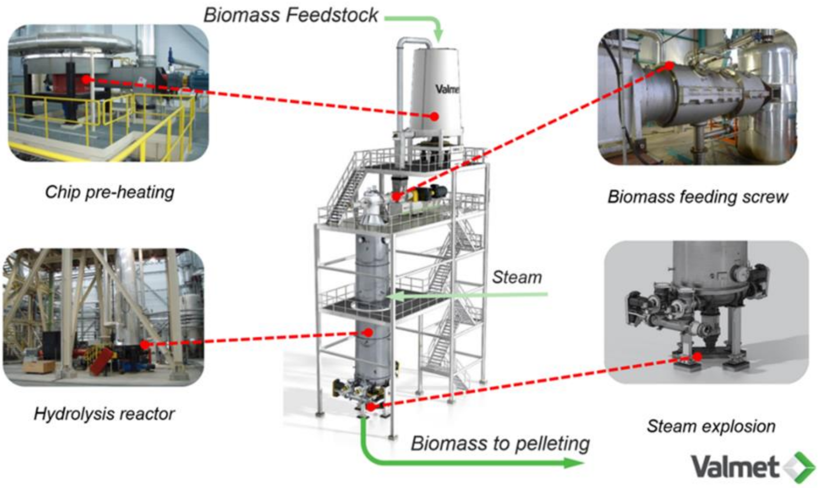


Figure 11. Black pellet plant by Valmet (2019). *Courtesy of Valmet*

2.5 Biomass handling and transportation before conversion

The dry content of hydrolysis lignin from the ethanol process may be as low as 30 wt%, depending on the conditions. Lignin is pressed to 50 wt% dry content in pellet form in all lignin scenarios. Further drying to 90 wt% is carried out in one scenario. Intermediate storage is needed at the ethanol production site and at the biochar production site if these locations are different. In one scenario, the ethanol and pyrolysis plants are at the same site, and the lignin pellets can be transported by a conveyor to the biochar production process.

Handling lignin in pellet form is less risky than in dry pulverized form, which presents an explosion risk. Lignin pellets are distinctively more hydrophobic than white pellets made from lignocellulosic biomass. However, with a moisture content of 50 wt%, or less, there is a risk of lignin pellet deterioration due to residues (including biological activity) from the ethanol production process; this presents challenges for the storage and transportation of the material and requires minimization of the time between drying and pyrolysis.

Bark is obtained from the debarking line of the pulp mill. The existing handling, conveying, and storage facilities as well as bark handling know-how of the working staff can be utilized. The biochar production site is assumed to be in close connection with the pulp mill.

Black pellets, converted from bark by steam explosion (SE), are also assumed to be produced at the pulp mill site, and bark from the pulp mill can be utilized as the raw material. The bark handling, conveying and storage facilities are present at the mill site, whereas the biochar production unit is assumed to be in a different location. Intermediate storage of black pellets is therefore needed in close connection to the pulp mill site and at the biochar production site.

The transportation needs for raw biomass will depend on the selected scenario evaluated in this work. Railway transport is assumed for both raw material and for the produced biochar.

2.6 Biomass upgrading for injection of pulverized coal into blast furnace

2.6.1 Biomass drying

To avoid thermal ballast and its effects on yield in pyrolysis, the drying step is necessary to achieve a biomass moisture content of no more than 10 wt%.

Lignin pellets, if delivered in moisture content of 50 wt%, and bark therefore require drying to a moisture content of less than or equal to 10 wt% before entering the slow pyrolysis unit. Black pellets are delivered at a moisture content of close to 10 wt%, so no pretreatment before pyrolysis is required.

The dryer can be stand-alone or integrated with an industrial plant, such as a pulp mill, district heating network, combined heat and power (CHP) plant, or other industrial operations that provide enough heat for biomass drying. Sources of heat could be, e.g.:

- exhaust gases, e.g. from furnace boiler / from combustion of pyrolysis side products or biomass residues
- high pressure (HP) steam from power plant
- low pressure (LP) steam, e.g. from CHP plant
- steam from combustion of, e.g., pyrolysis side products / biomass residues
- warm air, e.g. from air-cooling of surplus hot process water or from air-cooled condenser

Different drying methods with a variety of heat sources can be used in connection with thermochemical conversion processes. Several dryer types for biomass exist, such as the band dryer, rotary dryer, steam rotary dryer, pressurized fluid-bed dryer and pneumatic steam dryer. Key aspects regarding the selection of the dryer include the capacity requirement, available heat source(s), available heat temperature level, and the drying medium. Performance data for different dryers used for biomass are presented in *Table 10* (Fagernäs et al. 2010).

Table 10. Performance data for different dryers for biomass (Fagernäs et al. 2010).

	Band dryer	**Rotary dryer**	**Steam rotary dryer**	**Pressurized fluid-bed dryer**	**Pneumatic steam dryer**
Biomass feed	Sawdust, wood shavings, wood chips	Sawdust, wood chips, bark	Sawdust, wood processing	Wood chips	Sawdust, bark, forest residues
Dry feed [tonnes/h]	8-9	6-7	5-6	9	25
Moisture [wt%] Inlet / Outlet	50-60 / 10-15	50-60 / 10-15	50-60 / 10-15	50-60 / 10-15	50-60 / 10-15
Drying medium	Air, flue gas (90-120 °C)	Air, flue gas (250-400 °C)	Steam -ambient pressure	Recycled LP steam (3-4 bar)	Recycled LP steam (3-4 bar)
Heating medium	Hot air	Flue gas	HP steam (6-10 bar)	HP steam (26 bar)	HP steam (7-26 bar)
Capacity [tonnes of H_2O/h]	10	7-8	6-7	5-40	25
Evaporation energy demand [MJ/kg of H_2O]	4-5	4-5	3-4 (With energy recovery: 0.8-1)		2-3 (With energy recovery: 0.5-0.7)

The widely used unit for biomass feedstocks is the atmospheric band conveyor dryer, also called the belt dryer. The drying medium, hot air or clean enough flue gas, is blown through a thin layer of biomass on a horizontally moving permeable band. The dryer can be a single-stage single-pass, multi-stage single-pass or multi-pass design. Dryer types that can utilize low pressure steam or even hot water as a heating medium to heat up the drying air (drying medium) are beneficial. Band conveyor dryers are easy to control (moisture content, maximum temperature, residence time) and, due to the thin layer of biomass on the band, the drying uniformity is good. The main drawback of band conveyors is their extensive space requirement.

The band conveyor dryer was selected as the basis for the scenario calculations presented in this work due to its versatility and possibility to utilize low temperature heating media. According to SWISS COMBI (2019), their belt dryer can utilize different hot water streams in cascade mode starting from temperature levels as low as 50 °C up to 120 °C. Another possibility is to utilize saturated steam at pressure levels between 1.5 - 4 bar(a). According to Hannula & Kurkela (2013), the specific energy consumption is ca. 1 100 kWh (4 MJ) per tonne of H_2O evaporated, which is in line with the evaporation energy demand by Fagernäs et al. (2010) shown in *Table 10*. This energy consumption was used as the basis in the scenario calculations.

The operating principle of the band conveyor dryer is presented in *Figure 12* (Fagernäs et al. 2010; Bennett 2011).

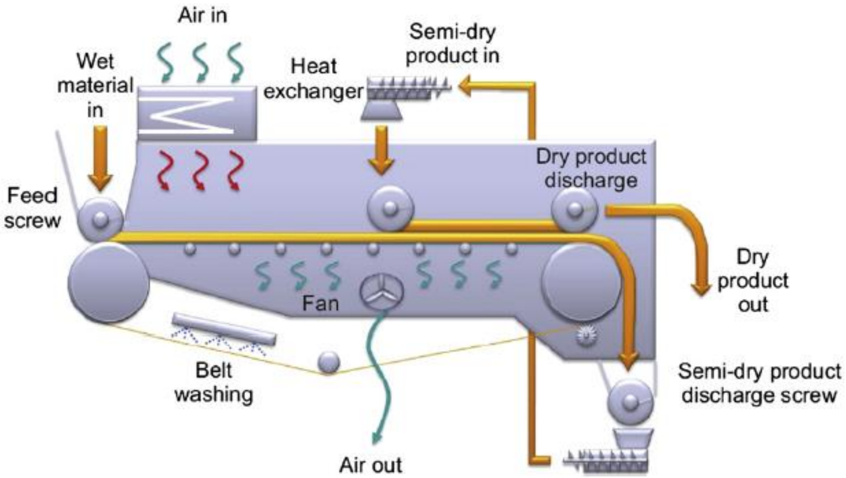


Figure 12. KUVO belt dryer for biomass drying by Metso (Fagernäs et al. 2010; Bennett 2011).

2.6.2 Slow pyrolysis

The biomaterial is led through grinding and feeding to the pyrolysis unit at a moisture content of ca. 10 wt%. In slow pyrolysis, gradual heating to 300-600 °C with a residence time from 15 minutes to several hours takes place in absence of air (oxygen) (*Table 6*).

Many different designs of industrial charcoal pyrolysis unit exist. The charcoal kiln has conventionally been the most common design. Today, the most popular unit is based on a retort design. The different names given to charcoal kilns or retorts are based on slight variations in design, but the principles of operation are similar (Tutturen 2013). The commercial processes typically operate near atmospheric pressure, and most units apply wood as the feedstock. The main principles of charcoal production include internal heating, external heating, and heating of the recirculated gas (Antal et al. 2003).

Internal heating is employed in Missouri kilns (Argentine and Brazilian 'beehive' kilns). These processes apply partial combustion of the feedstock. The rectangular and poured concrete structured Missouri kiln is filled with wood by front-end loaders. Air flow into the kiln is regulated, and afterburners take care of the emissions. Many Missouri kilns have, however, been unable to meet emissions regulations and have ceased to operate (Antal et al. 2003).

External heating is employed, for example, in the Van Marion retort (VMR), which consists of two chambers for interchangeable cylinders and a central combustion chamber. The chambers operate in a cyclic manner, where combustible vapours from the pyrolysis in one cylinder are burned and the heat generated is delivered to the other cylinder, filled with fresh wood, to dry and initiate the pyrolysis reactions. The cylinder full of hot charcoal is removed, and replaced with a new cylinder filled with fresh wood. The airtight cylinder is left to cool before unloading the charcoal. VMR is of an old design and does not exist in the market anymore (Antal et al. 2003; Tutturen 2013).

In the Reichert process, a batch-fed retort, heating is based on a recirculated gas principle. The retort represents the first successful attempt to transmit heat to the charge inside the retort without having to heat through the metal walls of the retort itself (FAO 1985). The heat is transferred to the wood by blowing recirculated heated inert or fuel gas (e.g. non-condensable pyrolysis gases) through the charge inside the retort. Flue gas could, in principle, also be used although this would dilute the excess pyrolysis gases, which are to be burned to generate process heat (FAO 1985). One solution is that the hot pyrolysis gases generated are condensed and the remaining non-condensable gases are heated and returned to the retort; excess gases are burned to heat up the gases returned to the retort and to pre-dry the feed (Antal et al. 2003). In the Reichert process, the wood particles must be small enough and yet not too small to impede the gas circulation. When carbonization is complete the bottom of the retort is opened and the charcoal is dropped into closed steel containers, where cooling takes place. If required, rapid cooling can be achieved by passing cold inert gas through the retort (FAO 1985).

FAO (1985) claimed the Lambiotte or SIFIC process, originally developed in the early 1940s, to be perhaps the most successful technology for the continuous carbonization of slab and roundwood to produce conventional lump charcoal useable for all purposes. The pre-dried wood is lifted (by e.g. conveyor) to the top of the retort and is dumped into a double bell gate allowing the wood to enter the retort while preventing significant escape of retort gases. As the wood drops slowly downwards through the retort, a counter-current flow of inert hot gas dries the wood and raises it to the carbonizing temperature. Small charcoal batches are removed from the base of the retort through a pair of interlocking gates. The pass through can be controlled by the operator to suit the moisture content of the wood and fixed carbon in the charcoal. The percentage of fines formation is about the same as charcoal produced in conventional brick kilns. Charcoal cooling is necessary due to the contact with air. This is done by blowing cool inert or fuel gas into the bottom of the retort. This gas heats and rises in the retort. The heated gas is drawn off at the middle of the retort just below the point where the hot gas for converting the wood to charcoal is blown in. The hot gas is usually produced by burning combustible gas and the resulting flue gas is blown into the retort just above the exit point of the cooling gas stream. The gas passes up the retort, heating up the charge. The gas released from the head of the retort is set at a sufficient temperature level to prevent condensation. Hot pyrolysis gases are partly burned to provide hot inert gas for heating the charge and the rest is cooled and scrubbed to remove tar and passed in at the base to cool the charcoal. This gas stream is mixed with the rest of the gas and burned to produce the hot inert gas. The large retorts at the Wundowie ironworks in Australia, for example, burn the off-gases for power generation and use clean cold blast furnace gas as the cooling and heating medium for the retorts. The process has, however, a notably high investment cost (FAO 1985).

A smaller CISR-Lambiotte retort has been developed to simplify the original Lambiotte process. This retort is heated by hot inert gas from burning part of the recycled retort gases and vapours. This provides enough energy to complete the drying of the wood, raise it to spontaneous decomposition temperature, drive-off surplus tar trapped in the structure of the charcoal, and cover the heat losses from the retort. The remaining retort gas is heated by cooling the charcoal, and again mixed with the rest of the retort gas to be burned. The moisture content of the wood entering the retort must be ca. 30% or less. The carbonization temperature is determined by the quality requirements of the charcoal. Wood feed is typically ca. 7 000 tonnes per year (FAO 1985). There are several CISR carbonization plants operating in Europe (Vamvuka 2011).

Lambiotte and Cie (2019) have developed a unique carbonization technology, the automatic continuous carbonization retort, type SIFIC/CISR, capable of producing 2 000 – 6 000 tonnes of charcoal per year. According to Lambiotte and Cie, the process has a very high production yield. Pyrolysis gases are burnt inside the retort without the need to burn wood or charcoal. In this process, wood is dried in an initial zone at the top of the retort, after which it undergoes carbonization at controlled temperatures in a second zone. The charcoal is cooled in the carbonization retort by cooled gas and discharged at ambient temperature. The charcoal is said to be pure and highly homogeneous with a very high carbon content (80-90% fixed carbon), a moisture content of ca. 3 to 4%, and maximum volatile matter content of 12%.

A similar concept has been applied by Lurgi, which forms part of the Silicon Metal Complex in Bunbury, Western Australia, producing ca. 27 000 tonnes of charcoal per year from local hardwood, in two retorts (Vamvuka 2011). Outokumpu Technology (now Outotec) acquired Lurgi Metallurgie in 2001. A schematic of the Lurgi process is described in *Figure 13*.

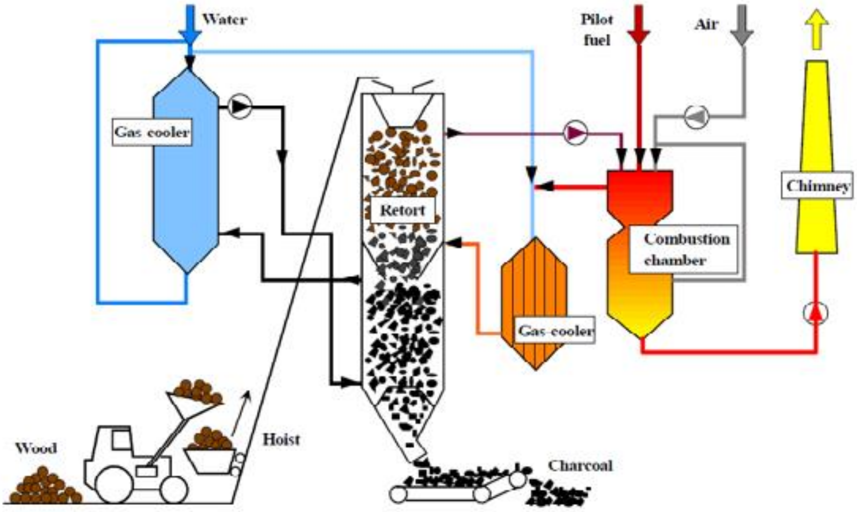


Figure 13. Lurgi process for producing charcoal (Sun et al. 2011).

The rotary hearth furnace, also known as the Herreshoff roaster, developed in the metallurgical industry for roasting sulphide ores, is a proven method for carbonizing small particle size wood and bark. The charcoal is produced in powdered form. The furnace consists of four to six circular refractory hearths stacked one above the other. Each hearth has a central hole through which passes a hollow shaft and a set of rabble arms are fixed to each hearth. These arms are hollow, enabling the whole raking system to be cooled by blowing air through it. The ploughs attached to the arms turn the feedstock over and move it across the hearths. The material slowly moves through the system while being constantly turned over to be exposed to the combustion air passing through the furnace from the bottom to the top. Once the furnace is lit it must operate continuously. The rate of air is regulated so that the wood carbonizes and leaves the furnace as fine charcoal. Gases emerging from the top of the furnace can be burned directly. The charcoal leaving the furnace is cooled by passing the charcoal slowly through a horizontal steel cylinder, the walls of which are externally cooled by a water spray. The capacity of wood or other residues in the larger units is ca. ten tonnes dry residue per hour. The resulting fine charcoal needs to be pelletized, resulting in extra costs. Herreshoff roasters have proven successful when attached to large saw and plywood mills (in southern USA), having an adequate raw material supply, and a market for barbecue briquettes (FAO 1985).

Weaver (2011) reviewed the Pyrec process as part of the IEA Bioenergy ExCo66 Workshop for soil improvement applications. The concept of an inclined gasifier with a double screw was designed in 2005, and the first full-scale plant started operation in Switzerland in 2009. For small-scale applications the Pyrec 500 plant has been developed (*Figure 14*). The Pyrec 500 has an annual mass flow rate capacity of 1 500 tonnes of dry solids. Slow pyrolysis takes place at temperatures between 350-500°C. For small-scale units it is recommended to use white pellets to avoid emission problems. The produced charcoal has a fine pore structure.

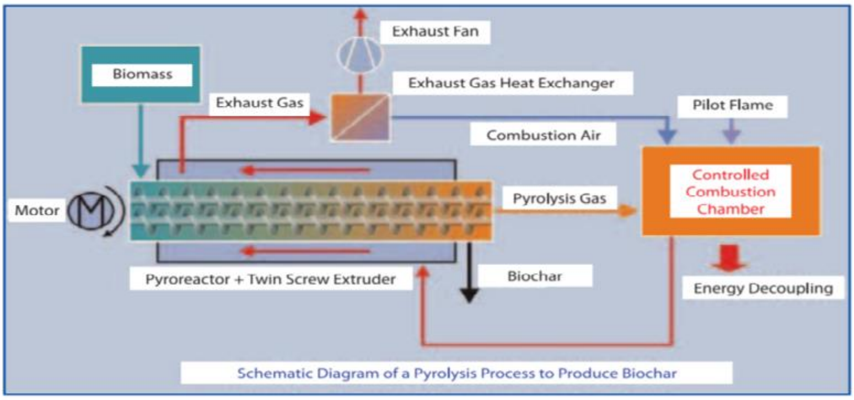


Figure 14. Schematic of a Pyrec 500 plant (Weaver 2011).

Cooksey et al. (2016) reported an auto-thermal pyrolysis technology piloting unit targeted at biomass-derived charcoal for metal production. It is designed, constructed and commissioned at CSIRO's laboratories (Melbourne). The pilot plant has been operating since 2013 and has a capacity of 1 000 tonnes of charcoal per year. It is designed for processing small-sized feedstock, such as wood waste or forest residues, in continuous operation (in pilot phase up to 4 hours continuous operation, targeting >24 hours of operation). The pyrolysis gas and condensate by-products are not diluted. There are no heat transfer limitations on scaling up the reactor, since no supply of external heat to the material is needed other than low-grade heat for drying the feed material. A schematic of the process is visualized in *Figure 15*.

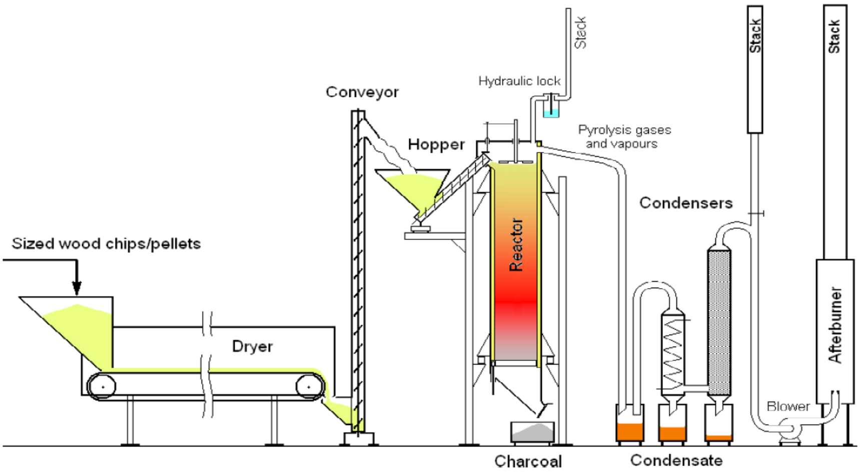


Figure 15. Process diagram of CSIRO auto-thermal pyrolysis technology (Cooksey et al. 2016).

In this work no specific pyrolysis technology was assumed for the subsequent heat and mass balance scenarios. Low-grade heat (hot water and low pressure steam) is used for drying the feedstock and hot pyrolysis gases are assumed to be partially burned to provide the necessary heat for maintaining the pyrolysis reactions. The remaining hot pyrolysis gases are further burned to provide steam for the steam turbines generating electricity and for drying in the applicable scenarios, or used as a fuel gas to replace other fuels, for example in the lime kiln. Biochar is cooled with an applicable system and pelletized for transportation and handling.

Feed-related technical issues need to be evaluated further, such as how the lignin feed would behave in the pyrolysis process in question. Even if the lignin is pelletized, feed-specific challenges may arise (e.g. melting and sticking to surfaces). Additionally, the suitability of the technology for necessary upscaling and for meeting the charcoal quality requirements needed to replace PCI coal need to be verified.

2.6.3 Handling and pulverizing of biochar

For scenarios where biochar is produced at the iron and steel manufacturing site, no off-site transportation or preprocessing of the biochar are needed.

If the pyrolysis site is located elsewhere, the biochar must first be pelletized for transportation. Binder(s) may be needed to increase the durability of the biochar pellets. One option could be to utilize the condensable constituents (tar) containing fraction of the pyrolysis gases as binder components. Intermediate storage and loading systems for the pellets are needed at the pulp or pulp and paper mill site.

At the iron and steel manufacturing site, biochar pellets are unloaded from the train to intermediate storage and further transported to biochar pellet silos situated in connection with a PCI coal grinding plant, where coal is pulverized by grinding.

The coal handling, pulverizing and injection systems (*Figure 16*) are assumed to be also compatible with biochar, with minor adjustments and modifications where required.

Before starting the operation, the grinding plant is inertized and heated up by process gas, which could be nitrogen or carbon dioxide. Some amount of oxygen must be present to lower the dew point temperature. Typical oxygen content in the process gas is between 8% and 10%. Process gas is heated by a burner. The temperature profile inside the system may vary, but should not be lower than 65 °C. After the operation is started, additional inert gas is no longer fed to the plant, other than to the fine coal/biochar silo for safety reasons. Process gas dries the pulverized coal/biochar particles in the system. The pulverizer units also contain classifiers that return any oversized particles back to the grinding area, which consists of grinding plates and rollers. Particles follow the process gas flow. After the classifiers, the accepted particles are screened and transferred to a fine coal/biochar silo (Loesche-Group 2013). The stream is then divided into multiple injection vessels and pressurized by inert gas. The injection system piping leads to the tuyeres, where the pulverized coal/biochar is injected through a lance to the blow pipe, and further to the blast furnace.

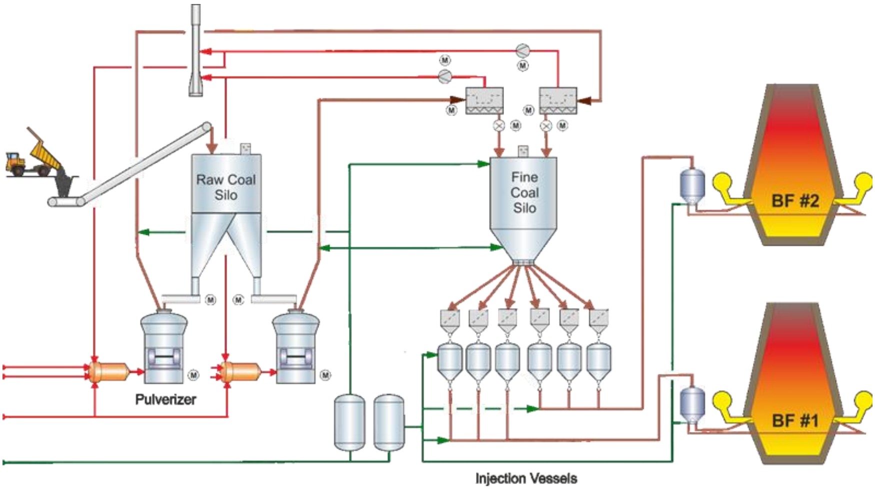


Figure 16. Handling, pulverizing and injection system of coal by SSAB Raahe (2019b). Courtesy of SSAB

Biochar handling, pulverizing and feeding into the blast furnace are not considered in the scenarios. The PCI plant is adopted as-is and assumed to be applicable also for biochar with the required adjustments and possible minor modifications.

Different parameters are also considered in Toloue Farrokh et al. (2019) for storing biochar for applications like blast furnace injection:

- Pyrolysis improves the biomass behaviour in material handling and storage since it increases bulk density and hydrophobicity.
- A good storage should ensure safe, low-cost storage with minimum effect on the physical and chemical properties of the stored material.
- Particle size and density are important parameters. Low density materials occupy more space affecting the storage area required.
- In case of high share of fine particles, the dust formation in the plant area will be a problem. In windy conditions, fine particles with low density are spread easily. Moisture increase of the stored materials may be needed to avoid this effect. On the other hand, moisture at sub-zero conditions may cause problems in silos, when frozen and unfrozen raw material get together.
- As a rule of thumb when the share of fines (particle size of less than 1mm) is more than 30% the material flow inside the silo would cause issues.
- Ignitibility of stored material should be considered to prevent the risks.

Toloue Farrokh et al. also evaluated the suitability of lignin and black pellet chars for blast furnace injection and compared these to fossil-based reductants that are currently used. They took into consideration also the effects of raw biomass in the grinding process, which can be found from their report. Here, the focus is on biochar:

- Coal is denser than the biomass, or even charcoal. When the material is grinded, the powdered material bulk density difference will increase, affecting the flow design and storage in injection silos of the blast furnace.
- Pulverized coals (PC) have varying chemical properties depending on the coal source. The typical carbon content of fossil coal is 80–90%, ash content 10% and the heating value of the pulverized coals is around 30 MJ/kg.
- The chemical properties of biochars produced from wood-based feedstock depend on the end-temperature of the heat treatment. As the end-temperature increases, the heating value of the pyrolyzed biomass approaches the heating value of pulverized coal.

- The important property of biochar powder is its permeability, which can be used to estimate the effectiveness of pneumatic transfer, where particles are separated and carried by the carrying gas. The higher the permeability is the less material is transferred by the carrying gas. Thus, low or moderate permeability values are favoured. The permeability of the lignin char (pyrolysis at 500°C and 650°C) and black pellet char (at 500°C) samples was quite similar and close to PC values. For lignin char with pyrolysis temperature of 300°C, the permeability was significantly higher, leading to a more cohesive behaviour.
- Lignin samples (at 300°C and 650°C) displayed an easy-flowing property and the lignin and black pellet samples at 500°C were classified as a free-flowing powder. To mention, extreme free-flowing property of a powder may not be considered desirable as it may cause problems in the transfer line equipment (more details in Toloue Farrokh et al).

3. Methods

3.1 Experimental proof and arrangements -Bio-reducer production

The raw materials, applied methods, and experimental arrangements carried out by the University of Oulu are presented in detail in Toloue Farrokh et al. (2019).

Hydrolysis lignin and black pellets were selected for use as raw materials as well as a fossil coal sample as a reference material.

The following experiments were carried out as described in Toloue Farrokh et al. (2019):

- Slow pyrolysis
- Density measurement
- Water uptake
- Surface and pore analysis
- Flow behaviour of the fuels
- XPS
- Combustibility
- Drop Tube Furnace combustion
- CO_2 reactivity test

3.2 Thermochemical modelling of blast furnace

Dynamic modelling of the blast furnace was used to study the chemical reactions in the furnace when using different bio-based materials to replace part of the pulverized coal injected to tuyeres (thermodynamic simulation of selected pyrometallurgical bioreducing operations). As an additional goal, the time-dependent thermochemical behaviour of the reaction zones, such as the cohesive zone in the furnace, were also modelled.

3.2.1 Blast Furnace Model

The blast furnace model is based on the CROM simulator first developed for the submerged arc furnace (SAF) in the SIMP project (ferrochromium production at Outokumpu Tornio steel mill) (Penttilä 2017). The first version of CROM was a steady-state flowsheet simulator with simplified process models (one or two calculation steps per process). The model was later updated as a dynamic, one-dimensional counter-current process model for the preheater and smelter part of the SAF process (with a total of 32 calculation nodes in the axial direction). To be applicable to the blast furnace, the CROM simulator was updated to a dynamic procedure with a new particle model supporting multiple particles and particle size classes. The flowsheet supports multiple process units (called blocks) and multiple flows (called connections) between the blocks. The flowsheet solver is a so-called sequential modular solver, i.e. the process units must be solved in a certain order (partitioning and sequencing of the flowsheet). This is necessary as the number of unknown variables (phase constituents) could be hundreds, and using an equation-based solver was not an option. The solver can also handle multiple recycle flows by identifying so-called tear flows. The temperature and composition of the tear flow must be iterated at each time step. Each process can have its own time steps, in which case the simulator is responsible for calling the process units in proper order and with proper time steps. The simulated time duration can be from seconds to days. As a simple option, the blast furnace flowsheet only contains a charge feeder and the blast furnace, without any recycle flows.

The CROM simulator was programmed using Intel Visual Fortran with Microsoft Visual Studio 2015. It is a command prompt program without a user interface. Input values are transferred via files. A graphical user interface for viewing the simulation results and changing the input values while the simulation is running is under development.

3.2.2 Thermodynamic system

A thermodynamic system consists of a number of phases. The phases are divided into three groups – gaseous, mixture, and invariant (also known as pure) phases. Each phase has one or more constituents. These phase constituents have compositions expressed as amounts of components. A component is a system-wide entity. Usually, components are elements, but it is also possible for them to be stoichiometric combinations of elements. A thermodynamic system also contains relevant thermodynamic data, such as Gibbs energies and enthalpies, entropies and heat capacities for all the phases and phase constituents (usually defined as a set of temperature-dependent Gibbs energy equations). The thermodynamic system can be compiled from thermodynamic databases such as FactSage (Bale et. al. 2002).

In the blast furnace, the solid feeds consist of particles such as lumpy ore, pellets, limestone and coke. Ore is a mineral containing mainly chromium, iron, aluminium, magnesium in the form of various oxide phases (Fe_2O_3(s), Fe_3O_4(s), various silicates, etc.). Pellets are sintered concentrates prepared from the lumpy ore. Limestone contains mainly calcite ($CaCO_3$(s)) and is used for slag forming with silica (fluxing). Coke contains mainly carbon (C(s)), moisture (H_2O(l)) and ash (various aluminosilicates and other oxide phases) and is used for the reduction of metal oxides to metals.

Each particle has its own unique phase composition and can be handled as a separate thermodynamic system. However, all of the particles can also be combined together as one system, i.e. a bulk system. This greatly simplifies the calculation and makes it faster, but also makes it more difficult to control the physical interaction between the particles and their phases.

The elements included in the blast furnace model are: Al, C, Ca, Cr, Fe, H, K, Mg, Mn, N, Na, Ni, O, P, S, Si. Phases included are: ideal gas, liquid steel, austenite, ferrite, various carbides (cementite, M7C3), liquid oxide (slag) and silicate solutions (olivine, orthopyroxene) and a number of pure condensed hydroxides, carbonates, sulphates and oxides (Al_2O_3(s), SiO_2(s), etc.). The phases were selected from the FactPS (FactSage pure substances database), FToxid (oxide database for slags, glasses, ceramics, and refractories) and FSstel (steel database) databases in FactSage. In total, the model contains are around 200 phases and more than 300 phase constituents.

The equilibrium composition of the thermodynamic system can be calculated with ChemApp (Petersen and Hack 2007). ChemApp a is thermodynamic programming library for C, Fortran, Pascal, Visual Basic and other languages. In this project, the Intel Visual Fortran compiler with Visual Studio 2015 was used.

3.2.3 Blast furnace reaction zones

Near the bottom of the furnace is the active coke zone, where coke and air react to produce red-hot coals (*Figure 17*). Carbon is in excess at this point and throughout the furnace. Hence, the principal product of combustion is carbon monoxide. The conversion of iron ore to metallic iron takes place in the reduction zone. The metallic iron produced enters the fusion (cohesive) zone where temperatures are sufficiently high to melt it. The molten material percolates through the active coke and stagnant coke zones and eventually collects in the bottom of the hearth, where it is periodically tapped off as pig iron for further processing into steel. The limestone in the charge decomposes into calcium oxide and carbon dioxide as it passes through the reduction zone.

The calcium oxide combines with silicate impurities present in the iron ore to produce molten slag in the fusion (cohesive) zone. The slag drips through the coke and collects as less dense liquid in the hearth (Treptow & Jean 1998).

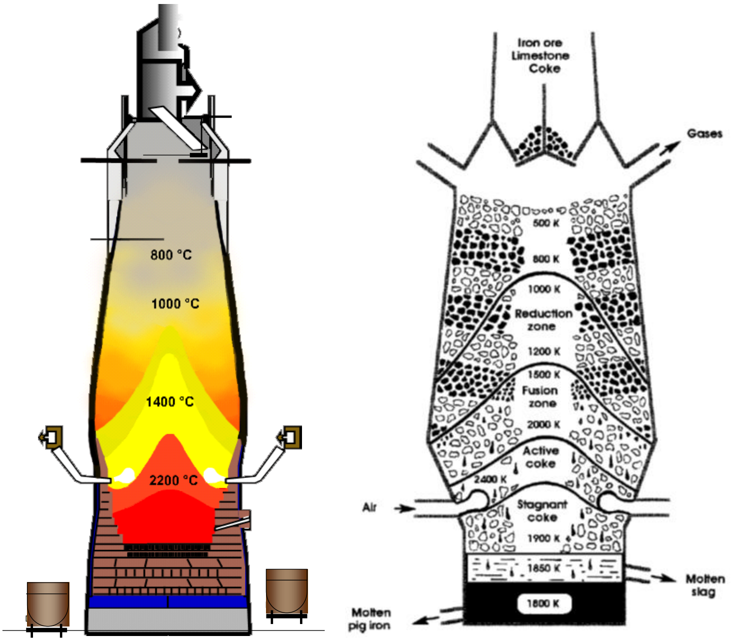


Figure 17. Blast furnace reaction zones.

The carbon monoxide produced in the active coke zone rises through the furnace and comes into contact with the ore. The reduction sequence involves three basic steps (iron used as an example):

$$3Fe_2O_3(s) + CO(g) \rightarrow 2Fe_3O_4(s) + CO_2(g) \tag{3.2.1}$$

$$Fe_3O_4(s) + CO(g) \rightarrow 3FeO(s) + CO_2(g) \tag{3.2.2}$$

$$FeO(s,l) + CO(g) \rightarrow Fe(s,l) + CO_2(g) \tag{3.2.3}$$

Instead of calculating these reactions by using their reaction stoichiometries with reaction rate equations as the mechanistic models do, a more general method is used. All reactions in the furnace are calculated by combining kinetics with equilibrium calculation using the Gibbs energy minimization method. Here, equilibrium calculation determines the reaction products but the kinetics determines the rate at which these reactions can take place. With this method, one does not need to know exact reaction paths, but it is crucial to include all the relevant phases in the thermodynamic system. As the reacting phases in the blast furnace are contained within relatively large particles, the solid-solid and gas-solid kinetics play an important role in constraining the rate of overall reactions.

3.2.4 Reaction kinetics

As mentioned above, the reactions in the blast furnace model are calculated using the Gibbs energy minimization method, which gives the equilibrium composition, but the fraction of the phases taking part in this equilibrium calculation (reactive fraction) are in turn calculated with time-dependent kinetic equations. Other fractions of the phases are considered inert (inert fraction). Finally, the equilibrium composition of the reactive phases and the inert fractions are combined to yield the current local phase composition. *Table 11* shows the included solid-solid kinetic models.

Table 11. Solid-solid kinetic models (Khawam & Flanagan 2006).

Kinetic model		***f*(α)**	
Power law	P2	$2\alpha^{1/2}$	
Power law	P3	$3\alpha^{2/3}$	
Power law	P4	$4\alpha^{3/4}$	
Avrami-Erofeyev	A2	$2(1-\alpha)[-\ln(1-\alpha)]^{1/2}$	
Avrami-Erofeyev	A3	$3(1-\alpha)[-\ln(1-\alpha)]^{2/3}$	
Avrami-Erofeyev	A4	$4(1-\alpha)[-\ln(1-\alpha)]^{3/4}$	
Contracting area	R2	$2(1-\alpha)^{1/2}$	$k' = k/R$
Contracting volume	R3	$3(1-\alpha)^{2/3}$	$k' = k/R$
1-D diffusion	D1	$1/2\alpha$	$k' = k/R^2$
2-D diffusion	D2	$-1/\ln(1-\alpha)$	$k' = k/R^2$
3-D diffusion	D3	$3(1-\alpha)^{2/3}/[2(1-(1-\alpha)^{2/3})]$	$k' = k/R^2$
First-order	F1	$(1-\alpha)$	
Second-order	F2	$(1-\alpha)^2$	
Third-order	F3	$(1-\alpha)^3$	

Variable α in the kinetic model is the conversion of the reactant phase to the product phase:

$$\alpha = 1 - \frac{c_t}{c_0} \tag{3.2.4}$$

where c_0 and c_t are the initial and current mass concentrations of the reactant phase. Normally, the initial concentration of the phase is same as its concentration in the charge, but as any phase could take part in multiple reactions as reactant and/or product, then the initial concentration is taken as the maximum concentration of the phase at any given time and position in the furnace. The conversion as a function of time is calculated as:

$$\frac{d\alpha}{dt} = kf(\alpha) = A_0 e^{-E_a/RT} f(\alpha) \tag{3.2.5}$$

where A_0 and E_a are the frequency factor and the activation energy of the Arrhenius equation, and T is the temperature and R is the gas constant. For certain models the rate constant k is dependent on the geometry, i.e. the particle size. For example, $k' = k / R^2$, where R is the radius of the particle.

3.2.5 Particle model

Many different particles can exist in the blast furnace: lumpy ore, sintered pellets and metallurgical coke are the most common. The particles are usually a few centimetres in diameter on average. Each particle has its own particle size distribution (PSD) and phase composition that could even vary from size to size as reactions are faster with smaller particles (if the reaction rate constant of the kinetic model is dependent on the geometry). A general particle model has been implemented where kinetically constrained equilibrium reactions can be calculated separately for each particle and particle size class in the material. There might be only one particle with one particle size class (equivalent to a bulk system), or there can be as many as five different particles each with fifteen different size classes (all particles must have the same size classes, i.e. the same lower and upper size boundaries).

The gas phase taking part in the constrained equilibrium calculation (determined by the gas-solid mass transfer equation) is divided between the particles and their size classes based on the fractions of their effective surface areas. This enables indirect reduction reactions in the blast furnace between the CO gas and the metal oxides. Other important part of the particle model is the particle-particle interaction, i.e. sintering and the formation of the melt phases at higher temperatures (liquid steel and oxide slag). This also enables direct reduction reactions between the solid carbon and the metal oxides (between coke and ore/pellet particles). The sintering model determines the fractions of particles and their size classes that are first mixed and then redistributed between the particles in a certain way. Usually this is done so that maximum size of the last particle is selected as the target particle size; its mass fraction starts to increase as others decrease (meaning that the particles are fused together as one). Formed liquid phases are also divided between the particles in a similar way to the gas phase.

3.2.6 Enthalpy balance

Solving the kinetically constrained equilibrium of the particle system involves solving the enthalpy balance between the initial and final phase compositions (states) of the particles. An iteration routine was implemented where the temperature of the final state is solved with a root-finding algorithm combining binary search and false position methods, and interpolation around the discontinuity points (enthalpy function is discontinuous at the phase transition temperature and thus the phase composition needs to be interpolated between the two compositions around the discontinuity).

Parameters of the enthalpy balance routine include initial phase compositions of the particle sizes, time step and the enthalpy change (i.e. the net heat into the particle system). The reactive fractions of the phases for each particle size are first calculated at a given temperature, and then the equilibrium compositions are calculated between these reactive fractions. Finally, the equilibrium compositions are combined with inert fractions and the enthalpies of the final phase compositions of the particles are calculated. The temperature iteration is continued until the final enthalpy of the particle system equals the initial enthalpy plus the enthalpy change. Only five to ten iterations are required at lower temperatures where the reactions do not take yet place or are still very slow (enthalpy change is close to linear). In contrast, more than twenty iterations may be required at higher temperatures where many endo- or exothermic reactions can take place (enthalpy change is nonlinear with one or many discontinuities around the solution temperature).

3.2.7 Blast furnace calculation grid

The blast furnace is divided into calculation nodes in the axial and radial directions (forming a two-dimensional calculation grid). Each calculation node contains material comprising one or more particles, a gas and two liquid phases (liquid metal and oxide slag). Each particle, the gas and the liquid phases have calculated densities and volume fractions in the node. Particles have intra-particle voidage, i.e. porosity, and inter-particle voidage, i.e. the void between particles. The porosity of a particle is dependent on the state of the particle reactions (decomposition reactions increases porosity due to weight loss). Voidage is generally considered constant by the model, but it can be a function of particle size distribution (as each size class can have its own voidage). Formed liquid phases fill the voidage (wetting), thus reducing the void volume fraction, especially when flowing down to a node below.

Each node has its own particle, liquid and gas temperatures and masses of the particles, gas and two solids at the start of each new time step. Calculation starts from the bottom row of nodes. First, the net heat into a calculation node is calculated during the time step (from heat transfer equations between the adjoining nodes and furnace wall). Then, any gases ascending from nodes below are added (except for the bottom row). Next, the enthalpy balance of the node is solved by iterating its temperature at the same time as the kinetically constrained equilibrium reactions for the particles in the node are calculated (also dependent on the initially unknown temperature). The same procedure is then repeated for the other rows of nodes by updating the gas flow and distributing it between the nodes in the radial direction (determined by the pressure change in each direction).

After solving the new temperatures and phase compositions in each node, the volumes of the material in the nodes are calculated starting from the bottom row. The volume changes then determine the volume of material flowing down from the nodes above. Any liquid phase present in the node above fills the voidage between the remaining particles, and heavier liquid metal replaces the slag, if present (causing the slag to float on top of the liquid metal). Thus, the material slowly descends in the furnace at the same rate as the volume changes due to melting and periodical tapping.

New material is charged to the top row of nodes by varying the ore-coke ratio from the centre node to the wall node. Charging is done so that the material surface level remains roughly constant.

Also a steady-state version of blast furnace has been implemented. In the steady-state model, the furnace can be divided into up to three radial and 30 axial calculation nodes for both the material bed and the gas sides. By default, each radial node has same cross-sectional area and volume. The gas fills the void between the solid particles and the formed liquid phases. The charging and the tabbing of the reactor are now continuous processes. The local volume elements (nodes) of the bed and the gas are described as open thermochemical systems, which transforms heat and mass with each other. The equilibrium states of the volumes are calculated by minimizing their Gibbs energy and by taking into account the heat and mass transfer between the volumes and the surroundings (currently only estimated heat loss). The time-dependent reactions in the material bed and the gas flow are taken into account in the Gibbs energy minimization by constraining the fractions of phases taking part in the equilibrium calculation (e.g. inert and reactive parts) as described in the earlier chapter.

When calculating the material bed side the temperatures and the mass flow rates of the gas side are not known. Problem is now a so-called two point boundary value problem with a fixed boundary. With the blast furnace model, its solution comprises an iterative calculation where first the mass and energy balance equations are solved for the material bed side by keeping the gas side temperature profile constant. After solving the bed side the gas side is calculated this time by keeping the bed side temperatures as constant. This is repeated until the temperature profiles converge (usually about ten iterations are needed).

3.3 Production scenarios

The evaluated scenarios are divided according to (i) stand-alone and (ii) pulp mill integrated production of biochar.

Stand-alone production refers to the selected scenarios where the biochar pyrolysis unit is located on the site of a steel/iron plant on the north-west coast of Finland. These scenarios are L1 (Lignin 1st scenario), L2 (Lignin 2nd scenario) and BP (Black Pellets scenario). The raw materials (hydrolysis lignin or black pellets) are transported to the production site from two separate locations in eastern Finland. The biochar manufacturing process is not integrated with the steel plant itself, although the produced biochar is used on site to replace part of the PCI coal injected in the blast furnace. The pyrolysis gases are burned to provide steam. The foremost use of the steam is drying the raw biomass, while the surplus of steam is valued by setting the electricity generation yield to 35% of this available heat load (of surplus of steam). The power plant itself is not included in the scenario estimation.

Pulp mill integrated biochar production refers to the selected scenarios where the production of biochar takes place at the pulp mill, utilizing integration possibilities. These scenarios are L3 (Lignin 3rd scenario) and Ba (Bark scenario). The raw material, hydrolysis lignin or bark, is available on site at the pulp mill. The locations of these typical Nordic Kraft pulp mills are on the west coast of Finland and in eastern Finland. The data from the pulp mills considered are not based on exact operational data. The produced biochar is transported to the steel/iron plant to replace part of the PCI coal. Excess pyrolysis gases, both condensable and non-condensable, are fed to the lime kiln of the pulp mill, thus replacing the original lime kiln fuel. In drying, it is assumed that 50% of the energy need is provided by hot water, assumed to be available at the pulp mill, and 50% by steam. Some steam is available from the pyrolysis flue gas, which is generated by burning part of the pyrolysis gases to cover the endothermic reactions taking place during pyrolysis. The rest of the steam requirement for drying is assumed to be available from the pulp mill. No excess steam is available.

Pressing and/or drying costs of produced lignin before transportation (L1 and L2), or pressing costs (before drying, which is in direct connection to the pyrolysis process) (L3), are not taken into account and are assumed to be covered by the ethanol plant, which produces lignin as a side product. The pressing/drying costs are assumed to be covered by the higher heat content of the pressed/dried raw material when compared to the heat content before pressing. Evaluation of the raw material is based on this heat content, thus also the cost of the pressed raw material is higher.

A summary of the considered scenarios is presented in *Table 12*.

Table 12. Different scenarios selected for the evaluation.

Scenario	Integrated with a pulp mill	Raw material and moisture as received	Distance [km] and raw material or product transported	Use of pyrolysis gas	Electricity generation
L1	No	Lignin 50%	200 / Lignin	Internal	Yes
L2	No	Lignin 10%	200 / Lignin	Internal	Yes
L3	Yes	Lignin 50%	160 / Biochar	Lime kiln	No
BP	No	Black pellets 10%	275 / Black pellets	Internal	Yes
Ba	Yes	Bark 60%	275 / Biochar	Lime kiln	No

3.4 Modelling of biomass drying and slow pyrolysis

This section focuses on the modelling of the drying and pyrolysis process, and the subsequent *Section 3.6* broadens the focus taking into account also the processing locations and transport, possible electricity generation, optional fuel replacements in the lime kiln, and subsequent benefits and losses due to the integration in calculations of the production costs.

Hydrolysis lignin, black pellets and softwood bark are the raw materials studied for producing biochar to replace part of the PCI coal at the steel/iron plant. The process models required are built within the project in the Balas® simulation software (http://balas.vtt.fi/). Experimental work within the project provides elemental analysis and heating values of the raw materials and products. Consequently, three optional models are presented for stand-alone production, and two models for pulp mill integrated production of biochar.

3.4.1 Process description of the models

In the models providing the data for the stand-alone production scenarios, the pyrolysis and burning of pyrolysis gases are modelled to be contained within a combined isothermal reactor unit, and thus no pyrolysis gases are produced as side products. The scheme of the respective stand-alone setup is given in *Figure 18*. Heat is recovered from the flue gases (pyrolysis and burning of the pyrolysis gases) in the boiler section of the model. The flue gas end temperature is set at 130 °C. Steam generated in the model, from the boiler section and the combined reactor unit, is set to 2 bar (a). The energy requirement for drying and the heat losses (drying, pyrolysis and boiler section) are estimated. The same amount of steam is condensed in the steam network of the model. The surplus steam is valued as electricity by configuring the electricity generation yield as 35% for the available thermal load. This is described more closely in *Section 3.6.* The low steam temperature for the calculation is not critical, since the overall thermal load is not sensitive with regard to the selected modelling temperature. The real steam pressure, which can be generated by the burning of the pyrolysis gases, will gain a much higher level (same heat load as in the model) and is further assumed to be sufficient for steam turbines at the power plant.

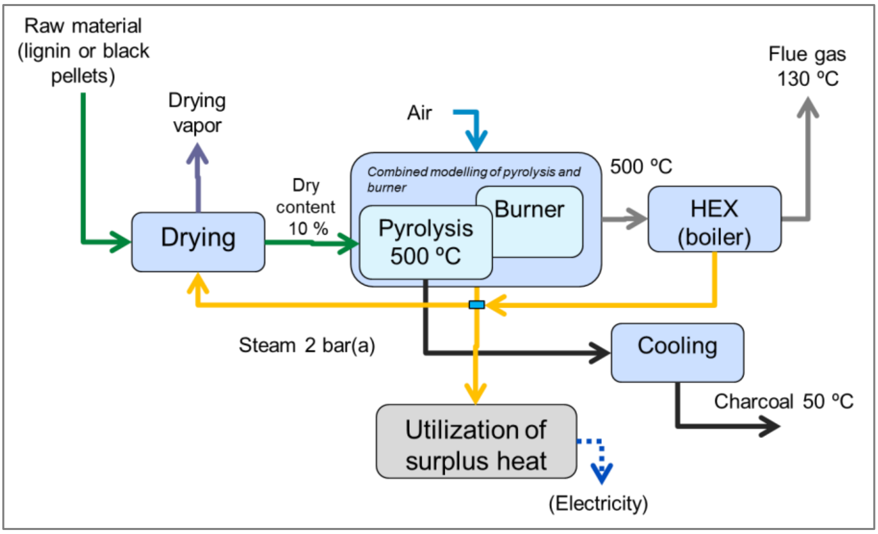


Figure 18. Schematic process diagram of the Balas® simulation models providing data for the stand-alone production scenarios. Pyrolysis and burning of pyrolysis gases are combined into one isothermal reactor unit; surplus heat is used for electricity generation.

In the models providing the data for the pulp mill integrated production scenarios (*Figure 19*), pyrolysis is calculated separately in an isothermal reactor unit. The model thus allows the use of the pyrolysis gases for further utilization as fuel, for example in the lime kiln. Part of the pyrolysis gases are combusted in the burner unit to provide the energy required for the endothermic reactions of the pyrolysis itself. Heat is recovered as 2 bar (a) steam from the resulting flue gases. The steam is further used for drying the feed biomass. The model also calculates the additional energy requirement for drying, which is provided from the pulp mill (as hot water and steam). No excess energy/steam is available.

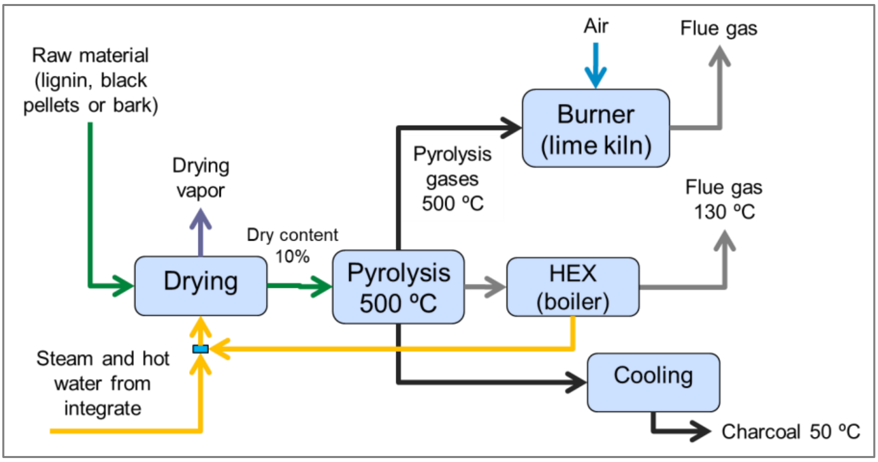


Figure 19. Schematic process diagram of the Balas® simulation models providing data for the pulp mill integrated production scenarios. Pyrolysis is modelled by an isothermal unit, and pyrolysis gas is led to a separate burner unit. No steam is left for electricity generation. Most of the energy for drying is provided externally from the pulp mill.

3.4.2 Assumptions

The production scenarios under examination are presented in *Section 3.3* and in *Table 12*, where L stands for the raw material hydrolysis lignin, BP for black pellets, and Ba for bark; the pulp mill integrated scenarios are denoted as L3 and Ba.

It is assumed that ca. 10 wt% of PCI coal in the blast furnace can be directly replaced with biochar. While a 10 wt% substitution of coal with biocarbon is expected to be unproblematic, there would in principle be no limitation on increasing the fraction of biocoal even up to 100% if certain technical limitations are solved, such as sizing of the PCI coal handling and pulverizing line and re-designing the co-feed of coal and biocoal to the blast furnace. As the PCI coal feed is approximately 220 kg per tonne of crude iron, the corresponding replacement with biochar would be 22 kg of biochar; and as the annual PCI coal feed is ca. 450 000 tonnes, the amount of replacement biochar would be 44 200 tonnes. This amount is also assumed to closely match the capacity of the future plant, which will produce the required lignin raw material as a side product (scenarios L1, L2 and L3). The main product of this future plant is ethanol, produced from sawdust. The same annual capacity of 44 200 tonnes of biochar is also applied to the BP scenario with black pellets as the raw material. However, for the Ba scenario (bark as a raw material) the capacity is set according to the bark amount assumed to be available at the pulp mill.

The models in which pyrolysis and burning of pyrolysis gases are included within the combined isothermal reactor unit (*Figure 18*) provide data for the stand-alone production scenarios, which are for lignin feed material L1 and L2, and for black pellets BP. The mass and molar balances of the elements of raw materials and biochar, and mass yield of biochar are given, from which the stoichiometric reactions for raw material, biochar and flue gas components (CO_2, NO_2, H_2O and SO_2) are then formulated. The simulation then calculates the exothermic energy from this combined reactor unit, while the pyrolysis itself is endothermic.

The models in which pyrolysis is calculated in the separate isothermal reactor unit and pyrolysis gases are separated for further utilization as fuel (e.g. in the lime kiln), provide the data for the pulp mill integrated production scenarios, which are for lignin feed material L3 and for bark Ba (*Figure 19*). In these models, the pyrolysis gases (condensable and non- condensable) are expressed by artificial model components, which are created by deducting biochar from the pyrolysis feed material at the elemental level. The resulting elemental compositions of these model components, lignin and bark pyrolysis gas, are presented in *Table 14*. The actual pyrolysis gases were not analysed within the project. The model uses higher heating value (HHV), and for the pyrolysis gas component this is estimated by applying the element-based weight factors introduced by Channiwala and Parikh (2002). The calculations are presented in *Table 16.*

Table 13 presents the parameters, and their sources, applied in the modelling. It should be noted that for scenario Ba (bark as a raw material) the capacity is set according to the bark amount assumed to be available at the pulp mill. The University of Oulu provided the scenario parameters for the yield, raw material dry content as received, and heating values for raw material and biochar, based on the laboratory analyses performed.

Table 13. *Parameters applied in modelling the scenarios.*

		All	L1	L2	BP	L3	Ba	Source
Biochar amount for injection	[k-tonnes/a]		44.2	44.2	44.2	44.2	96.0	Industry
Yield of biochar, dry-based	[wt%]		45.0	45.0	42.6	45.0	40.0 *)	Univ. of Oulu
Process in operation	[h]	8 000						Estimation
Raw material dry content as received	[wt%]		50	90	89.6	50	40.0 *)	Univ. of Oulu
Dry content before pyrolysis (after drying)	[wt%]	90						Estimation
Pyrolysis temperature	[°C]	500						Univ. of Oulu
Biochar temperature after cooling	[°C]	50						Estimation
Higher heating value (HHV) of the raw material, dry-based	[MJ/kg HHV]		25.22	25.22	20.84	25.22	20.84*)	Univ. of Oulu
Lower heating value (LHV) of the raw material, dry-based	[MJ/kg LHV]		23.87	23.87	19.65	23.87	19.65*)	Univ. of Oulu
Higher heating value (HHV) of the biochar	[MJ/kg HHV]		33.12	33.12	29.62	33.12	29.62*)	Univ. of Oulu
Lower heating value (LHV) of the biochar	[MJ/kg LHV]		32.36	32.36	29.04	32.36	29.04*)	Univ. of Oulu
Losses in pyrolysis: share of heat content of produced pyrolysis gases (own use subtracted)		2						Estimation
Total energy consumption in drying, including losses, as steam	[kWh/tonne H_2O]	1100						Hannula & Kurkela (2013)
Heat losses in drying	[%]	43						Calculations
Heat losses in the steam boiler (radiation)	[%]	0.4						Estimation
Excess air coefficient in burning		0.001						Estimation
Boiler blowdown	[wt%]	4.0						Estimation
Flue gas end temperature	[°C]	130						Estimation
Steam pressure	[kPa]	200						Estimation
Boiler approach T	[°C]	9.77						Estimation
Correction factor for the enthalpy of vaporization for water, at 25 °C, per 1 wt% of moisture	[MJ/kg]	0.0443						Alakangas et al. (2016)

**) The same heating values are assumed for Ba (bark) as for BP (black pellets) and biochar products. The bark dry content as received is an estimate.*

Table 14 presents the elemental compositions of the different modelling components applied as ash free. Ash is treated separately as an inert component. The ash entering the pyrolysis ends up in the biochar. Lignin and black pellet raw materials and the resulting biochar compositions are based on the laboratory analyses performed by the University of Oulu. It is also assumed that the bark composition and the resulting biochar composition is the same as those for the analysed black pellets and resulting biochar. The artificial single model components for the pyrolysis gases (condensable and non-condensable) are created based on the elemental balances of the pyrolysis raw material and the resulting biochar. The real compositions of bark and pyrolysis gas were not analysed.

Table 14. Elemental compositions of the different modelling components applied as ash free and the scenarios in which the components are applied. Molar compositions are scaled to 1000 g.

Modelling component	**C [mole]**	**H [mole]**	**N [mole]**	**O [mole]**	**S [mole]**	**L1 and L2**	**BP**	**L3**	**Ba**
Lignin	51.18	61.98	0.50	19.68	0.03	X		X	
Black pellet	46.23	57.17	0.31	23.90	0.01		X		
Bark	*As black pellet*								X
Lignin biochar	71.94	35.53	0.88	5.41	0.04	X		X	
Black pellet biochar	73.60	30.01	0.82	4.62	0.01		X		
Bark biochar	*As black pellet biochar*								X
Lignin pyrolysis gas	34.32	83.47	0.19	31.28	0.02			X	
Bark pyrolysis gas	30.06	73.21	0.02	35.29	0.01				X

Table 15 gives the calculated and applied feed streams and fixed stoichiometry given for the reactions. The reaction stoichiometries are set to meet molar element balances.

Table 15. Calculated and applied feed streams and fixed stoichiometry of the reactions. Note that the feed streams presented are the dry-based modelling components, whereas ash is treated separately as an inert component.

Modelling component	**Unit**	**L1 & L2**	**BP**	**L3**	**Ba**
Feed, dry-based (modelling components, ash treated as separate component)					
Black pellets	[wt%]		95.4		
Lignin	[wt%]	99.7		99.7	
Bark	[wt%]				95.4
Ash	[wt%]	0.3	4.6	0.3	4.6
Stoichiometry of pyrolysis and/or burning					
Conversion of raw material components (ash-free)	[wt%]	100	100	100	100
Black pellets	[wt%]		-100		
Lignin	[wt%]	-100		-100	
Bark	[wt%]				-100
O_2	[wt%]	-70.2	-55.1		
N2	[wt%]		-0.01		
Biochar	[wt%]	44.8	39.8	44.8	37.1
CO_2	[wt%]	83.3	74.6		
H_2O	[wt%]	41.5	40.7		
NO_2	[wt%]	0.48	0.00		
SO_2	[wt%]	0.07	0.03		
Pyrolysis gas (artificial single modelling component)	[wt%]			55.2	62.9

For the pyrolysis gas components HHV is estimated by applying the element-based weight factors proposed by Channiwala and Parikh (2002). These calculations are presented in *Table 16*. The resulting HHV value estimation is higher for lignin (19.1 MJ/kg) than for black pellets (15.5 MJ/kg). It is to be noted that the elemental weight percentage for black pellets is outside the upper bound for oxygen, which may cause some error.

Table 16. Estimation of HHV values for hydrolysis lignin and for black pellets pyrolysis gas by applying weight factors for each element as presented by Channiwala and Parikh (2002).

	C	**H**	**O**	**N**	**S**	**A**	**HHV [MJ/kg]**
Weight factors	0.349	1.178	-0.103	-0.015	0.101	-0.021	
	[wt%]	[w%]	[wt%]	[wt%]	[wt%]	[wt%]	
min	*0.00*	*0.43*	*0.00*	*0.00*	*0.00*	*0.00*	
max	*92.25*	*25.15*	*50.00*	*5.60*	*94.08*	*71.40*	
Lignin pyrolysis gas	41.2	8.4	50.0	0.3	0.1	-	19.1
Bark pyrolysis gas *)	36.1	7.4	*56.5*	0.0	0.0	-	15.5

**) Based on black pellet raw material and on the resulting biochar analyses*

3.5 Biochar production integration with a pulp mill

The analysis of the integration of biochar production with a Kraft pulp mill is based on the process simulation, in addition to the pyrolysis modelling explained above. This analysis is conducted using the reference softwood Kraft pulp mill model presented in *Figure 20*. The model comprises a single-line market pulp mill with wood handling, continuous cooking, two-stage oxygen delignification, ECF bleaching (D_0-E_{op}-D_1-P), pulp drying, seven-stage evaporation with tall oil recovery, recovery boiler, re-causticizing and condensing turbine. The reference mill, based on the Future Pulp Mill (FUPU) concept, is described in detail by Kangas et al. (2014). Some outside boundary limit areas, such as chemical preparation, air separation unit and water and wastewater treatment plant, are excluded from the model. Two options for bark treatment have been previously evaluated using the reference mill model: i) gasification of bark (Kangas et al. 2014) and ii) combustion of bark in a multi-fuel boiler (Onarheim et al. 2017).

The overall block flow diagram of the reference softwood Kraft pulp mill is shown in *Figure 20*. Wood consumption of the FUPU mill is 5.7 m^3/adt (air-dried tonne) of pulp. The dry content of the incoming wood is ~50%. Roundwood is debarked, yielding 470 kg of bark with a dry content of 40% per air-dried tonne of pulp. Lignin and hemicelluloses are dissolved from the wood chips during the cooking stage, where NaOH and NaS are used as cooking chemicals. The pulp yield during cooking is 47%. Delignified pulp is bleached with oxygen, chlorine dioxide, and hydrogen peroxide. Finally, the pulp is dried, giving a final yield of 44% from the chips.

Chemical recovery is an important part of the Kraft pulp mill process. The aim is to utilize the dissolved wood as energy and to recycle cooking chemicals back to the fibre line. Used cooking chemicals, i.e. black liquor, are concentrated in evaporators from 17% dry content to 80% dry content. Strong black liquor is combusted in the recovery boiler and 22 GJ/adt of pulp super-heated steam is gained. Cooking chemicals (Na_2CO_3 and Na_2S) are simultaneously obtained as smelt. The smelt is dissolved and green liquor is produced. The green liquor is converted back to cooking chemicals at the re-causticizing plant where lime reacts with the sodium carbonate.

In the lime cycle, the rotary lime kiln converts the used lime mud back to lime. The energy consumption of lime kiln is 1.7 GJ/adt of pulp. Various fuels can be used for firing lime kiln from heavy-fuel oil to natural gas. Within the reference model, gasified bark is used as lime kiln fuel. The total steam consumption of reference mill is less than 10 GJ/adt of pulp and the typical electricity consumption is 600 kWh/adt of pulp. Excess steam is converted to electricity. A stand-alone Kraft pulp mill produces 760 kWh/adt of pulp surplus electricity, which can be sold to the grid. The detailed process parameters of the FUPU reference mill are available in the literature (Kangas et al. 2014; Onarheim et al. 2017).

The reference mill is comparable to another well-known reference model, the FRAM model (Berglin and Lovell 2011; ÅF-Engineering 2011), and typical industrial parameters (European IPPC Bureau 2015). The FUPU model was developed here using the Wingems 5.3 modelling tool (Valmet 2016). Additional analyses, such as economics, were performed with Excel spreadsheets, and HSC Chemistry 9 (Outotec 2017) and e!Sankey (ifu Hamburg 2015) were utilized for illustrating the results.

The reference Kraft pulp mill model has been successfully used for analysing new pulp mill concepts such as energy mill, sulfur lean pulping, and high kappa cooking (Kangas 2016), but also for estimating the possibilities of biogenic carbon capture from pulp and board mills (Onarheim et al. 2017). Within this study, the reference model is used for estimating the integration effects of biochar production on Kraft pulp mill energy balances.

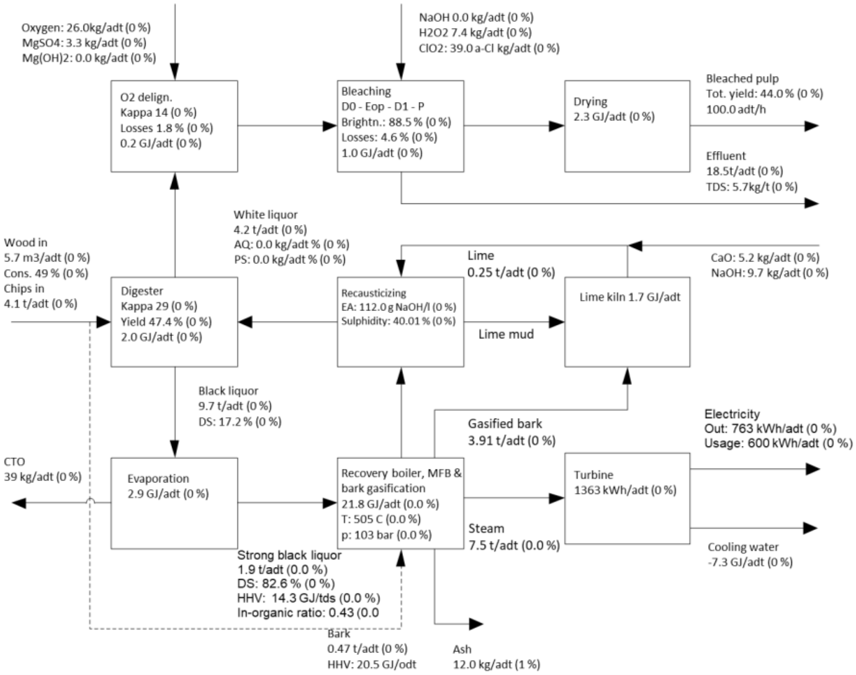


Figure 20. Reference softwood Kraft pulp mill (FUPU), major areas and key process parameters. Based on Kangas et al. (2014); some modifications based on Onarheim et al. (2017). Wingems 5.3 used as the modelling tool.

Two alternative scenarios for the pulp mill integration are formulated: i) Integrated pulp and paper mill with a production capacity of 800 000 air-dried tonnes of Kraft pulp (L3 scenario), and ii) Market Kraft pulp mill with a larger production capacity of 1.2 million air-dried tonnes of Kraft pulp per year (Ba scenario).

A schematic block-flow diagram of an integrated pulp and paper mill (baseline for L3) is illustrated in *Figure 21*. The total bark yield is 400 000 tonnes per year with a dry content of 40%. The bark is combusted in a multi-fuel boiler and the lime kiln is fired mainly with bio-oils. Bio-oil consumption is 33 000 tonnes per year. The paper mill uses heat from the boilers and excess heat is also used for district heating in the nearby community. A back-pressure turbine is used for this scenario. The net excess electricity sold outside the integrate is 100 GWh per year.

The market pulp mill is illustrated in a block flow diagram in *Figure 22* (baseline Ba). The total bark yield is 600 000 tonnes per year with a dry content of 40%. Half of the bark is gasified and the lime kiln is fired with the resulting syngas. Remaining bark is combusted in a multi-fuel boiler. A significant amount of excess heat is produced and a condensing turbine is used to convert the heat to electricity. The net electricity output is 1000 GWh per year.

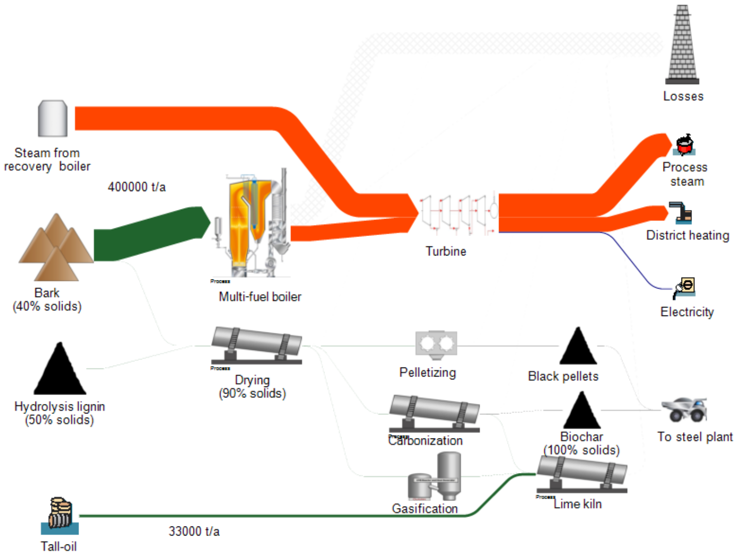


Figure 21. Reference integrated pulp and paper mill (L3 reference). Kraft pulp production is 800 000 air-dried tonnes per year. Bark is combusted in a multi-fuel boiler. Lime kiln is fired with bio-oil. Heat is utilized for district heating.

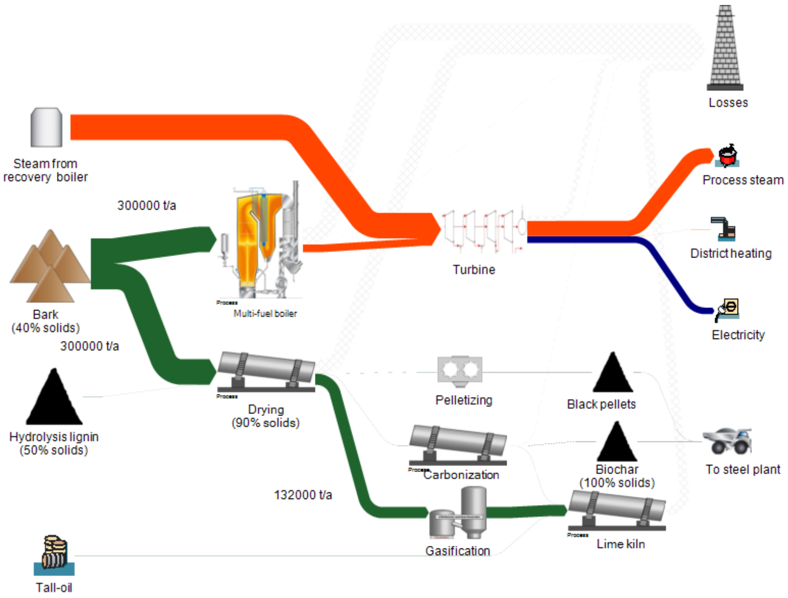


Figure 22. Reference Kraft pulp mill producing 1.2 million air-dried tonnes of softwood Kraft pulp per year (Ba reference). 50% of bark is gasified and lime kiln is fired with bark gas; 50% of bark is combusted in a multi-fuel boiler. Excess heat is converted to electricity in a condensing turbine.

3.6 Business modelling and prefeasibility: biochar production cost

3.6.1 Assumptions

Our estimations of scenario-specific biochar production costs are based on data provided by modelling, laboratory analyses of raw materials and biochars, and on additional parameters necessary for the estimation of production costs. These figures are presented in *Table 17*. Other costs are based on a percentage of the selling price (*Table 18*). The selling price is fixed for all scenarios. In reality, some variations may occur, since one producer may be able to sell biochar at a cheaper price than another due to possibly lower production costs.

A cost calculation template developed for VTT for the manufacture of biochar was used to assess the different scenarios, and was fine-tuned to also take into account the integration effects estimated by VTT experts.

The following figures are based on the laboratory analyses of the University of Oulu:

- Humidity of raw material, as received
- Lower heating value (LHV) of raw material, dry-based
- Biochar production yield, dry-based
- Lower heating value (LHV) of biochar

These and heating values based on higher heating (HHV) values, among other results, are explained in detail in the University of Oulu report (Toloue Farrokh et al. 2019).

Modelling work provided the following estimations:

- Pyrolysis gases production yield, self-consumption subtracted
- Lower heating value (LHV) of pyrolysis gases
- Total energy consumption of drying, including losses and internal use of energy
- Internal energy for drying from heat recovery
- Losses in electricity generation (at the pulp mill power plant)
- Losses in district heat generation (at the pulp mill power plant)
- Ideal external energy input in the drying and pyrolysis system (no losses)
- Estimated energy losses in the system
- Excess heat load for electricity generation

Additionally, the following assumptions are made, as given in *Table 17*:

- Energy shares for raw material drying: hot water / low pressure steam
- Light fuel oil share of the heat load at the lime kiln (valid only for scenario L3)
- Biochar production capacity based on available hydrolysis lignin or bark. For black pellets the same production capacity was selected as for lignin.

Following estimations are based on industrial experience within the project:

- Process time in operation
- Labour, total person years (operating labour, supervisory, production engineers and maintenance). Differences between scenarios L3 and Ba are explained below. Less operating labour and maintenance staff are required due to integration benefits and existing staff in the integrate.
- Electricity generation yield: Electricity / thermal load

Further comments related to the L3 and Ba scenarios (*Table 18*):

- Maintenance staff costs are assumed to be included in the capital expenditure (CAPEX) related maintenance cost, 3% of CAPEX
- No need for handling raw materials (lignin or bark) as pellets, contrary to L1, L2 and BP scenarios, where the raw material is first transported to the pyrolysis plant. This is taken into account in the operating labour need and in the investment estimation.
- Any savings or extra costs due to the pulp mill integration are allocated to the biochar production cost

Biochar yield for black pellets is calculated to be 42.6 wt% by the University of Oulu (scenario BP). Furthermore, bark yield is estimated to be slightly lower at ca. 40%. For comparison, Solar et al. (2018) conducted slow pyrolysis experiments on woody biomass waste from forestry activities and received a yield of 32.8 wt% for biochar at a pyrolysis temperature of 500 °C.

Table 17. Assumptions made, or resulting from the process modelling, for the calculations of production cost estimations for scenario-specific biochar.

Assumptions for biochar production cost estimation	**Lignin** **L1**	**Lignin** **L2**	**Lignin** **L3**	**Black pellets** **BP**	**Bark** **Ba**
Moisture of raw material, as received [%]	50	10	50	10.4	60
Lower heating value (LHV) of raw material, dry-based [MJ/kg]	23.9	23.9	23.9	19.7	19.7
Biochar production yield, dry-based [wt%]	45	45	45	42.6	40
Biochar production capacity [k-tonnes/a]	44.2	44.2	44.2	44.2	96.0
Lower heating value (LHV) of biochar [MJ/kg]	32.4	32.4	32.4	29.0	29.0
Pyrolysis gases production yield, self-consumption subtracted [wt%]	🟩	🟩	53.9	🟩	57.3
Lower heating value (LHV) of pyrolysis gases [MJ/kg] (500 °C)	🟩	🟩	15.2	🟩	12.5
Process in operation [h]	8 000	8 000	8 000	8 000	8 000
degree of capacity utilization [%]	100	100	100	100	100
Labour, total [person years] = Operating labour + Supervisory + Production engineer + Maintenance	27= 15+ 5+ 1+ 6	27= 15+ 5+ 1+ 6	16= 10+ 5+ 1+ 0	27= 15+ 5+ 1+ 6	16= 10+ 5+ 1+ 0
Energy for raw material drying: as hot water [%] / as low pressure steam [%]	🟩	🟩	50/50	🟩	50/50
Total energy consumption of drying, including losses and internal use of energy [kWh/tonne of H_2O]	1 100	🟩	1 100	🟩	1 100
Internal energy for drying (as LP steam) from heat recovery, share of total energy consumption of drying [%]	100	🟩	7.6	🟩	4.7
Losses in electricity generation [kWh/tonne of steam]	🟩	🟩	35	🟩	110
Losses in district heat generation [kWh/tonne of steam]	🟩	🟩	620	🟩	
Fuel oil share of heat load at the lime kiln [%]	🟩	🟩	20	🟩	

Assumptions for biochar production cost estimation	**Lignin**	**Lignin**	**Lignin**	**Black pellets**	**Bark**
	L1	**L2**	**L3**	**BP**	**Ba**
Ideal external energy input in the drying and pyrolysis system (no losses), share of raw material heat load as received [%] [**)]	▨	▨	10.1	▨	21.2
Estimated energy losses in the system, as [%]	13.8	4.7	2.4 [*)]	5.1	1.9 [*)]
Excess heat load for electricity generation, as [%]	18.3	▨	None	▨	None
Electricity generation yield: electricity/thermal load [%]	35	35		35	

[*)] *The losses are internal system losses. If external energy is used for drying (L3, Ba) the related drying losses are not included in the figure. Also, losses in the burning of produced pyrolysis gases (L3, Ba) are not accounted for, being outside the system boundaries. Burning takes place at the lime kiln.*

3.6.2 Applied prices, logistics information and fixed costs

In addition to the figures presented in *Table 17*, price estimations, logistics information and figures for estimating the fixed costs are also needed for the production cost calculations. These are listed in *Table 18*.

The applied electricity price of 35 €/MWh, agreed with the project consortium, is based on the principle that in Finland power plant owners may purchase electricity at prime rates. The market price was notably higher than the reported electricity prices for non-household consumers in Finland in 2017 (second semester) at 67.6 €/MWh (Eurostat 2018). The Eurostat figure includes electricity production and network costs, as well as all non-recoverable taxes and levies for medium size industrial consumers (annual consumption between 500 and 2000 MWh).

Raw material or biofuel price is based on the price history of forest chips, VAT excluded (Statistics Finland 2018b).

Light fuel oil (sulphur free) price (VAT excluded) in *Table 18* includes excise tax of 24.39 cent/l from 1 January onwards (Finnish Petroleum and Biofuels Association 2018a). The density of 0.845 kg/l and the lower heating value of 11.74 MWh/tonne are applied. Oil price is based on the April 2018 price level (Finnish Petroleum and Biofuels Association 2018b). In reality, heavy fuel oil would be used as a lime kiln fuel instead of light fuel oil. As an example, the heavy fuel oil lower heating value, at a sulphur content less or equal to 0.5%, is 11.53 MWh/tonne (Statistics Finland 2018c). The import price for the heavy fuel oil was ca. 510 €/tonne in December 2018 and excise taxes (including strategic stockpile and oil pollution fees) 29.07

cent/kg (Statistics Finland 2018d). The resulting heavy fuel oil price is 69.5 €/MWh, which is somewhat cheaper than the light fuel oil price (78.6 €/MWh) applied.

Revenue income steam for the producer is based on an average district heating energy fee of 63.79 €/MWh for an apartment building of 80 apartments, assuming that district heat is provided from a combined heat and power production plant (Finnish Energy 2018) and that the producer gets approximately 45% of this fee.

Labour, maintenance, and insurance cost shares were based on the CAPEX estimation, other cost shares were based on sales, transportation related handling fee [€/tonne] and railway transportation costs [€/tonne/100 km], and CAPEX estimates were based on accumulated industrial information during the project and personal communication.

Further comments related to the scenarios:

- The Ba investment estimate is much higher than the other scenarios due to the higher production capacity, as described in *Subsection 3.4.2*
- In Ba, in contrast to BP, it is assumed that there is no need for a raw material handling line as the bark handling line is part of the pulp mill. This is taken into account in the investment cost estimation.
- For L3, the raw material handling line investment cost estimate is lower than in scenarios L1 and L2 as it is assumed that lignin is transported by conveyor at the integrated plant, whereas in L1 and L2 the raw material pellets arriving at the pyrolysis site after transportation need a handling line.
- Biochar must be pelletized before transportation to the steel/iron mill location. This is noted in the investment cost estimates in scenarios L3 and Ba. What adhesives or binders should be used for pelletizing remains unresolved. One solution could be to utilize part of the condensable gases, the tar-containing fraction, as a binder. Currently, the pyrolysis gases, both condensable and non-condensable fractions, are used as a fuel in the lime kiln.
- In the investment estimates for both L3 and Ba, the drying process investment estimate is included, as in L1, but not in L2 or in BP where there is no need for drying.

Table 18. Price estimations, logistics information and fixed-cost figures for the calculation of production cost estimations for scenario-specific biochar.

Assumptions for biochar production cost estimates	**Lignin**	**Lignin**	**Lignin**	**Black pellets**	**Bark**
	L1	**L2**	**L3**	**BP**	**Ba**
Biochar sales price estimate: 330+10% margin [€/tonne]	363	363	363	363	363
Electricity: Purchase and sales price [€/MWh]	35	35	35	35	35
Raw material or biofuel price: Based on heating value [€/MWh]	20	20	20	20	20
Light fuel oil price [€/MWh]			78,6		
Hot water cost: Inside the integrate for drying [€/MWh]			0		0
District heating price for the producer: 45% of the consumer price [€/MWh]			28,7		
Labour cost [€//person/month] Operating labour and maintenance 100%, supervisory 125%, production engineer 150%	4 402	4 402	4 402	4 402	4 402
Maintenance [% of CAPEX] Includes maintenance staff [Yes /No]	3 No	3 No	3 Yes	3 No	3 Yes
Insurance [% of CAPEX]	2.5	2.5	2.5	2.5	2.5
Other costs [% of sales]	3	3	3	3	3
Transportation costs:					
- Handling fee [€/tonne]	1	1	1	1	1
- Railway transportation [€/tonne/100 km]	3.5	3.5	3.5	3.5	3.5
- Transportation distance [km]	200 Lignin	200 Lignin	160 Biochar	275 Black pellets	275 Biochar
Investment cost estimate (CAPEX) [M€]	24.9	22.7	27.4	23.5	51.4
Opportunity cost of capital [%]	5.0	5.0	5.0	5.0	5.0
Investment period [a]	15	15	15	15	15

3.7 Assessment of environmental impacts

The environmental impacts of the studied systems are evaluated by life cycle assessment (LCA). The LCA calculations are performed for the hydrolysis lignin case (L3) and bark case (Ba). The impacts of the new systems are compared with the conventional production systems that are assumed to take place if the biochar production facilities are not implemented.

3.7.1 Introduction to life cycle assessment

LCA is a standardized method (ISO 14040, ISO 14044) that is widely used to quantitatively assess the effects of an activity on the environment. By definition, LCA takes into account the whole life cycle of a product, process or service. This means calculating the impacts from raw material acquisition to end-of-life (waste disposal) or to material recycling. This approach is called cradle-to-grave or cradle-to-cradle, respectively. The assessment can also be performed for part of the life cycle, typically excluding the use phase and end-of-life (i.e. cradle-to-gate).

Life cycle assessment consists of four phases: goal and scope definition, inventory analysis, impact assessment, and interpretation, as illustrated in *Figure 23*.

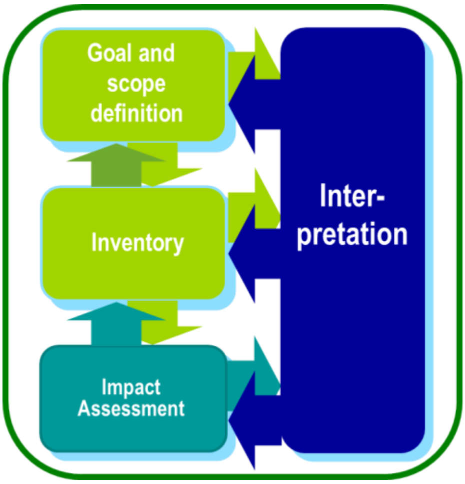


Figure 23. The four phases of LCA based on ISO 14040

In the first phase, the goal of the assessment is defined, i.e. what will be assessed and why. In addition, the scope of the assessment is defined by outlining the systems under study. This includes defining the product system and its function, the functional unit, system boundaries, data requirements, and assumptions.

The life cycle inventory (LCI) phase includes data collection and a balance calculation for all unit processes in the life cycle. The results are presented as inputs and outputs of the entire system.

The LCI results are converted into potential environmental impacts in the impact assessment (LCIA) phase. The inventory data is associated with specific environmental impact categories and category indicators using a selected impact assessment methodology.

The final phase of LCA is interpretation of the results, which is based on all three previous phases. The outcomes of the phases are continuously interpreted, thus serving to redefine the goal and scope of the study, which leads potentially to an improved scope definition and more accurate results. Finally, the interpretation phase reflects on the results with respect to the goal and scope of the study and draws conclusions and provides recommendations based on the outcomes.

3.7.2 Goal and scope definition

The goal of the assessment was to determine the environmental impact of integrating the examined biochar production systems with a pulp mill. Due to the numerous trade-offs between different products and different options for dealing with reduced energy flows and replaced energy sources, it is important to quantitatively assess the overall impacts of the scenarios.

The scope of assessment covers the hydrolysis lignin case (L3) and the bark case (Ba) in which biochar production is integrated with a pulp mill. The system boundaries are set to cover all affected unit processes in pulp production, new production processes and raw materials, and the changes in energy flows.

The reference cases represent systems where the pulp mills operate without biochar production. Bark is used as a fuel to produce energy that is utilized both internally and externally. The energy for the lime kiln is produced from bio-oil and bark in L3 and Ba, respectively. Energy that is not needed internally is sold as electricity or district heating. Integrating biochar production with the pulp mill changes the material and energy flows. The biochar is produced by slow pyrolysis, which yields, in addition to biochar, pyrolysis gases that can be burned to produce energy. This is utilized in the lime kiln, resulting in reduced need for other energy sources.

The impacts are calculated based on the annual volumes of the production systems, i.e. the functional unit of the study is the annual production volume of the system yielding multiple products. This means that the results show the absolute results of the systems for one year, and the difference between the reference and the biochar production scenario describes how the impacts could change if the biochar production system was implemented.

3.7.3 Life cycle inventory

The mass and energy balances for the studied systems are based on the research done within the project. The chosen assessment approach aims at calculating the differences in impacts, i.e. all absolute flow values are not considered. More detailed information about the life cycle inventory can be found in *Section 9.1*.

3.7.4 Impact assessment

The impact assessment is focused on assessing the impact on climate change, i.e. the impact category chosen for the study is global warming potential (GWP100). The category is based on the characterization factors defined by The Intergovernmental Panel on Climate Change (IPCC). The assessment was performed by using the LCA software SULCA 5.1.

The impact assessment results are presented in *Section 9.2*.

4. Bioreducer production results

The University of Oulu used several characterization methods to investigate the suitability of biochars for iron and steel industry, described in Toloue Farrokh et al. (2019). Slow pyrolysis experiments were carried out, as well as other experiments, to define:

- mass and energy yields
- chemical, elemental and ash compositions

Following physical analyses were also performed to define and investigate:

- particle sizes and shapes
- microstructures
- densities
- surfaces and pores
- water uptake
- flow and transport properties
- combustibility and reactivity of biochars

Tolouefarrokh at al. summarized the obtained results as following: Properties of biochars as a sustainable resource for blast furnace injection are related to the biomass source material and pretreatment conditions like pyrolysis temperature. Heat treatment of lignin and black pellet (made of bark) in a slow pyrolysis process cause different changes in the biochar. The decomposition reactions, including devolatilization, carbonization and repolymerization, take place over a temperature range. The properties of produced char depend on these reactions affecting the proceeding processes in which the char will be used.

- Slow pyrolysis improves physical and chemical properties of lignin with the best results for the char obtained in pyrolysis temperature of 500 °C
- Lignin and black pellets char, obtained in pyrolysis temperature of 500 °C, powders were more spherical and had higher bulk density displaying lower compressibility and permeability properties along with lower cohesion and aeration energy of the samples studied. These were also categorized as a free-flowing powders based on their flow function indices and, thus, were assumed to behave well in blast furnace injection system from storage and material handling to injection into blast furnace.
- Thermogravimetric indices (see in detail Toloue Farrokh et al.) revealed that the combustibility of lignin chars decreased as the temperature of pyrolysis increased. However, the combustibility of lignin char samples obtained pyrolysis temperatures of 300 °C and 500 °C was higher than that of pulverized coal. The ash content of the lignin chars was significantly lower compared to pulverized coal. The ash and fixed carbon content affect to the reactivity. Lignin char

samples showed higher reactivity towards CO_2 than pulverized coal but increasing the pyrolysis temperature lead to lower reactivity. The reactivity of lignin chars was much lower than that of black pellet.

- Black pellet char combustion started and ended in a very low temperature range which shows its high combustibility among all studied samples. Black pellet char had an ash content as high as pulverized coal, but with higher basicity because of higher CaO and K2O levels. Higher reactivity was observed compared to lignin chars and pulverized coal which, aside from its biomass nature, might be due to higher calcium oxides content of the ash.
- Combustion and reactivity behaviour of char samples are influenced by several properties of individual chars and cannot be estimated by using any single parameter of the chars.

5. Results of blast furnace modelling

5.1 Reactor operations in terms of charge and PCI composition

The steady state version of the CROM simulator was used to analyse the multicomponent thermochemistry within the blast furnace during operation. “Masuuni 1” at SSAB Raahe steel mill was used as a test case with its reported operational parameters. The height of the furnace is ~28 meters and its total volume is ~1450 m^3.

Excel and Visual Basic was used to make an interface for designing and executing the simulation model and for viewing the results of the calculations. *Figure 24* shows the flowsheet of the blast furnace with material bed (red boxes), gas (white circles) nodes, and the connections (flows) between them.

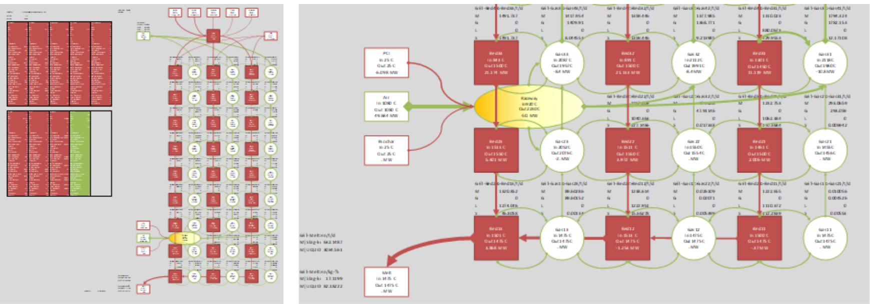


Figure 24. Blast furnace flowsheet for the steady-state version of the CROM simulator. The picture on the right hand picture shows a detail of the flowsheet.

The blast furnace was divided into three radial (centre, middle and wall) and nine axial (heart (2 nodes), bosh&belly (2 nodes), stack (4 nodes) and throat (1 node)) calculation nodes.

The measured average production rate of the hot metal (pig iron) in the furnace was 2943 tonnes per day. *Table 19* shows the flow rates and the compositions of the feed flows taken from the furnace operation report given by SSAB. Charge is composed of Pellett1, Pellett2, Briquette, Others and Limestone feeds. The charge is mixed with coke so that coke/ore ratio is highest at the centre node (in radial direction). PCI and Air are blown through the tuyeres to the heart of the furnace. No biochar was not used in this simulation to replace part of the PCI but the (unused) Biochar composition in *Table 19* was taken from analysis of the lignin sample pyrolyzed at 500 °C by University of Oulu (Toloue Farrokh et al. 2019).

After the combustion of the PCI with the air in the raceway, the hot gas (~2200 °C) is distributed between the three radial nodes. Largest fraction, 40% goes to the centre node, 30% goes to the middle node and the rest (20%) goes to the wall node. This distribution has not been verified.

Table 19. Flow rates (thm = tonnes of hot metal) and composition of the feed flows.

Feeds	**Pellet 1**	**Pellet 2**	**Briquette**	**Others**	**Lime-stone**	**Coke**	**PCI**	**Biochar**	**Air**
T/C	25	25	25	25	25	25	25	25	1080
P/bar	1	1	1	1	1	1	1	1	1
Mass/t/thm	0.8	0.49	0.125	0.095	0.07	0.335	0.144	0	1.23
N_2	0	0	0	0	0	0.5	1.5	1.13	70.08
H_2	0	0	0	0	0	0.5	3	3.56	0
H_2O	0	0	5.68	0	0	0	0	0	0
O_2	0	0	0	0	0	0.5	3	8.6	29.92
CO	0	0	0	0	0	0	0	0	0
CO_2	0	0	0	0	0	0	0	0	0
C_Graphite(s)	0	0	10.75	0	0	87.6	83.18	85.9	0
Al_2O_3_gamma(s)	0.435	0.34	1.49	1.58	0	3.1	2.5	0.014	0
$CaCO_3$_ Aragonite_Pmcn(s)	0	0	0	0	97	0	0	0.078	0
CaO_Lime(s)	0.33	0.2	13.79	28.15	0	0.28	0.2	0	0
$Ca(OH)_2$_Portlandite(s)	0	0	0	0	0	0	0	0	0
$CaSO_4$_Anhydrite(s)	0	0	0	0	0	0	0	0.078	0
Cr_solid(s)	0	0	0	0	0	0	0	0	0
Cr_2O_3_solid(s)	0.05	0.028	0	0	0	0	0	0	0
Fe_bcc(s)	0	0	16.6	35.53	0	0	0	0	0
FeO_Wustite(s)	0.415	0.635	8.3	0	0	0	0	0	0
Fe_3O_4_Magnetite(s)	1.63	1.317	0	0	0	0	0	0	0
Fe_2O_3_hematite(s)	93.12	89.64	31.6	23.16	0	0.49	0.5	0.104	0
K_2CO_3_Solid_A(s)	0	0	0	0	0	0	0	0.082	0
K_2O_solid(s)	0.13	0.03	0.82	0	0	0.17	0.25	0	0
$MgCO_3$_magnesite(s)	0	0	0	0	2	0	0	0	0
MgO_Periclase(s)	1.3	1.5	1.12	1.58	0	0.11	0.2	0.016	0
Mn_Solid_Alpha(s)	0	0	0	0	0	0	0	0	0
MnO_solid(s)	0.15	0.12	0.7	0	0	0.005	0.005	0.11	0
Na_2O_Solid-A(s)	0.05	0.04	0.58	0	0	0.05	0.055	0	0
Na_2SO_4_Solid_IV_(orth(s)	0	0	0	0	0	0	0	0.233	0
P_Solid_(white)(s)	0	0	0	0	0	0.095	0.1	0	0
P_2O5-H(s)	0.02	0.017	0	0	0	0	0	0.031	0
S_Orthorhombic(s)	0	0.003	0.2	0	0	0.65	0.7	0.033	0
Si_solid(s)	0	0	0	0	0	0	0	0	0
SiO_2_Quartz(l)(s)	1.9	5.93	8	10	1	5.8	4.6	0.031	0
Ti_Solid_Alpha(s)	0	0	0.21	0	0	0	0	0	0
TiO_2_Anatase(s2)	0.22	0.2	0	0	0	0.14	0.2	0.000	0
V_solid(s)	0	0	0	0	0	0	0	0	0
V_2O_5_solid(s)	0.25	0	0.16	0	0	0.01	0.01	0	0
Check sum	100	100	100	100	100	100	100	100	100

Figure 25 shows the calculated average composition profiles for the carbon and the main iron containing phases in the material bed. Material bed flows from right (furnace top) the left (furnace bottom). The shown values are the average over the cross-section of the furnace (centre, middle and wall together). First, the hematite is reduced to magnetite when the temperature exceeds ~500 °C. Then rest of the hematite and magnetite are reduce to wustite and wustite is finally reduced to metallic iron. When the temperature of the material bed exceeds ~1150 °C, the liquid slag and metal are starting to form. Remaining carbon is accumulated on the top of the melt in the heart of the furnace (forming dead man's zone). The calculated production rate of hot metal (LIQUID) was 3035 tonnes per day, and the production rate of the slag was 208 kg per tonnes of hot metal (the measured average value was 212.6 kg/thm). The calculated rate of the blast furnace gas was 181 636 Nm^3/h (at 0 °C) (the measured average value was 177.8 Nm^3/h). The calculated dust load in the gas was 58.7 tonnes per day (the measured average value was 25.9 tonnes per day).

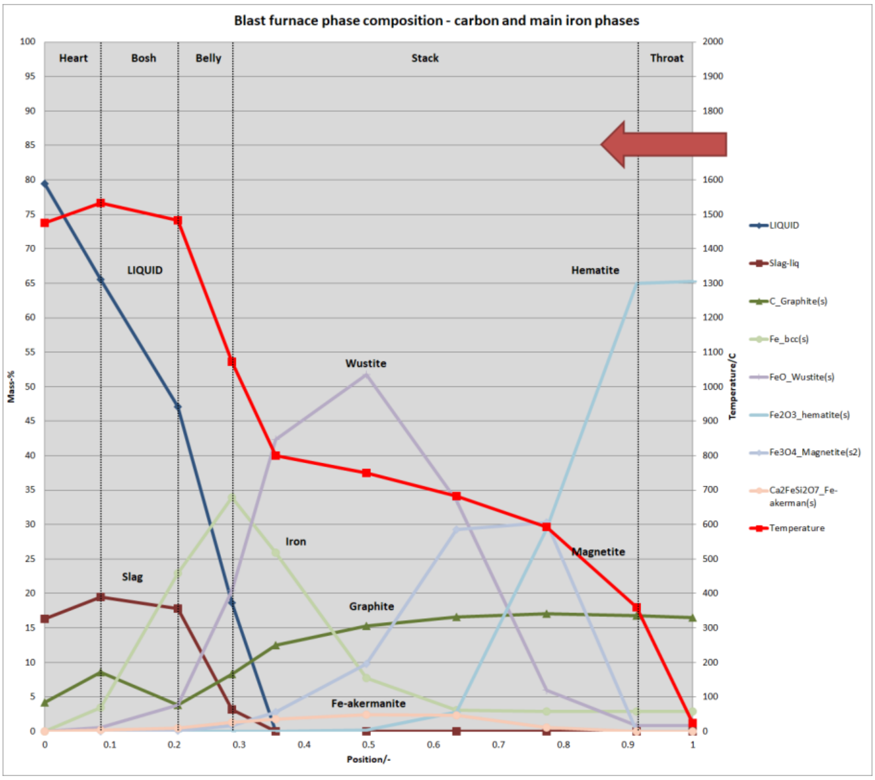


Figure 25. Calculated average composition profiles for the carbon (C_graphite(s)) and the main iron containing phases in the material bed.

Figure 26 shows the calculated potassium circulation inside the furnace between the material bed and the gas. The material bed flows from the right to the left. Potassium rate in the bed is increased as the dust in the gas is mixed with bed until at the hot bottom part of the furnace the potassium is vaporized and transferred to the gas side again. The circulation is dependent on the mass transfer of gas species and dust particles between the bed and the gas.

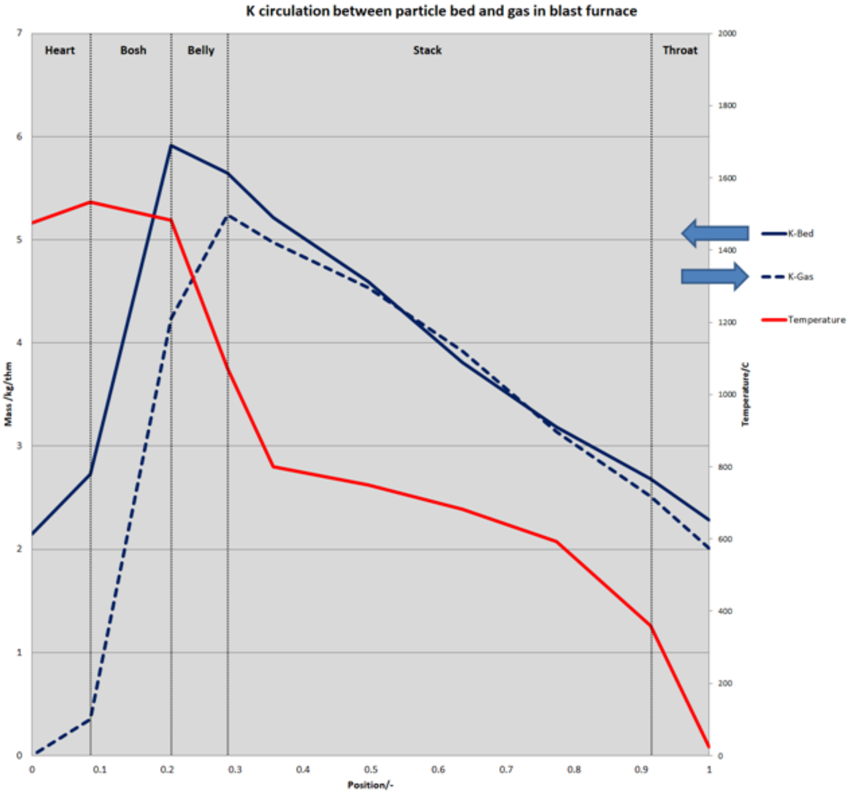


Figure 26. The calculated bed temperature profile and the potassium circulation inside the furnace between the material bed and the gas [kg/thm] (thm = tonnes of hot metal).

Table 20 shows the elementary compositions of the two exit flows of the furnace (gas+dust and tabbed melt (LIQUID+Slag), as well as the compositions of the LIQUID and slag phases.

The composition of the carbon in the hot metal is practically same as its solubility limit in the melt. More potassium exits the furnace via the gas flow than via the tabbing. This ratio is affected by the mass transfer rate between the bed and the gas flows. In this test case 8% of the gas species and 50% of the solid phases in the gas flow were mixed with the bed at each axial location. And all gas species and 0.1% of the solid phases in the bed were mixed back to the gas flow after calculating the kinetically constrained equilibrium in the bed. The selected kinetic model was 3-D diffusion (D3) and the frequency factor 4E-5 and zero activation energy were used for all the solid phases. Formation of wustite (FeO(s)) was prevented at temperatures lower than 650 °C and formation of solid iron (Fe_BCC(s)) was prevented at temperatures lower than 800 °C by excluding them from the equilibrium calculation.

Table 20. Compositions of the two exit flows (gas and tabbed melt (LIQUID+Slag)) of the furnace.

Products	**Gas [kg/thm]**	**Melt [kg/thm]**	**Liquid**	**wt-%**	**Slag**	**wt-%**
Ni	0.000	0.000	Ni	0.000	Na_2O	0.265
Fe	0.401	943.391	Fe	94.334	K_2O	0.100
Mn	0.003	2.052	Mn	0.198	Al_2O_3	9.380
Cr	0.000	0.363	Cr	0.036	SiO_2	34.693
V	0.002	1.239	V	0.109	$NaAlO_2$	0.403
Ti	0.036	2.294	Ti	0.065	$KAlO_2$	0.937
Ca	0.060	61.313	Ca	2.67E-06	CaO	40.410
K	1.607	0.967	K	0.000	FeO	0.021
S	0.405	3.006	S	0.014	MgO	10.182
P	0.033	0.529	P	0.053	MnO	0.041
Si	0.641	41.163	Si	0.677	CrO	1.86E-04
Al	0.390	11.376	Al	0.002	Ti_2O_3	0.606
Mg	0.062	13.025	Mg	2.51E-05	TiO_2	0.616
Na	0.493	0.656	Na	0.000	Na_2S	0.006
O	790.885	82.396	O	6.62E-05	K_2S	0.002
N	855.954	0.004	N	3.66E-04	Al_2S_3	0.236
C	376.885	53.036	C	4.511	SiS_2	0.910
H	6.712	0.000	H	0.000	$NaAlS_2$	0.010
					$KAlS_2$	0.021
					CaS	0.888
					FeS	4.45E-04
					MgS	0.243
					MnS	8.68E-04
					Ti_2S_3	0.014
					TiS_2	0.015

The model allows for follow-up of the chemical and phase changes as well as local energy and mass balances in different parts of the furnace as function of the feed composition. The model also simulates the recirculation of volatile species such as alkali metal compounds in the shaft.

6. Results of biomass drying and slow pyrolysis modelling

6.1 Mass and energy balances

Material balances for the different scenarios are presented in *Table 21*. The amount of bark received (Ba scenario) is much higher than *Raw materials as received* for the other scenarios. This is based on the volume of bark assumed to be available at the pulp mill. Additionally, the bark moisture content of 60 wt% is higher than that of the other raw materials. Accordingly, the biochar production amount (output) is higher for the Ba scenario, which is more than double the output of the other scenarios. *Air for burning* (feed) and *Flue gases* (output) are remarkably lower for the L3 and Ba scenarios. This is because in these scenarios not all pyrolysis gases are directly burned, moreover only a small share is burned to cover the endothermic reactions of pyrolysis. *Pyrolysis gases* (output) is the net gas amount produced for further use in the lime kiln.

Table 21. Material balances resulting from the modelling. These values are applied in calculating the production cost scenario estimates.

FEED	**Lignin L1 [k-tonne/a]**	**Lignin L2 [k-tonne/a]**	**Lignin L3 [k-tonne/a]**	**Black pellets BP [k-tonne/a]**	**Bark Ba [k-tonne/a]**
Raw material, as received	196	109	196	116	600
Air for burning	328	328	60	273	131
Sum (feed):	**525**	**438**	**257**	**388**	**730**

OUTPUTS	**Lignin L1 [k-tonne/a]**	**Lignin L2 [k-tonne/a]**	**Lignin L3 [k-tonne/a]**	**Black pellets BP [k-tonne/a]**	**Bark Ba [k-tonne/a]**
Biochar (at 50 °C)	44.2	44.2	44.2	44.2	96.0
Pyrolysis gases	*Burned*	*Burned*	53.0	*Burned*	137
Flue gas	393	393	72.1	344	164
Vapour from drying	87.3		87.3	0.52	333
Sum (outputs):	**525**	**438**	**257**	**388**	**730**

Energy balances based on higher heating values (HHV) are presented in *Table 22*. The Balas® simulation software uses HHV values and provides the energy related data in this form. As shown in *Table 21*, the bark heat content entering the system is much higher than the raw material heat content for the other scenarios. Similarly, the heat content of the biochar in the Ba scenario is higher, but less than 2 times the heat content in the other scenarios, while on a tonnage basis it is more than double. This difference is due to the lower HHV value estimation for bark-based biochar than the respective figure for the L1, L2 and L3 scenarios presented in *Table 13*. In scenarios L3 and Ba no excess steam is produced, yet some amount of steam is generated (output: *Internal energy for drying*) from the flue gases as a small share of the pyrolysis gases are burned to cover the endothermic reactions in pyrolysis. This steam is used for drying prior to pyrolysis, thus lowering the amount of steam needed from the pulp mill.

Scenarios L1, L2 and BP are self-sufficient in drying energy (marked with grey background) because all of the *Pyrolysis gases* are burned and heat is regenerated from the resulting flue gases as steam, which is then partly used for drying. The scenarios also provide excess steam as a surplus (output *Excess energy*), which is later used to generate electricity. Also, the output *Other losses* is higher for these scenarios due to losses from drying, burning and heat recovery.

Table 22. Energy balances resulting from the modelling. Expressed as the higher heating values, in accordance with the applied Balas® simulation software.

FEED	**Lignin L1** [MW HHV]	**Lignin L2** [MW HHV]	**Lignin L3** [MW HHV]	**Black pellets BP** [MW HHV]	**Bark Ba** [MW HHV]
Raw material, as received	86.4	86.1	86.4	75.2	175
Drying / preheating	14.5 internal	0.74 internal	14.5	0.97 internal	54.5
Air for burning	0.29	0.29	0.05	0.24	0.12
Sum (feed):	**101**	**87.1**	**101**	**76.4**	**229**

OUTPUTS	**Lignin L1** **[MW HHV]**	**Lignin L2** **[MW HHV]**	**Lignin L3** **[MW HHV]**	**Black pellets BP** **[MW HHV]**	**Bark Ba** **[MW HHV]**
Biochar (at 50 °C)	50.9	50.9	50.9	45.5	97.8
Pyrolysis gases			31.5		67.9
Internal energy for drying / preheating (steam)	14.5	0.74	1.10	0.97	2.54
Excess energy (steam)	13.3	27.1	None	21.7	None
Flue gas losses (130 °C)	6.47	6.47	1.19	6.27	2.89
Vapour losses from drying	8.14		8.14	0.05	31.1
Cooling losses of biochar	1.14	1.14	1.14	1.09	2.35
Other losses (pyrolysis, boiler section, drying)	6.68	0.79	6.88	0.79	24.9
Sum (outputs):	**101**	**87.1**	**101**	**76.4**	**229**

The respective simplified energy balances based on lower heating values (LHV), used in the calculation of production costs estimates, are presented in *Table 23*. The feed *Thermal load received to the system* and output *Excess energy for drying (steam)* are neglected for scenarios L1, L2 and BP because the steam is used internally. There is no direct additional cost associated with this arrangement.

Table 23. Simplified energy balances derived as lower heating values. These values are applied in the calculation of production cost scenario estimates.

FEED	**Lignin L1 [MW LHV]**	**Lignin L2 [MW LHV]**	**Lignin L3 [MW LHV]**	**Black pellets BP [MW LHV]**	**Bark Ba [MW LHV]**
Raw material, as received	73.1	80.5	73.1	69.8	133
Thermal load received to the system; raw material dried to 10% moisture	*internal*	*internal*	7.4	*internal*	28.3
Sum (feed):	**73.1**	**80.5**	**80.5**	**69.8**	**161**

OUTPUTS	**Lignin L1 [MW LHV]**	**Lignin L2 [MW LHV]**	**Lignin L3 [MW LHV]**	**Black pellets BP [MW LHV]**	**Bark Ba [MW LHV]**
Biochar (at 50 °C)	49.7	49.7	49.7	44.6	96.7
Pyrolysis gases			28.0		59.6
Excess energy for drying (steam)	*internal*	*internal*	1.10	*internal*	2.54
Excess energy (steam)	13.3	27.1		21.7	
Losses (L1& L2: drying losses included. Inside system boundaries) (L3 and Ba: drying losses excluded. Outside system boundaries)	10.1	3.76	1.74	3.5	2.48
Sum (outputs):	**73.1**	**80.5**	**80.5**	**69.8**	**161**

The various energy sources for drying are listed in *Table 24*. The integrated scenario (L3 and Ba) values are especially important for calculation of the production cost scenarios due to the costs associated with the supply of *External energy (steam)*. In these scenarios, 50% of the energy is assumed to be covered by hot water from the pulp mill, and the other half by steam, primarily using internally generated steam, with the remaining steam sourced from the pulp mill.

Table 24. Energy division for drying purposes from different sources. Integrated scenario (L3 and Ba) values are essential for the calculation of production cost scenarios.

Drying/ preheating energy division, losses included	**Lignin L1 [MW]**	**Lignin L2 [MW]**	**Lignin L3 [MW]**	**Black pellets BP [MW]**	**Bark Ba [MW]**
External energy (hot water)			7.23		27.2
External energy (steam)			6.12		24.7
Internal energy (steam)	14.5	0.74	1.10	0.97	2.54
Sum (drying/preheating energy):	**14.5**	**0.74**	**14.5**	**0.97**	**54.5**

6.2 Integration with a Kraft pulp mill

The Kraft pulp mill is seen as a viable option for the integration of biochar production. Currently, gasified bark is utilized as fuel for the lime kiln. Likewise, pyrolysis gas produced during carbonization could be combusted in the lime kiln or other boilers. This could provide a tempting alternative to stand-alone biochar processes or integration with steel mill processes. Two alternative scenarios are assessed. Biochar production integrated either to a i) stand-alone Kraft pulp mill or ii) pulp and paper integrate. Lignin and bark provide the raw materials for biochar production. These two scenarios are evaluated against two alternative reference mills (L3 reference and Ba reference), which are described in *Chapter 3.5*. The process integration analysis describes the effect of biochar production on the energy balance of the Kraft pulp mill. The findings are later used in *Chapter 7* when assessing the economic feasibility.

6.2.1 Biochar production from hydrolysis lignin in an integrated pulp & paper mill (Scenario L3)

Biochar production at the site of origin of lignin, integrated with a pulp and paper mill producing 800 000 air-dried tonnes of Kraft pulp per year, is illustrated in *Figure 27* (L3 scenario). The focus is on producing biochar and pyrolysis gases from lignin. The amount of lignin is 196 000 tonnes per year with 50% moisture content. The lignin is further dried to 10% moisture content before pyrolysis/carbonization. The biochar production volume is 44 200 tonnes per year and the biochar product is transported 160 km by railway to the iron/steel plant. The bark fraction, ca 400 000 tonnes per year at 60% moisture content, is combusted in the multi-fuel boiler without any changes in bark utilization vs. the reference pulp and paper mill. The reference mill (Baseline L3) is presented in *Section 3.5* and illustrated in *Figure 21.*

The hydrolysis lignin used is an additional raw material used by the integrated pulp and paper mill. Therefore, the effect of biochar production on the net electricity production of the integrate is minimal. The energy available for district heating is reduced by 50 GWh per year. This is due to the use of steam for bark drying. The amount of bio-oil used for firing the lime kiln is reduced from 33 000 to 20 000 tonnes/a as pyrolysis gas replaces part of the bio-oil.

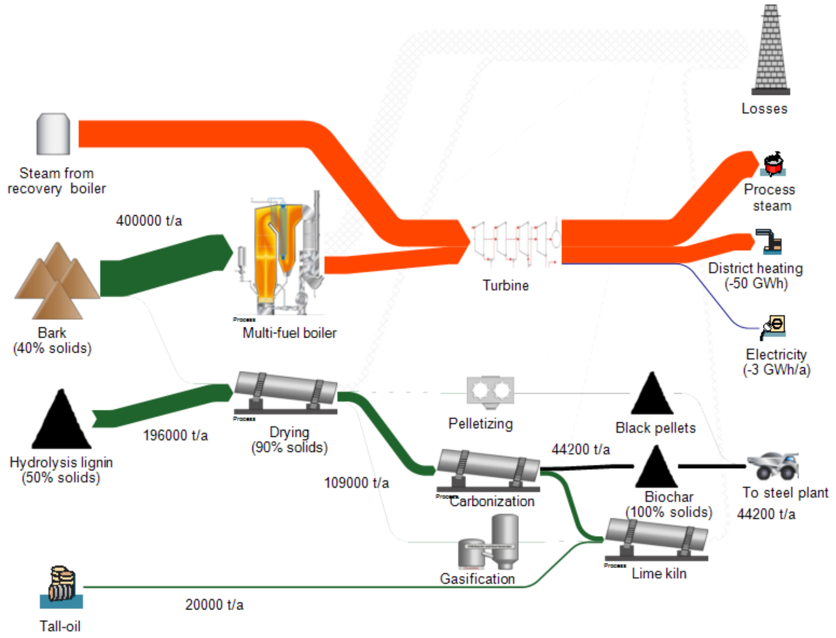
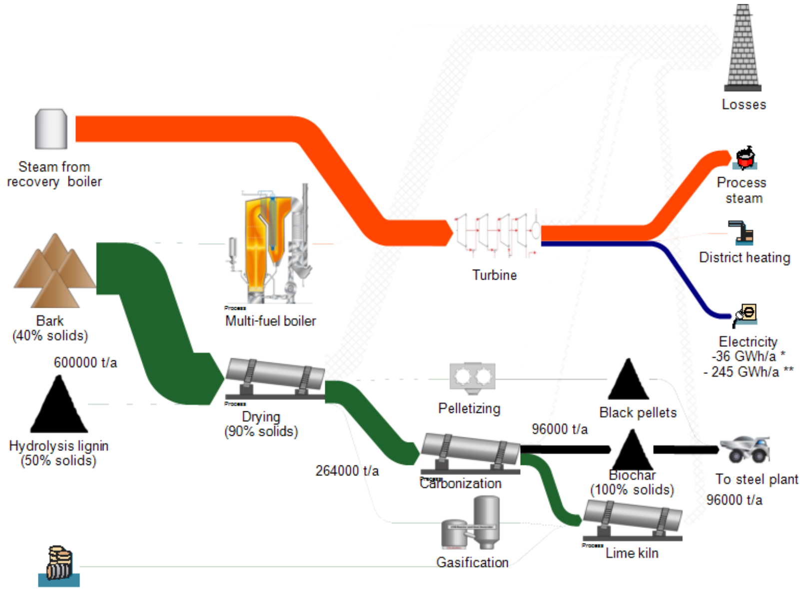


Figure 27. Integrated pulp and paper mill producing 44 200 tonnes of biochar per year (L3 scenario). Kraft pulp production is 800 000 air-dried tonnes per year. Bark is combusted in a multi-fuel boiler together with combustibles from other sources (e.g. bark, coal and wood). Hydrolysis lignin is carbonized and pyrolysis gases are used for firing the lime kiln together with bio-oil. Heat is utilized for district heating. (Baseline for L3 scenario is illustrated in Figure 21).

6.2.2 Biochar production from bark, integrated with a market pulp mill (Scenario Ba)

In the Ba scenario, biochar is produced from bark at an integrated pulp mill producing 1.2 million air-dried tonnes of softwood Kraft pulp per year (*Figure 28*). The total bark yield is 600 000 tonnes per year at 60% moisture content. The bark is used as a raw material for producing biochar and pyrolysis gases, instead of gasifying one half of the bark and burning the other half in the multi-fuel boiler, as in the baseline Ba reference mill. Pyrolysis gases are used as a fuel in the lime kiln. The bark is dried to 10% moisture content and then carbonized/pyrolysed, producing 96 000 tonnes of biochar per year. The biochar product is transported 275 km by rail to the iron/steel plant. The reference mill (Baseline Ba) is presented in *Section 3.5* and illustrated in *Figure 22*.

Net electricity production is reduced by 36 GWh per year if only the reduction related to biochar production is considered. Here it can be assumed that the multi-fuel boiler is able to accept additional biobased fuels in order to keep the production capacity constant. If the multi-fuel boiler is omitted (e.g. green-field plant) the effect is larger, with a reduction in electricity production of 245 GWh per year. This may be a valid case when considering bottlenecks of biomass availability in the future.



* Electricity reduction related to biochar production.
** Electricity reduction related to both biochar production and reduced combusted bark in the multi-fuel boiler.

Figure 28. Market pulp mill producing 1.2 million air-dried tonnes of softwood Kraft pulp per year (Ba scenario). Bark is carbonized and lime kiln is fired with pyrolysis gas; annual biocarbon production is 96 000 tonnes. Excess heat is converted to electricity in a condensing turbine. (Baseline for Ba scenario illustrated in Figure 22).

6.2.3 Black pellet production from bark, integrated with a market pulp mill (optional scenario Ba-BP)

The optional scenario of a market pulp mill with integrated black pellet production is illustrated in *Figure 29*. The entire flow of bark (600 000 tonnes/a, dry content 40%) is dried to 90% dry solids. Half of the dried bark is gasified and the lime kiln is fired with bark gas, as in the baseline Ba reference mill. The remaining half is processed to black pellet through thermal treatment and pelletizing. No biochar is produced. Black pellet production volume is 108 000 tonnes per year. A reduction in net electricity output is also observed. The reference mill is the baseline Ba mill (*Figure 22*).

Black pellet production uses steam for drying and for steam explosion of biomass. This will have effect on the energy balance when compared with the reference mill, although this is not assessed further here. The cost analysis for this additional option is also omitted as no biochar is produced.

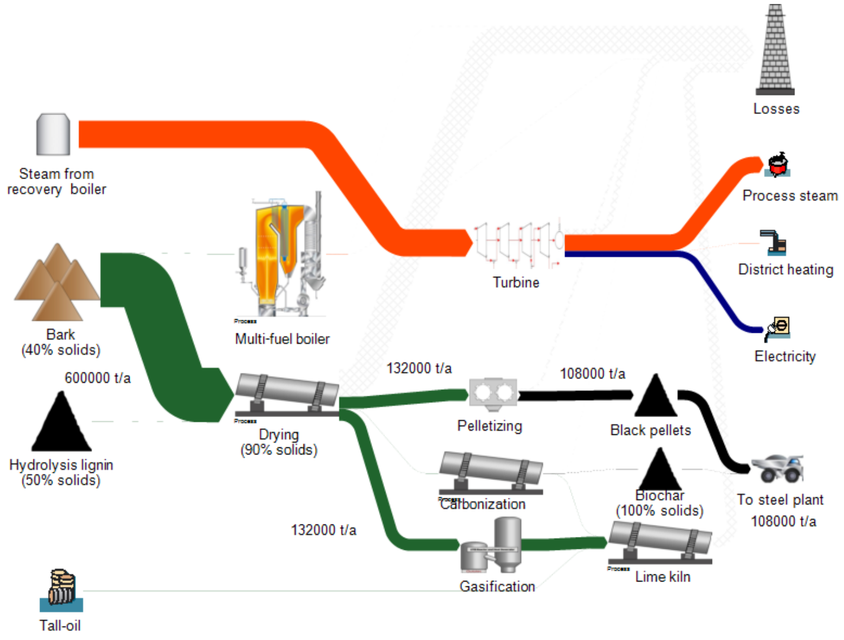


Figure 29. Market pulp mill producing 1.2 million air-dried tonnes of softwood Kraft pulp per year (optional Ba-BP scenario). 50% of bark is gasified and lime kiln is fired with bark gas; 50% of bark is processed to black pellets. Excess heat is converted to electricity in a condensing turbine.

6.2.4 Key process parameters for process integration with Kraft pulp mills

Two different Kraft pulp mill scenarios are evaluated. The largest differences between these two cases are: i) pulp production capacity (1.200 000 vs. 800 000 air-dried tonnes per year), ii) availability of excess biomass for biochar production (excess bark vs. hydrolysis lignin from external process), iii) turbine configuration (condensing vs. back-pressure turbine), and iv) utilization of low temperature heat (no usage vs. district heating). These variations in baselines should give a wider outlook regarding the possibilities for integration of biochar production with Kraft pulp mills.

Based on the process integration assessment, a list of key process parameters is given in *Table 25*. Parameters for both the baselines (references) and the evaluated integration scenarios are presented. An optional black pellet production scenario (Ba-BP) for a market pulp mill is also given.

Table 25. Key process parameters for process integration assessment. Kraft pulp mill production details and respective treatment of bark and hydrolysis lignin are presented. Applied reference mill model is described in Kangas et al. (2014) and Onarheim et al. (2017). (Baseline mills estimated based on Aluehallintovirasto 2017a; 2017b).

	Unit	**Pulp and paper mill**		**Market pulp mill**		
		Baseline L3	**Scenario L3 (biochar)**	Baseline	Scenario Ba-BP (black pellets)	**Scenario Ba (biochar)**
Kraft pulp mill						
Capacity	adt/a	800 000	**800 000**	1 200 000	1 200 000	**1 200 000**
Type of mill		Integrated P&P	**Integrated P&P**	market pulp	market pulp	**market pulp**
Chips	dry tonnes/a	1 600 000	**1 600 000**	2 400 000	2 400 000	**2 400 000**
Round wood	m^3/a	4 500 000	**4 500 000**	6 700 000	6 700 000	**6 700 000**
Bark	dry tonnes/a	160 000	**160 000**	240 000	240 000	**240 000**
Multi-fuel boiler						
Bark to boiler	tonnes/a	400 000	**400 000**	300 000	-	**-**
Dry content of bark	wt%	40%	**40%**	40%	-	**-**
HHV, bark	MJ/kg	21.8	**21.8**	21.8	-	**-**

	Unit	Pulp and paper mill		Market pulp mill		
		Baseline L3	**Scenario L3 (biochar)**	Baseline	Scenario Ba-BP (black pellets)	**Scenario Ba (biochar)**
Ethanol plant						
Hydrolysis lignin	tonnes/a	-	**196 000**	-	-	**-**
Dry content of hyd. lignin	wt%	-	**50%**	-	-	**-**
Carbonization or gasification						
Process		-	**slow pyrolysis**	gasification	gasification	**slow pyrolysis**
Raw material		-	**hydrolysis lignin**	bark	bark	**bark**
Dry content after drying		-	**90%**	90%	90%	**90%**
Raw material	dry tonnes/a	-	**98 000**	-	120 000	**240 000**
Biochar yield	wt%	-	**45%**	-	-	**40%**
Biochar/Black pellets prod.	dry tonnes/a	-	**44 200**	-	108 000	**96 000**
Lime kiln						
Capacity	tonne CaO /a	184 000	**184 000**	276 000	276 000	**276 000**
Heat demand	GWh/a	380	**380**	570	570	**570**
Fuel		bio-oil	**pyrolysis gas**	gasified bark	gasified bark	**pyrolysis gas**
HHV, Fuel	MJ/kg	41.8	**19.1**	6.5	6.5	**15.5**
Demand	tonnes/a	33 000	**72 000**	316 000	316 000	**132 000**
Losses						
Electricity	kWh/t steam		**-35**			**-110**
District heating	kWh/t steam					**-620**
Electricity, bark	GWh/a	-	**-**	-	-	**-209**
Electricity, steam	GWh/a	-	**-3**	-	-	**-36**
District heating	GWh/a	-	**-50**	-	-	**-**

adt = air-dried tonne

7. Biochar production costs

The estimated biochar production cost structures are presented for each scenario in *Figure 30*, *Figure 31, Figure 32*, *Figure 33* and *Figure 34*.

The capital cost is added as CAPEX, and refunds from fuel savings and sold electricity are also included. Production losses refers here to integrate scenarios where the use of steam in drying is assumed to reduce the steam load in the turbines, causing losses in electricity generation at the pulp mill's power plant. It also accounts for the reduction in thermal load generation for the district heating network in scenario L3. The electricity cost component refers to process electricity consumption. Fuel savings refers to the lime kiln fuel savings in scenarios L3 and Ba through replacement with pyrolysis gas (as equal to the thermal load available). The 'Other costs' cost component includes insurance (as a percentage of CAPEX) and other costs (as a percentage of sales).

In the tables presenting the cost effectiveness of replacing PCI coal with biochar it is estimated that burning PCI coal emits 2.375 tonnes of CO_2. The basis for this figure is explained in *Section 2.1*. Related emission trading costs are applied. For example, if the PCI coal price is 125 €/tonne and EUA trading is at 25 €/tonne of CO_2, the equivalent cost of biochar is 184 €/tonne.

It should also be noted that replacing blast furnace PCI coal with biochar may not be exactly 1:1 in tonnage terms. *Table 26* gives the heating values and ash contents for PCI and for lignin and black pellet based biochars. The heating value of lignin biochar is higher than that of PCI coal, indicating that the replacement ratio could be 0.94 kg / kg PCI coal to maintain energy balance. Similarly, for black pellet and bark biochars the replacement ratio would be 1.05 kg / kg PCI coal. If the ash content is omitted and the heating values are calculated only for the ash-free fraction, the biochars are at the same level. This indicates that lignin biochar benefits slightly when compared to black pellet or bark biochars due to ash content differences. However, there may be differences in reactivity, effects of ash components in the blast furnace and injected coal, or biochar participation in the reduction reactions. Future research in this area is needed. In the following cost comparisons of biochar costs with PCI costs, the replacement ratios are assumed to be 1:1 in tonnage terms due to these uncertainties.

Table 26. Ash contents and heating values for PCI and for lignin and black pellet based biochars. Values obtained from experiments made by University of Oulu.

	PCI	**biochar**	
Raw material	Coal	Lignin	Black pellet
Ash content [wt%]	10.1%	0.7%	10.8%
LHV, dry-based [MJ/kg]	30.5	32.4	29.0
Replacement ratio [kg/kg PCI]		94%	105%
LHV, ash free [MJ/kg]	*34.0*	*32.6*	*32.6*

7.1 Stand-alone production – biochar produced at end-user site

The scenarios and assumptions are described in detail in *Section 3.6*.

7.1.1 Scenario L1: Lignin transported at 50% moisture to the production site

In this scenario lignin is pressed to 50% moisture content before transportation 200 km by rail to the biochar production site, which is assumed to be in close connection with the iron/steel plant. Pressing costs at the site of origin of the lignin are not taken into account, as they are assumed to be integral to the ethanol plant process, see *Subsection 3.6.1.* These costs are assumed to be covered by the higher heat content of the dried raw material (thus higher valuation) when compared to the heat content before pressing. The pyrolysis gases are assumed to be burned for steam generation. The steam is used in the drying of the lignin to 10% moisture content before the pyrolysis, and the excess steam is assumed to be available for electricity generation. The electricity generation efficiency is set to 35% of the available steam thermal load.

Biochar production costs are estimated to be 405 €/tonne, taking also into account the use of excess steam for electricity generation (*Figure 30*). The cost effectiveness of replacing PCI coal with biochar is presented in *Table 27.*

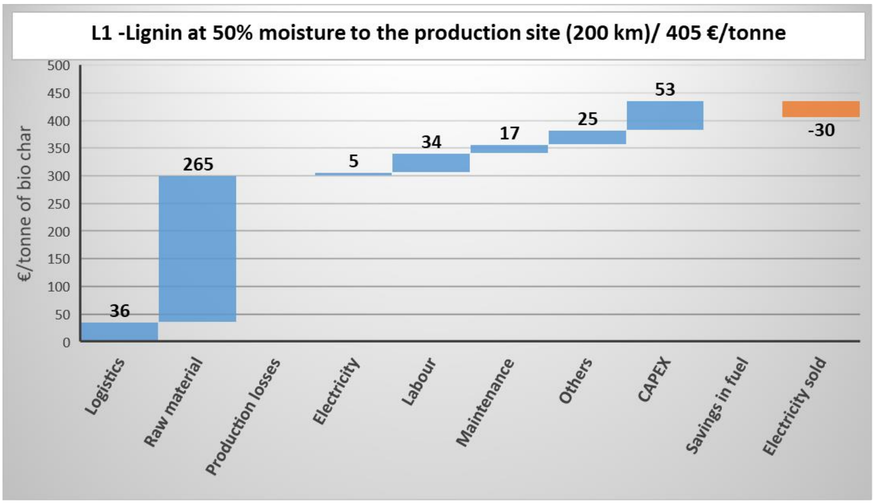


Figure 30. Production cost structure for the L1 scenario. Lignin is transported to the biochar production site at 50% moisture content. Transport distance 200 km.

Table 27. Cost-effectiveness of replacing PCI coal with biochar in scenario L1. Emission trading cost is included in the cost of burning PCI coal. Coloured cells indicate the viability of replacing PCI coal with biochar, blue meaning viable. Estimated production cost in the scenario: 405 €/tonne of biochar.

Cost of burning PCI coal, including emission trading EUA costs											
Coal price	Emission trading EUA price [€/tonne CO$_2$]										
[€/tonne]	0	5	10	15	20	25	30	35	40	45	50
	PCI coal and EUA [€/tonne]										
25	25	37	49	61	73	84	96	108	120	132	144
50	50	62	74	86	98	109	121	133	145	157	169
75	75	87	99	111	123	134	146	158	170	182	194
100	100	112	124	136	148	159	171	183	195	207	219
125	125	137	149	161	173	184	196	208	220	232	244
150	150	162	174	186	198	209	221	233	245	257	269
175	175	187	199	211	223	234	246	258	270	282	294
200	200	212	224	236	248	259	271	283	295	307	319
225	225	237	249	261	273	284	296	308	320	332	344
250	250	262	274	286	298	309	321	333	345	357	369
275	275	287	299	311	323	334	346	358	370	382	394
300	300	312	324	336	348	359	371	383	395	407	419
Cost – Less than 355 €/tonne					**Cost – Between 395 and 355 €/tonne**			**Cost – More than 395 €/tonne**			

7.1.2 Scenario L2: Lignin transported at 10% moisture to the production site

In the L2 scenario lignin is pressed and further dried to 10% moisture content before transportation by rail ca. 200 km to the biochar production site, which is assumed to be in close connection to the iron/steel plant. Pressing and drying costs at the site of origin of the lignin are not taken into account, being part of the ethanol plant process, as explained in *subsection 4.5.1.* The pyrolysis gases are assumed to be burned for steam generation. No further drying is needed at the biochar production site. Excess steam generated is assumed to be available for electricity generation. Electricity generation efficiency is set to 35% of the available steam thermal load.

The production cost estimate for scenario L2 is 379 €/tonne of biochar, taking also into account utilization of excess steam for electricity generation (*Figure 31*). The cost effectiveness of replacing PCI coal with biochar is presented in *Table 28*.

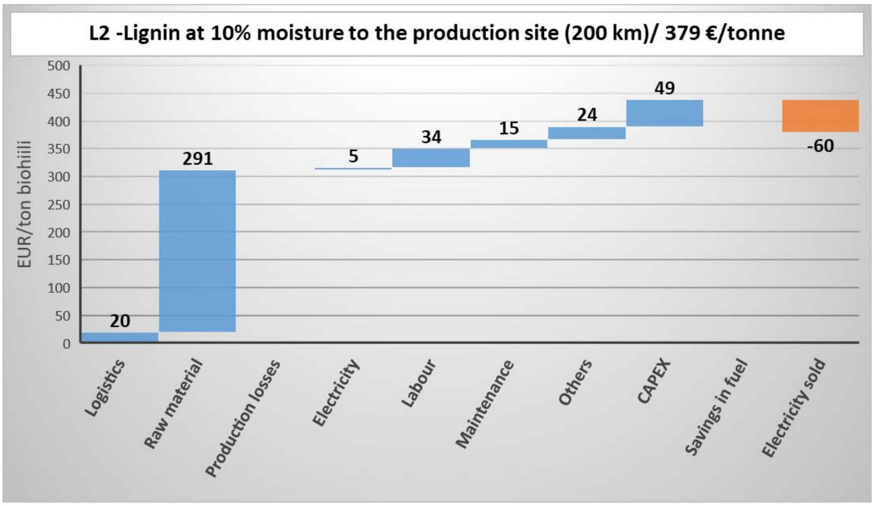


Figure 31. Production cost structure for the L2 scenario. Lignin is transported to the biochar production site at 10% moisture content. Lignin is dried at the location of origin. Transport distance 200 km.

Table 28. Cost-effectiveness of replacing PCI coal with biochar in scenario L2. Emission trading cost is included in the cost of burning PCI coal. Coloured cells indicate the viability of replacing PCI coal with biochar, blue meaning viable. Estimated production cost in the scenario: 379 €/tonne of biochar.

Cost of burning PCI coal, including emission trading EUA costs											
Coal price	Emission trading EUA price [€/tonne CO_2]										
[€/tonne]	0	5	10	15	20	25	30	35	40	45	50
	PCI coal and EUA [€/tonne]										
25	25	37	49	61	73	84	96	108	120	132	144
50	50	62	74	86	98	109	121	133	145	157	169
75	75	87	99	111	123	134	146	158	170	182	194
100	100	112	124	136	148	159	171	183	195	207	219
125	125	137	149	161	173	184	196	208	220	232	244
150	150	162	174	186	198	209	221	233	245	257	269
175	175	187	199	211	223	234	246	258	270	282	294
200	200	212	224	236	248	259	271	283	295	307	319
225	225	237	249	261	273	284	296	308	320	332	344
250	250	262	274	286	298	309	321	333	345	357	369
275	275	287	299	311	323	334	346	358	370	382	394
300	300	312	324	336	348	359	371	383	395	407	419
Cost – Less than 329 €/tonne				**Cost – Between 369 and 329 €/tonne**				**Cost – More than 369 €/tonne**			

7.1.3 Scenario BP: Black pellets transported at 10% moisture to the production site

In the BP scenario black pellets are transported at 10% moisture 275 km by rail to the biochar production site, which is assumed to be in close connection to the iron/steel plant. Pyrolysis gases are assumed to be burned for steam generation. No further drying is needed at the biochar production site. Excess steam generated is assumed to be available for electric power production. Electricity generation efficiency is set to 35% of the available steam thermal load.

The production cost estimate for the BP scenario is 363 €/tonne of biochar, taking also into account the utilization of excess steam for electricity generation (*Figure 32*). The cost effectiveness of replacing PCI coal with biochar is presented in *Table 29*.

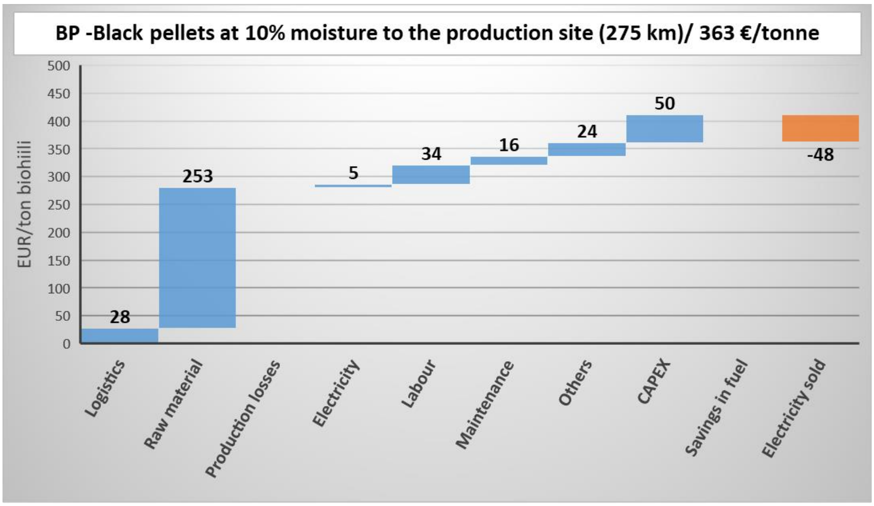


Figure 32. Production cost structure for the BP scenario. Black pellets transported to the biochar production site at 10% moisture content. No further drying is needed. Transport distance 275 km.

Table 29. Cost-effectiveness of replacing PCI coal with biochar in scenario BP. Emission trading cost is included in the cost of burning PCI coal. Coloured cells indicate the viability of replacing PCI coal with biochar, blue meaning viable. Estimated production cost in the scenario: 363 €/tonne of biochar.

Cost of burning PCI coal, including emission trading EUA costs											
Coal price	Emission trading EUA price [€/tonne CO$_2$]										
[€/tonne]	0	5	10	15	20	25	30	35	40	45	50
	PCI coal and EUA [€/tonne]										
25	25	37	49	61	73	84	96	108	120	132	144
50	50	62	74	86	98	109	121	133	145	157	169
75	75	87	99	111	123	134	146	158	170	182	194
100	100	112	124	136	148	159	171	183	195	207	219
125	125	137	149	161	173	184	196	208	220	232	244
150	150	162	174	186	198	209	221	233	245	257	269
175	175	187	199	211	223	234	246	258	270	282	294
200	200	212	224	236	248	259	271	283	295	307	319
225	225	237	249	261	273	284	296	308	320	332	344
250	250	262	274	286	298	309	321	333	345	357	369
275	275	287	299	311	323	334	346	358	370	382	394
300	300	312	324	336	348	359	371	383	395	407	419
Cost – Less than 313 €/tonne				**Cost – Between 353 and 313 €/tonne**				**Cost – More than 353 €/tonne**			

7.2 Pulp mill integrated biochar production

Two alternative scenarios for integrating biochar production with a Kraft pulp mill were evaluated: i) integrated pulp and paper mill (scenario L3) and ii) market pulp mill (scenario Ba). The scenarios and key integration parameters are described in *Section 6.2.*

7.2.1 Scenario L3: Lignin at 50% moisture, biochar transported to steel plant

In this scenario, hydrolysis lignin production is integrated with a pulp mill producing 800 000 air-dried tonnes of Kraft pulp per year (L3 scenario). The bark fraction, 400 000 tonnes per year at 60% moisture content, is combusted in the multi-fuel boiler, as in the baseline mill. The focus is on producing biochar and pyrolysis gases from lignin. The amount of lignin is 196 000 tonnes per year at 50% moisture content. The lignin is further dried to 10% moisture content before pyrolysis/carbonization. The biochar production volume is 44 200 tonnes per year and the biochar product is transported 160 km by rail to the iron/steel plant.

Pressing costs are not taken into account, being part of the ethanol plant process. Drying utilizes hot water and steam, each 50% of the necessary heat load. It is assumed that the pulp mill can provide the hot water and steam. Pyrolysis gases (net) are redirected for further use as a fuel for the lime kiln at the pulp mill, replacing original fuels. To cover the endothermic reactions during pyrolysis, a share of the generated pyrolysis gases are burned to provide the necessary heat load. From these flue gases some steam is generated for drying lignin, thus reducing the need for steam from the pulp mill.

The effect of biochar production on the energy balances of an integrated pulp and paper mill are following: i) annual net electricity production of the integrate is reduced by 3 GWh, and ii) the amount of available heat for district heating is reduced by ~50 GWh per year, see *Figure 27* and *Table 25*. These reductions are due to the increased demand for process steam within the biochar production processes. To compensate for these losses, additional biomass can be burned in the multi-fuel boiler if such biomass is available and if the boiler capacity is not exceeded.

The production cost estimate for scenario L3 is 275 €/tonne of biochar, taking also into account the use of net pyrolysis gases as a fuel for the lime kiln (*Figure 33*). The cost effectiveness of replacing PCI coal with biochar is presented in *Table 30*.

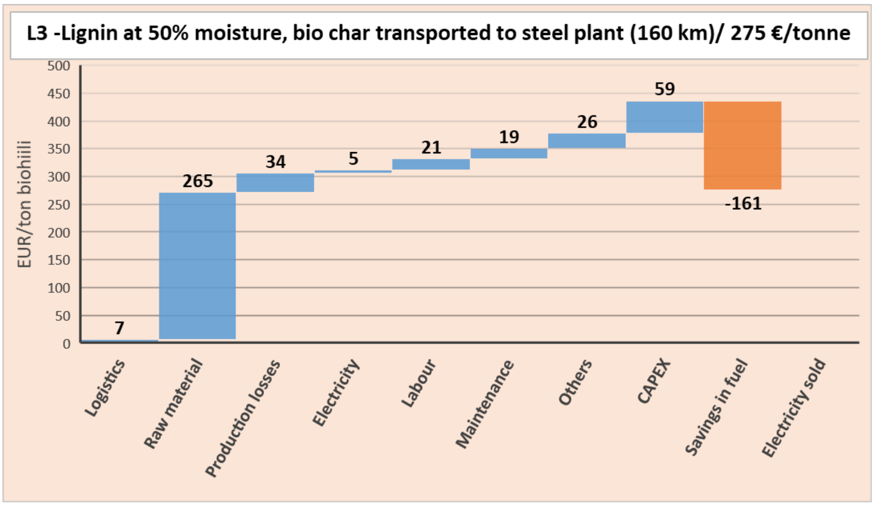


Figure 33. Production cost structure for the L3 scenario. Biochar produced is transported to the iron/steel plant. Transport distance 160 km.

Table 30. Cost-effectiveness of replacing PCI coal with biochar in scenario L3. Emission trading cost is included in the cost of burning PCI coal. Coloured cells indicate the viability of replacing PCI coal with biochar, blue meaning viable. Estimated production cost in the scenario: 275 €/tonne of biochar.

Cost of burning PCI coal, including emission trading EUA costs											
Coal price	Coal and EUA [€/tonne CO$_2$]										
[€/tonne]	0	5	10	15	20	25	30	35	40	45	50
	PCI coal and EUA [€/tonne]										
25	25	37	49	61	73	84	96	108	120	132	144
50	50	62	74	86	98	109	121	133	145	157	169
75	75	87	99	111	123	134	146	158	170	182	194
100	100	112	124	136	148	159	171	183	195	207	219
125	125	137	149	161	173	184	196	208	220	232	244
150	150	162	174	186	198	209	221	233	245	257	269
175	175	187	199	211	223	234	246	258	270	282	294
200	200	212	224	236	248	259	271	283	295	307	319
225	225	237	249	261	273	284	296	308	320	332	344
250	250	262	274	286	298	309	321	333	345	357	369
275	275	287	299	311	323	334	346	358	370	382	394
300	300	312	324	336	348	359	371	383	395	407	419
Cost – Less than 225 €/tonne				**Cost – Between 265 and 225 €/tonne**				**Cost – More than 265 €/tonne**			

7.2.2 Scenario Ba: Bark at 60% moisture, biochar transported to steel plant

Biochar is produced from bark at the original pulp mill site and integrated with the mill producing 1.2 million air-dried tonnes of softwood Kraft pulp per year (Ba scenario). The total bark yield is 600 000 tonnes per year at 60% moisture content. The bark is used as a raw material for producing biochar and pyrolysis gases instead of gasifying half and burning half in the multi-fuel boiler. Pyrolysis gases are used as a fuel in the lime kiln. The bark is first dried to 10% moisture content and then carbonized/pyrolysed producing 96 000 tonnes of biochar per year. The biochar product is transported 275 km by rail to the iron/steel plant.

Bark drying utilizes hot water and steam, each meeting 50% of the necessary heat load. It is assumed that the pulp mill can provide the hot water and steam. The pyrolysis gases (net) are redirected for further use as a fuel for the lime kiln at the pulp mill, replacing the gasified bark used in the baseline scenarios. To cover the endothermic reactions during pyrolysis, a share of the generated pyrolysis gases are burned to provide the necessary heat load. From these flue gases some steam is generated for drying lignin, reducing the need for steam from the pulp mill.

The energetic value of the bark is not converted to electricity but to biochar in this scenario. The effect of biochar production on the energy balance of a market pulp mill is a ca. 4% reduction in net electricity output of the mill due to steam taken for drying (from 1000 GWh/a to 964 GWh/a) and a further reduction of ca. 25% if the bark taken for pyrolysis is not replaced with other bio-combustibles (from 1000 GWh/a to 755 GWh/a), see *Figure 28* and *Table 25*. Here it is assumed that a multi-fuel boiler is available at the site, and reduction of bark is compensated by using additional biofuels within the boiler. If a greenfield mill is considered, a significantly smaller boiler may be considered if bark carbonization technology is chosen instead of combustion technology.

The production cost estimate for the Ba scenario is 252 €/tonne of biochar, taking also into account the use of net pyrolysis gases as fuel for the lime kiln (*Figure 34*). The cost effectiveness of replacing PCI coal with biochar is presented in *Table 31*.

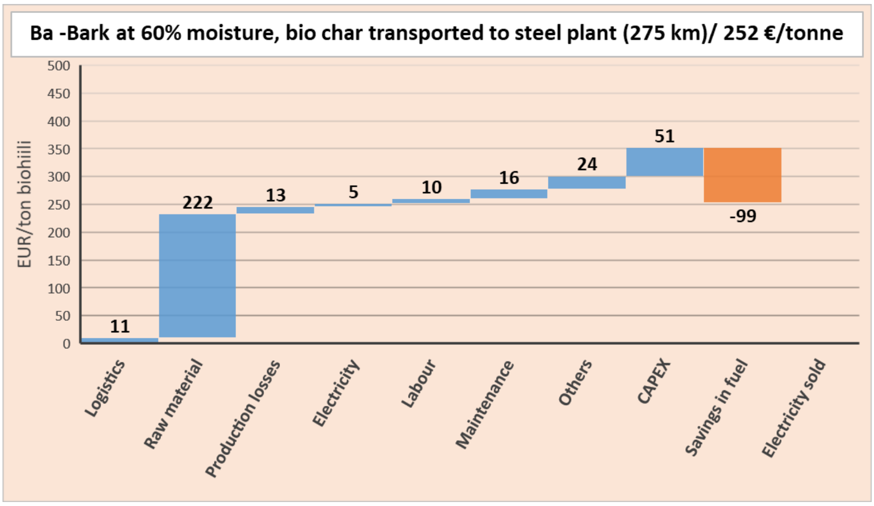


Figure 34. Production cost structure of the Ba scenario. Biochar produced (from bark) is transported to the iron/steel plant. Transport distance 275 km.

Table 31. Cost-effectiveness of replacing PCI coal with biochar in the Ba scenario. Emission trading cost is included in the cost of burning PCI coal. Coloured cells indicate the viability of replacing PCI coal with biochar, blue meaning viable. Estimated production cost in the scenario: 252 €/tonne of biochar.

Cost of burning PCI coal, including emission trading EUA costs											
Coal price	Emission trading EUA price [€/tonne CO$_2$]										
[€/tonne]	0	5	10	15	20	25	30	35	40	45	50
	PCI coal and EUA [€/tonne]										
25	25	37	49	61	73	84	96	108	120	132	144
50	50	62	74	86	98	109	121	133	145	157	169
75	75	87	99	111	123	134	146	158	170	182	194
100	100	112	124	136	148	159	171	183	195	207	219
125	125	137	149	161	173	184	196	208	220	232	244
150	150	162	174	186	198	209	221	233	245	257	269
175	175	187	199	211	223	234	246	258	270	282	294
200	200	212	224	236	248	259	271	283	295	307	319
225	225	237	249	261	273	284	296	308	320	332	344
250	250	262	274	286	298	309	321	333	345	357	369
275	275	287	299	311	323	334	346	358	370	382	394
300	300	312	324	336	348	359	371	383	395	407	419
Cost – Less than 202 €/tonne				**Cost – Between 242 and 202 €/tonne**				**Cost – More than 242 €/tonne**			

7.3 Biochar production cost sensitivity to raw material price

Raw material costs dominate the biochar production cost in every scenario, as visualized in *Figure 35* and *Figure 36*.

The baseline price for raw material in energy terms was selected to be 20 €/MWh. The tonnage based prices are back-calculated from the energy cost by applying the heating values at the given moisture content. The share of the raw material cost varies from 66% to 61% of the total costs while neglecting integration benefits and additional income from electricity generation in stand-alone production scenarios. If we take into account these benefits and additional incomes on the costs (by subtracting), the raw material cost share varies between 96% and 65%.

For example, if the bark price in Ba scenario would be 15 €/MWh (26.6 €/tonne), as nearly reached in July 2018 according to the PIX Forest Biomass Finland Index subindex for bark (Metsälehti 2018), the production cost would be only 197 €/tonne of biochar. This can be seen from the curve in *Figure 35*. The baseline production cost value for the raw material cost of 20 €/MWh was 252 €/tonne, thus the reduction in production cost is 55 €/tonne, and the production cost share of the baseline value is 78%.

The same result is also attained using the figures in *Table 32*, which give the respective changes in raw material cost (Δ €) for various scenarios, e.g. by taking the value 11.1 (Δ €/tonne production cost)/(Δ €/MWh raw material), multiplying this with 5 €/MWh (20 €/MWh-15 €/MWh) and subtracting the resulting figure of 55.5 €/tonne from the baseline production cost value of 252 €/tonne.

Furthermore, if we estimate a EUA price of 25 €/tonne of CO_2 (see *Section 2.1*), as was reached in 2018, the biochar production cost equivalent PCI coal price would be between 125 and 150 €/tonne (according to the figures in *Table 31*). If the market price of PCI coal is as above, it is economically beneficial to replace PCI coal with the biochar.

We can estimate the PCI coal price to be ca. 135 €/tonne, based on the coal price of 90 €/tonne (reached during 2018, without excise taxes) and further assuming that PCI coal is ca. 1.5 times the market coal price. Thus, the estimated PCI coal market price is in the range mentioned above, and replacing PCI coal with bark biochar is close to being an economically beneficial option.

The tonnage-based substitution mass ratio of PCI coal and biochar is set to 1:1, as explained above in *Chapter 7*.

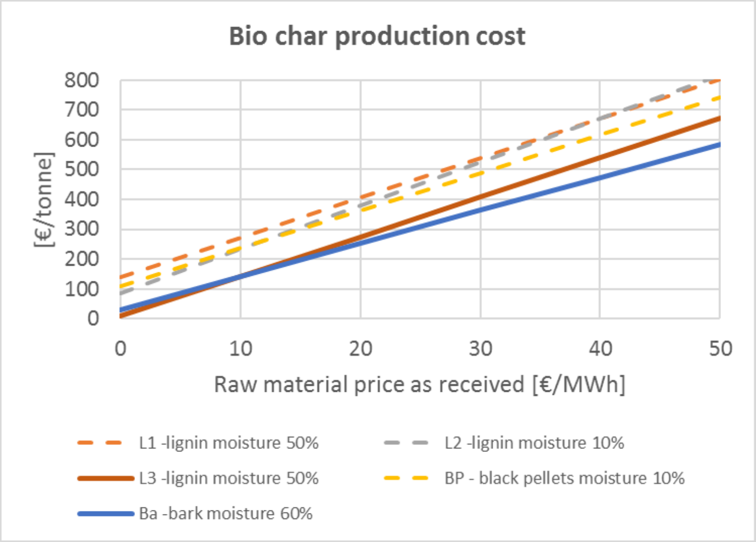


Figure 35. Effects of heat load based raw material prices on biochar production costs. Stand-alone production scenarios are visualized with dashed lines. Baseline situation is 20 €/MWh for each scenario.

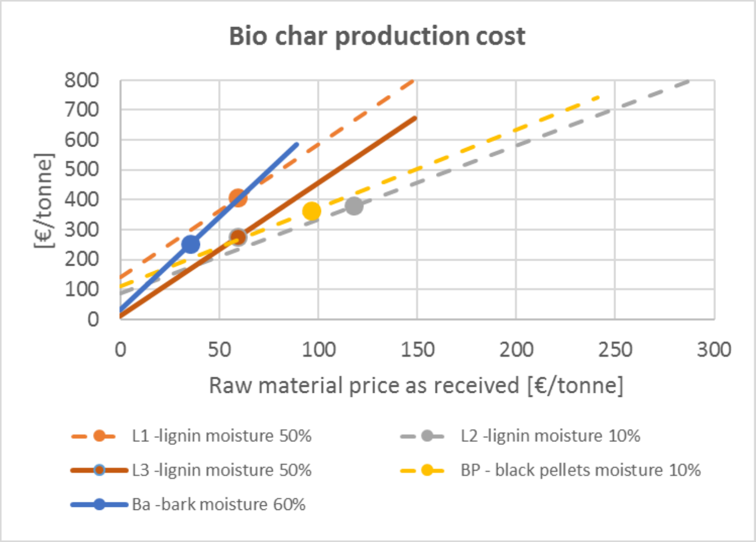


Figure 36. Effects of tonnage-based raw material prices on biochar production costs. Stand-alone production scenarios are visualized with dashed lines. The round data point markers represent the baseline scenarios, back-calculated from 20 €/MWh raw material price and expressed here in tonnage terms.

Table 32. Effect of raw material price change on biochar production cost for the evaluated scenarios. Raw material price change is expressed both in heat load and tonnage terms.

Scenario	Δ €/tonne production cost/ Δ €/tonne raw material	Δ €/tonne production cost/ Δ €/MWh raw material
L1, lignin moisture 50%	4.44	13.2
L2, lignin moisture 10%	2.47	14.6
L3, lignin moisture 50%	4.44	13.2
BP, black pellet moisture 10%	2.62	12.6
Ba, bark moisture 60%	6.25	11.1

7.4 Comparison of biochar production costs

The evaluated scenarios are compared in *Figure 37*. The pulp mill integrated scenarios L3 (lignin) and Ba (bark) gain remarkable benefits from integration when integration benefits and drawbacks are allocated to biochar production. These benefits and drawbacks are discussed further in *Chapter 8*.

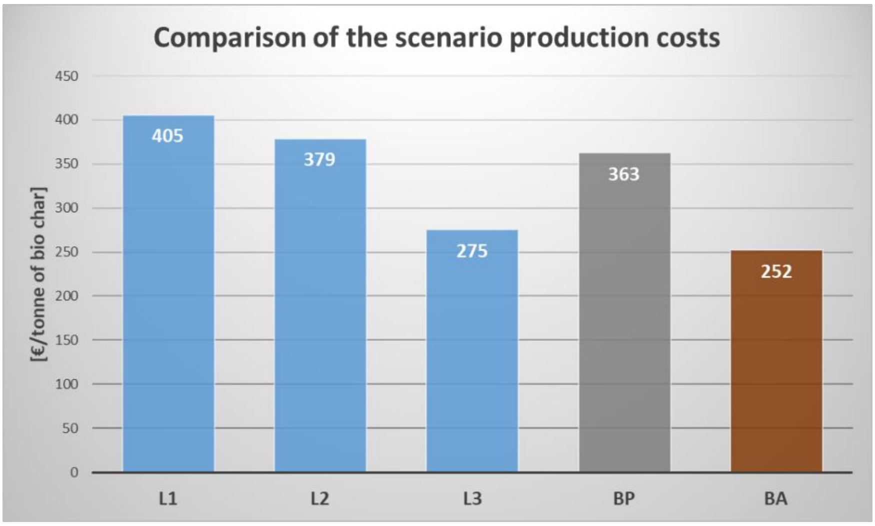


Figure 37. Comparison of the evaluated scenarios. Blue fill colour represents the lignin scenarios (L1, L2 and L3), grey colour the black pellet scenario (BP), and brown colour the bark scenario (Ba). Scenarios with pulp mill integration are L3 and Ba.

The scenario cost structures are compared in *Figure 38*, with fewer cost components than shown for each scenario separately. Total energy includes the sum of electricity and district heating production losses, process electricity consumption, fuel savings, and sold electricity. The cost component 'Other' is the sum of logistics, labour, maintenance and other costs.

'Raw material' costs, based on energy content as received, dominate the production costs. 'Total energy' is presented as a negative value, as its effect on total costs is positive for all scenarios. The L3 and Ba scenarios benefit most due to savings in original fuels at the lime kilns, as original fuel is replaced by pyrolysis gas. In 'Others' costs, logistic costs create variation due to the different transport distances and weights of the transported material (as raw material or as biochar). Variations in capital expenditure estimates are small.

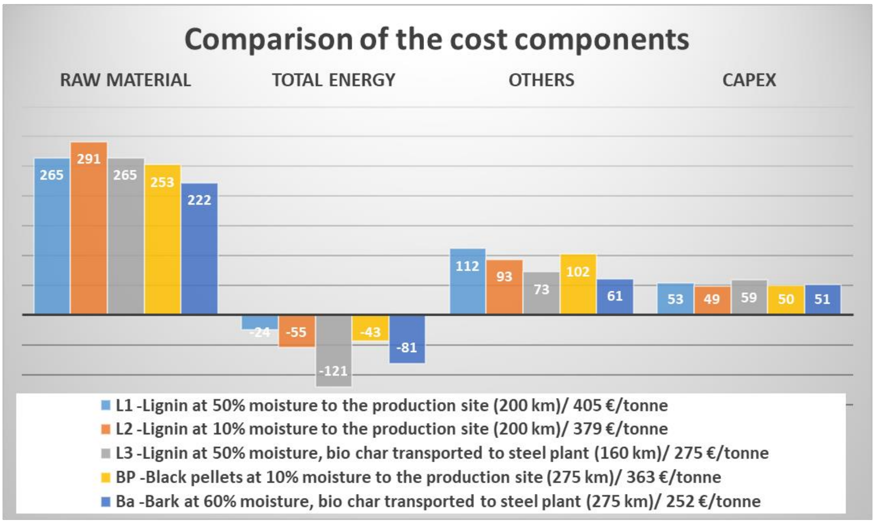


Figure 38. Cost structure comparison with fewer cost components than shown for each scenario separately.

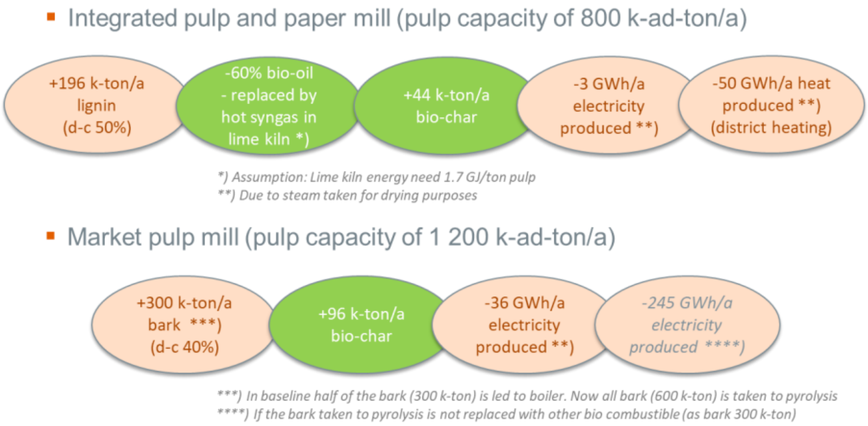
8. Effects of pulp mill or pulp and paper mill integration

An additional bio-product, biochar, can be produced from the excess biomass available at the pulp mill or the pulp and paper mill, such as bark, or side-product hydrolysis lignin from the ethanol production plant connected to the mill. In pulp mills and pulp and paper mills low-temperature excess heat (hot water) and excess steam is often available to be used for drying biomass prior to pyrolysis. Hot pyrolysis gases can be used to replace other fuel usages, such as in the lime kiln, thus releasing the original biofuels for other uses, or reducing the use of fossil fuels. The volume of biomass is reduced during pyrolysis and an energy-intensive, dense and stable stabilized product, biochar pellets, is obtained for efficient storage and transportation instead of storing and transporting large volumes of raw materials, such as hydrolysis lignin or bark.

Two pulp mill or pulp and paper mill integrated scenarios were studied, L3 (raw material hydrolysis lignin) and Ba (raw material bark), as presented in *Figure 39*. Based on estimated biochar production costs, both scenarios look promising, being close to economically beneficial solutions. The abovementioned integration effects apply to these scenarios. As benefits, hot pyrolysis gases are burned in the lime kiln, replacing original fuels, and excess low temperature heat as hot water and excess steam are utilized for biomass drying. Negative effects are caused by the use of steam for drying, which reduces the amount of steam used for electricity production (L3 and Ba) and for district heat generation (L3).

In Ba (lower case in *Figure 39*), all bark from the pulp mill is carbonized to biochar, whereas in the reference pulp mill half of the bark is burned in the boiler and half is gasified to provide syngas for the lime kiln. Steam usage for bark drying causes a reduction in electricity generation of ca. 36 GWh/a. Furthermore, if bark taken from the boiler is not replaced with other bio-combustibles, the reduction is ca. 245 GWh/a. For production cost evaluation, it is assumed that a multi-fuel boiler is available at the site, and reduction in bark is compensated by using additional biofuels in the boiler, and assuming that the cost of this fuel corresponds to bark. In production cost evaluation, the price of bark led to carbonization is set based on energy content. Furthermore, if a greenfield mill is considered, a significantly smaller boiler may be on option if bark carbonization technology is chosen instead of combustion technology. The capacity of the Ba scenario is remarkably higher than the other scenarios, based on the assumption that the needed amount of bark is available at the pulp mill. In the Ba scenario hot pyrolysis gases are assumed to be sufficient to cover the fuel needs of the lime kiln.

In L3 (upper scenario in *Figure 39*), hydrolysis lignin is used as the raw material for carbonization, producing biochar. The use of steam for biomass drying is estimated to cause a 3 GWh/a reduction in electricity generation. In the pulp and paper mill in question, a combined heat and power (CHP) plant also generates district heat, and a further reduction in district heat generation is estimated at 50 GWh/a. In the L3 scenario an estimated 60% of lime kiln original fuels (of which 20 wt% are assumed to be fossil-based heavy fuel oil) could be replaced by hot pyrolysis gases.



- Integrated pulp and paper mill (pulp capacity of 800 k-ad-ton/a)

**) Assumption: Lime kiln energy need 1.7 GJ/ton pulp*
***) Due to steam taken for drying purposes*

- Market pulp mill (pulp capacity of 1 200 k-ad-ton/a)

****) In baseline half of the bark (300 k-ton) is led to boiler. Now all bark (600 k-ton) is taken to pyrolysis*
*****) If the bark taken to pyrolysis is not replaced with other bio combustible (as bark 300 k-ton)*

Figure 39. Effects of pulp or pulp and paper mill integrated scenarios.

9. Environmental impact results

9.1 System boundaries and life cycle inventory of the integrated biochar production scenarios

9.1.1 System boundaries

The reference cases represent systems where the pulp mills operate without biochar production. Bark is used as a fuel to produce energy that is utilized both internally and externally. The energy for the lime kiln is produced from bio-oil and heavy fuel oil in L3, and from bark in Ba. Energy that is not needed internally is sold as electricity or district heating. Integrating biochar production with the pulp mill changes the material and energy flows. The biochar is produced by slow pyrolysis, which yields, in addition to biochar, pyrolysis gases that can be burned to produce energy. This is utilized in the lime kiln, resulting in reduced need for other energy sources. The system boundaries for the L3 reference case are presented in *Figure 40*.

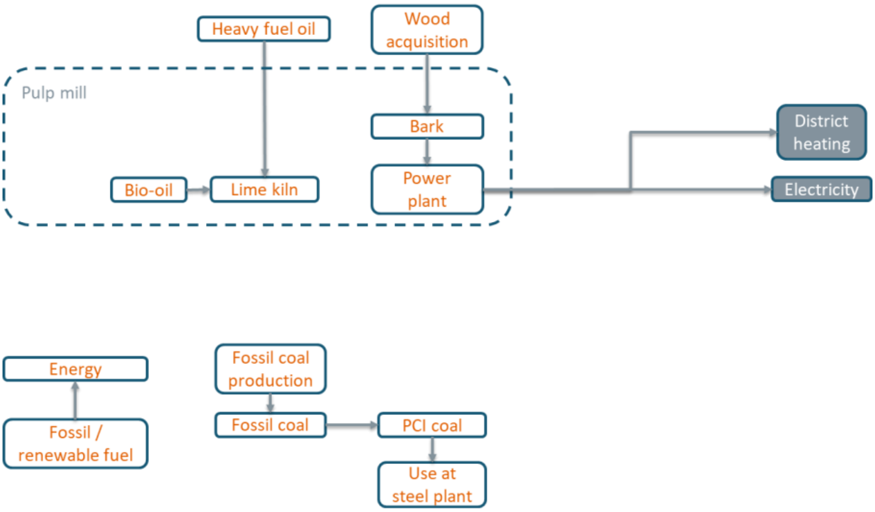


Figure 40. System boundaries for the L3 reference case. The pulp mill operates conventionally, and the excess energy is sold as district heating and electricity. The coal used in steel production is fossil coal.

In L3, part of the bio-oil and heavy fuel oil used in the lime kiln is replaced with the energy from pyrolysis gases, and the excess bio-oil can be used as fuel elsewhere. The main raw material for the pyrolysis process is hydrolysis lignin, a side stream from another process (sawdust refining to bioethanol). The lignin is available at 50% solids content and has to be dried before feeding it to the slow pyrolysis process.

This consumes heat, which reduces the amount of district heating and electricity that can be sold to external companies. The system boundaries of the scenario L3 are presented in *Figure 41*.

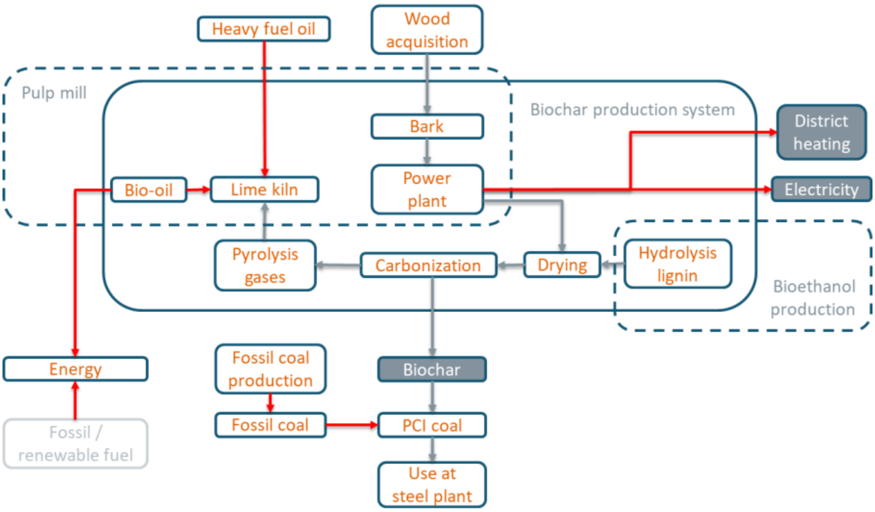


Figure 41. System boundaries for scenario L3. The affected flows are marked with red.

In the Ba reference case, half of the bark is burned in a multi-fuel boiler to produce process steam and electricity, and the other half is gasified to fuel the lime kiln. In the Ba case, all of the bark is pyrolysed, which fulfils the energy demand of the lime kiln in the form of pyrolysis gases and yields a new product – biochar. As a trade-off, less electricity is produced as the bark burned in the multi-fuel boiler is used for biochar production instead, and the energy demand for bark drying increases as all of the bark has to be dried. The system boundaries are illustrated in *Figure 42* and *Figure 43*.

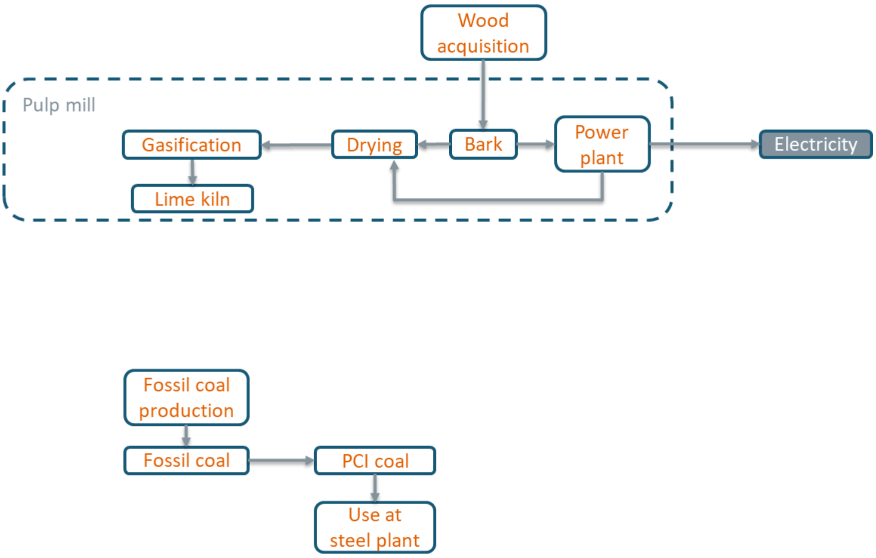


Figure 42. System boundaries for the Ba reference case. The pulp mill operates conventionally, and the excess energy is sold as electricity. The coal used in steel production is fossil coal.

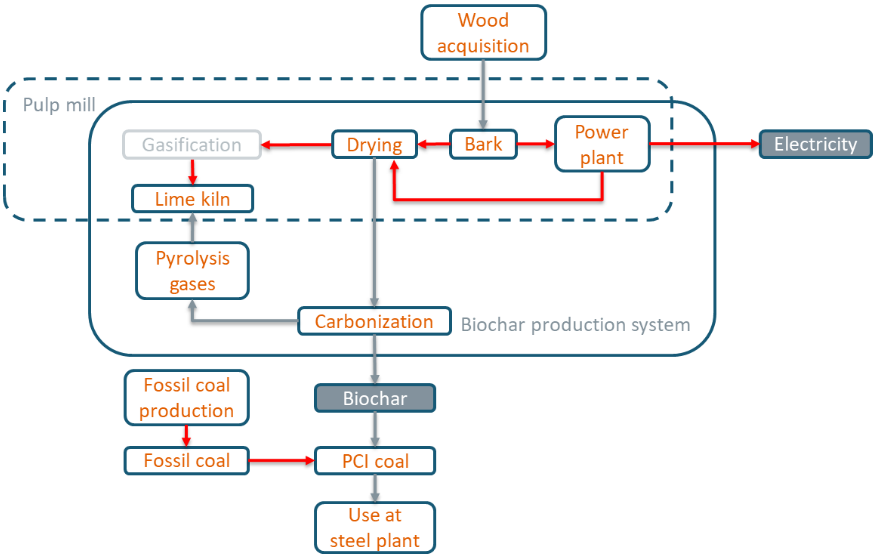


Figure 43. System boundaries for the scenario Ba. The affected flows are marked with red.

The system boundaries include the energy production that is required to replace the reduction in energy from the production site. The effect depends significantly on the replacing energy source, and the results are therefore calculated for two scenarios with different fuel options, using biofuels and fossil fuels as replacements. The same procedure is applied in the case of excess bio-oil, which is assumed to replace another energy source. In L3 and its reference, average Finnish grid electricity is assumed for both cases due to the small change in energy production.

9.1.2 Life cycle inventory

The life cycle inventory data of the considered biochar production scenarios and their reference cases are presented in *Table 33*. Absolute energy amounts are not considered, rather the focus is on the changes. Bio-oil as an output refers to the excess bio-oil that is not burned in the lime kiln but replaces another energy source elsewhere. Bio-oil is a product of the mill integrate, which means that it can be considered as a product, whereas the decrease in heavy fuel oil use means that less external energy needs to be bought. Extra bark to boiler refers to an additional Ba scenario 2, in which more bark is sourced to maintain a similar fuel feed to the multi-fuel boiler as in the reference case, while using bark as the raw material for biochar production.

Biochar replaces fossil PCI coal in steel production. To enable a proper comparison between the two carbon products, the replacement ratio has to be known. This is realized by replacing the fossil coal with the amount of biochar needed to achieve an equivalent heat content. For lignin-based biochar in L3, the replacement ratio is set at 0.94 kg / kg fossil PCI coal. For bark-based biochar in Ba, the ratio is set at 1.05 kg / kg fossil PCI coal (*Table 26*).

Table 33. Life cycle inventory data for scenarios L3 and Ba.

Life cycle inventory data	**L3**		**Ba**		
	Reference	Biochar scenario	Reference	Biochar scenario 1	Biochar scenario 2
Bark [tonnes/a] 40% solids	400 000	400 000	600 000	600 000	600 000
Extra bark to boiler [tonnes/a]					300 000
Bio-oil [tonnes/a]	29 400	29 400			
Excess bio-oil [MJ/a]		644 600			
Heat [MJ/a] (replaced by excess bio-oil)	644 600				
Hydrolysis lignin [tonnes/a] 50% solids		196 000			
Heavy fuel oil [MWh/a]	75 600	30 800			
Biochar [tonnes/a]		44 200		96 000	96 000
Fossil PCI coal [tonnes/a]	47 000		91 400		
District heating [MWh/a]		-49 700			
Electricity [MWh/a]		-2 800		-245 000	-35 600

9.2 Global warming potential results

The global warming potential results are calculated using the climate change impact category defined in the PEF (Product Environmental Footprint) initiative (European Commission 2019). The results are based on annual mass and energy flows.

In the figures below, the result columns are stacked based on different parts of the system in question, which are stated in the legend. *Bark*, *Bio-oil production*, and *Hydrolysis lignin* refer to production of the respective raw materials. *Heavy fuel oil (lime kiln)* refers to the production of the oil and its use in the lime kiln. *District heating* and *Electricity* refer to the impacts from energy production, which is needed to compensate for the reduction in energy production due to biochar production. *Heat* refers to the heat production achieved with the excess bio-oil in the biochar scenarios and with other fuels in the reference cases. *PCI coal production & use* refers to the production of PCI coal and its use at the steel plant.

9.2.1 Scenario L3

The global warming potential results for the lignin scenario L3 are presented in *Figure 44* and *Figure 45*. As the figures show, the overall results for the biochar scenario are clearly superior to the reference system. The main difference results from PCI coal use, as conventionally used fossil coal causes fossil carbon dioxide emissions, whereas biochar is considered carbon neutral.

In *Figure 44*, the results represent the least favourable situation for the L3 scenario. This means that the loss in district heating is compensated by producing the corresponding heat energy from peat, and the excess bio-oil is assumed to replace the heat energy produced from wood chips in the reference case.

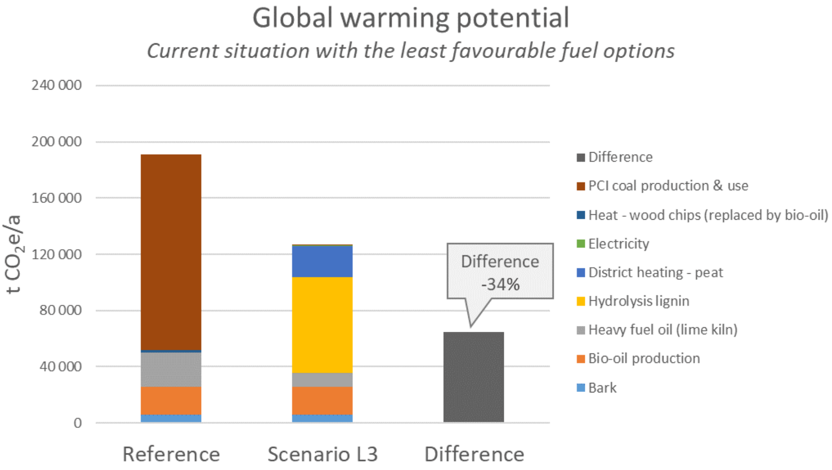


Figure 44. Global warming potential results for scenario L3. Results are calculated with unfavourable fuel options, i.e. the replacement energy sources give the least global warming mitigation benefit to the system. For example, the decrease in district heating is compensated by using peat as an energy source.

Equal amounts of bark and bio-oil are produced and used in both systems resulting in equal impacts. Less heavy fuel oil is used in the lime kiln in L3 than in the reference, which leads to lower impacts. The district heating produced from peat is responsible for almost a fifth of the L3 result.

The main contributor to the impact of L3 is the production of hydrolysis lignin. The high impact is due in part to the large amount used, but also to the lignin production process. The lignin is a by-product of another process where the main impacts are caused by raw material production and partly fossil-based energy. The impacts of lignin and other products of the process are allocated based on their energy contents.

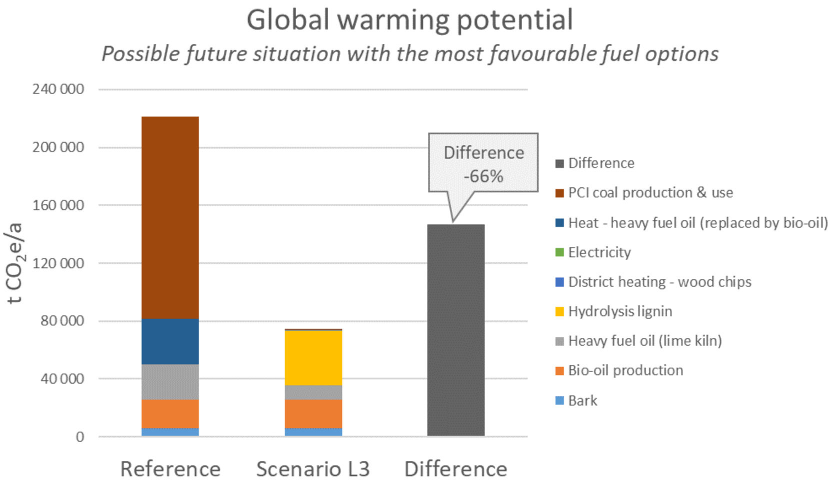


Figure 45. Global warming potential results for scenario L3. Results are calculated with favourable fuel options, i.e. the replacement energy sources give the maximum global warming mitigation benefit to the system. For example, the decrease in district heating is compensated by using wood chips as an energy source.

The results presented in *Figure 45* are similar to the results in *Figure 44*, but the assumptions concerning the replacement fuels are more favourable to the biochar scenario. The excess bio-oil is assumed to replace heavy fuel oil in heat production, and the decrease in district heating production is compensated with wood chips as an energy source. In addition, the impact of hydrolysis lignin is calculated using only bio-based energy for its production, which reduces the environmental burden that the lignin carries. However, the lignin is still the main contributor to the global warming impact of the biochar scenario.

9.2.2 Scenario Ba

The global warming potential results for the bark case (Ba) are presented in *Figure 46* and *Figure 47*. As the figures show, most of the impacts caused in the reference case are due to fossil PCI coal production and use, whereas the impact of the biochar scenario is dependent on the fuel that is used to compensate the loss in electricity generation due to biochar production.

In *Figure 46*, the results are calculated with average Finnish grid electricity (Energy Authority 2019) used to compensate the loss in electricity production. This results in large decrease in the impact due to the low share of fossil fuels in the electricity profile. The difference is 239 000 tonnes of CO_2-equivalent per year.

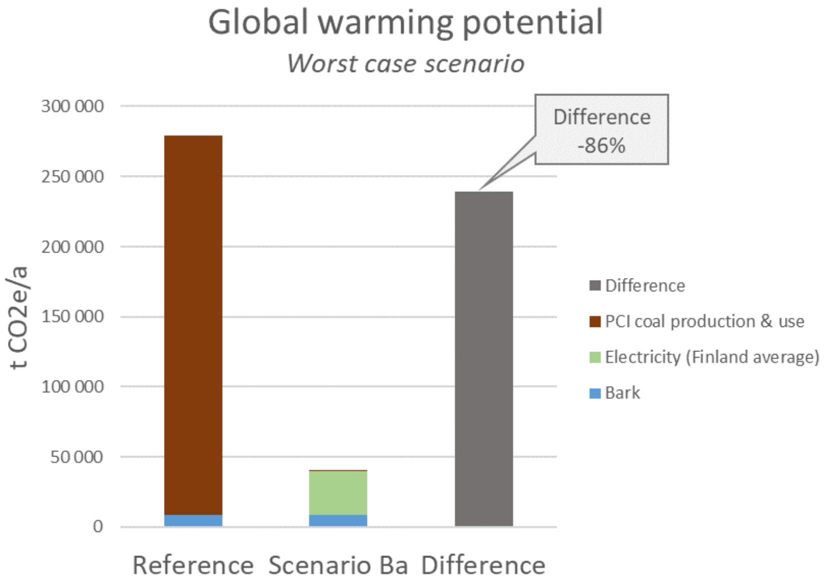


Figure 46. Global warming potential results for the Ba scenario and its reference. Results represent the worst case scenario in which reduced electricity production is compensated with average Finnish grid electricity.

Figure 47 represents a system where decreased electricity production at the production site is mostly avoided by buying more biofuel for combustion in the power plant. The loss in energy production is compensated with biomass burned in a combined heat and power plant. The impact reduction potential of this optimized system is 265 000 tonnes of CO_2-equivalent per year.

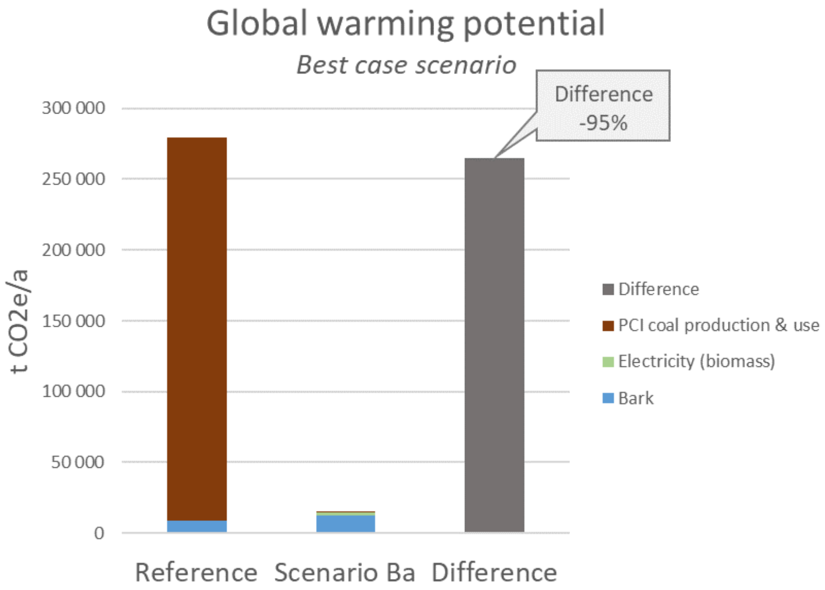


Figure 47. Global warming potential results for the Ba scenario and its reference. Results represent the best case situation where decreased electricity production is mostly avoided by buying more biofuel for the power plant, while the remaining decrease in electricity production is compensated with biomass as an energy source.

9.2.3 Overall results

Based on the assessment results, it can be concluded that the impact on climate change is lower in both scenarios L3 and Ba compared to the reference cases. This is mainly due to the fossil carbon dioxide emissions occurring from fossil coal use in steel production, which can be avoided by using biochar. However, the impact reduction potential depends largely on the whole system that is affected when the biochar production process is implemented.

Especially the energy flows of the overall systems are affected in the scenarios considered. The decreased energy production need to be compensated with another energy source, and the impact can vary largely depending if the replacement energy source is fossil or renewable. In addition, the impact can vary based on the raw material choices. If a raw material carries a large environmental burden, it can affect greatly the overall impact of the system, which can be seen in scenario L3.

10. Future challenges for high-volume supply of biochar

10.1 Biochar potential from forest industry side streams

The present Finnish strategy for forest biomass utilization is targeted at harvesting up to 80 Mm^3 of wood per year, up from the present ca. 70 Mm^3, and enhancing the utilization of side streams and forest residues. The strategy is based on the recent analyses of the Natural Resource Institute Finland (LUKE). Within the forest industry, a part of these side streams is used for necessary utilities to provide steam and power. A significant fraction could, however, be used for other purposes, such as making biocarbon.

Table 34. An estimate of the biocarbon potential of the present usage of lignocellulosic raw material in Finland.

2017	Wood vol. [Mm^3/a]	Side-stream share [vol-%]	Side-stream vol. [Mm3/a]	Share of in-house use [vol-%]	Potential side-stream vol. [Mm3/a]	Basic density [kg/m3]	Yield [wt%]	Biocarbon potential [k-tonnes/a]
Pulpwood bark -pine	17.2	12.5%	2.2	50%	1.1	300	40%	129
Sawtimber bark -pine	11.0	11.0%	1.2	50%	0.6	300	40%	73
Pulpwood bark -spruce	10.6	12.5%	1.3	50%	0.7	365	40%	97
Sawtimber bark -spruce	14.4	11.0%	1.6	50%	0.8	365	40%	116
Wood chips -sawmill	25.4	29.0%	7.4	80%	1.5	430	42%	266
Sawdust -sawmill	28.4	14.0%	4.0	40%	2.4	430	42%	431
Sum					**7.0**			**1111**
Forest chips			6.3	70%	1.9	457	41%	354
SUM					**8.9**			**1465**

Basic density = Dry mass [kg] divided by green solid volume [m^3]

In *Table 34*, an estimate of the biocarbon potential of the present usage of lignocellulosic raw material in Finland is given. The estimated amount of available biocarbon from present-day sources is ca 1.5 million tonnes per year. Logging of softwood (pine and spruce) data is derived from LUKE statistics for 2017 (LUKE 2018). The bark percentage is derived from the respective LUKE report by Lehtonen et al. (2016). In-house use of biorefineries and sawmills is based on industrial experience within the project. Biocarbon yield estimates are derived from experimental work conducted by the University of Oulu on black pellets. Basic densities are obtained from Alakangas et al. (2016).

With a forest biomass increment by growth of ca 4.5% per year in Finland, the potential of lignocellulosic biochar, if used for replacing fossil carbon dioxide sources, would roughly compensate for the loss of such annual growth caused by harvesting. Furthermore, as biochar is a renewable wood product of the forest industry and is used in metal processing as a direct substitute for fossil coal, the production of biocarbon will strengthen the CO_2 balance of both the forest and metal industries (*Figure 48*).

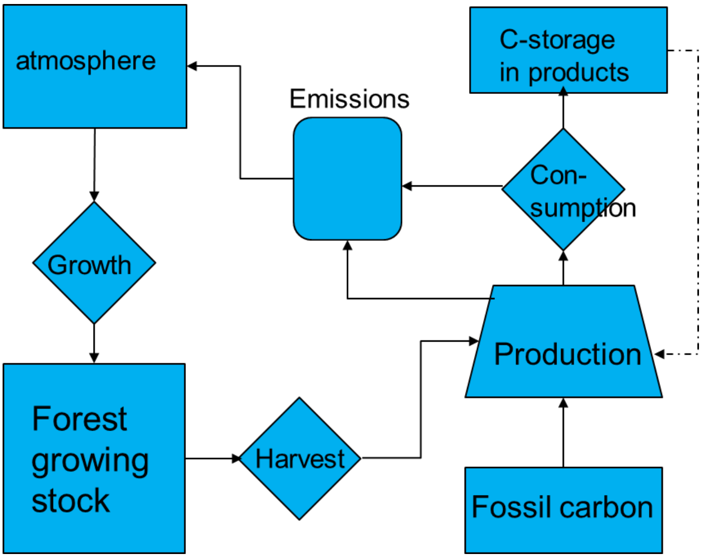


Figure 48. Industrial carbon cycle on forest biomass basis (adapted from Lintunen et al. (2016), CO_2 capture from emissions not included). Biocarbon can be used for permanent C-storage or to replace fossil carbon in production.

10.2 Local availability of side streams

The procurement of sufficient volumes of biomass for use as biocarbon in steelmaking remains a foremost challenge in Finland. Unless organized in a concerted and concentrated manner, as in scenarios lignin (L3), bark (Ba) and black pellets (BP), the gathering of sufficient biomass to the steel mill site will require the integration of harvesting, stevedoring and transportation of forest residues or side stream fractions from within a relatively large rural area. In *Figure 49* this is exemplified by estimating sawn timber from sawmills within a 200 km radius of the Raahe steel mill on the north-west coast of Finland. The side-streams of sawmills, the bark and sawdust, form the potential output of biochar producing.

R< 100 km
490 000 m^3

R< 200 km
2 190 000 m^3
(In Sweden:
1 000 000 m^3)

Figure 49. Sawn timber within a 200 km radius of the Raahe steel mill (Kyytsönen 2016).

Based on the above scenario, the bark and sawdust -based biochar supply potential from sawmills within a 200 km radius of the SSAB Raahe steel mill would be in the range of 15-25% of the total powdered injection coal requirement of the steel mill (Kyytsönen 2016). Considering that the capacity of a single sawmill is typically ca. 200 000 m^3 of timber per year, it would be a subject of a further feasibility analysis to determine whether the bark and sawdust -based biochar production of such mills should be organized within individual mills or through cooperation between several units and the metal manufacturer.

Due to the larger scale of operations, similar challenges to the above would be encountered at pulp mills and biorefineries, but at the individual plants scale. The scenario studies indicate that it would be generally more economical to locate the pyrolysis units within the sites of the biomass refining plants to avoid the transportation of raw (un-dried) chips or bark.

As for hydrolysis lignin, deterioration of the material during transport and intermediate storage is an additional challenge and location of the ethanol production plant in close connection with the pyrolysis process and, for example, pulp mill would provide synergy. The produced biochar and its conveyance to the steel mill would follow the assumptions of the bark (Ba) or black pellets (BP) scenarios, while, for example, the same transport means that are used to bring pulpwood to the biorefinery site would be utilized for rail connections for biochar or black pellets.

10.3 Use of bio-oil as a side product

Biochar can be efficiently produced at locations where sufficient amounts of raw biomass are available. The product, biochar, can be used as a bio-reducer in metal production, as a biofuel to replace fossil-based combustibles, as a soil conditioner, and also in water treatment applications.

The feasibility of production is largely dependent on the utilisation of the gaseous and liquid side products of the pyrolysis process, which account for nearly two thirds of the original biomass feedstock. Solar et al. (2018) have presented an experimental setup and results for the slow pyrolysis of woody biomass waste from forestry activities, with or without thermo-catalytic treatment, for separating the non-condensable and condensable fractions of the pyrolysis gases. The following options may be considered for further treatment of the gases:

- Convert hot syngas to bio-oil and non-condensable gas fractions.
- Non-condensable gases contain H_2, CO, lesser amounts of CO_2, and light hydrocarbons. End use possibilities include replacing fossil-based combustibles, and as an intermediate product for generating hydrogen, alcohols, FT (Fischer-Tropsch) petroleum, FT diesel, olefins, oxo chemicals, ammonia and synthetic natural gas (SNG) (Bain 2004).
- The valuable bio-oil (recovered from condensable gases) can be further processed, e.g. to liquid biofuels to replace fossil-based fuels. The tar-containing fraction of the condensable gases could also be used as a binder for briquetting or pelletizing biochar for easier handling during transport.

In this report it has been assumed that the pyrolysis gases, both condensable and non-condensable fractions, can generally be burned to produce steam, or to replace other fuels in the lime kiln of the pulp mill. It is, however, also possible to separate the condensable fraction and obtain bio-oil, which may be utilized, for example, in oil refinery, depending on the composition, or further processed to a water-containing (acidic) fraction for use, for example, in ash removal, and to a tar-containing (organic) fraction, which may be utilized as a chemical or, for example, as a component of a binder, as explained above. Treatment of the excess condensable vapour will, however, be site and case dependent and various options for its end use remain a topic of further research.

10.4 Options for future work

The production costs presented here are estimates and are based partly on assumptions and calculations made by key experts in the field. Yet, more precise investment estimations are needed from vendors, as well as their views regarding large-scale technical feasibility. Accordingly, the presented figures can, and should be, challenged by future R&D work.

Possible topics of further research are listed below:

- In using pulverized biochar as a replacement for PCI coal, what are the differences in terms of biochar reactivity, ash component effects in the blast furnace, and participation in reduction reactions, when compared to PCI coal?
- Piloting experiments of the slow pyrolysis process with regard to combustion properties and analyses of hot pyrolysis gas (heating values, different chemical components, condensable and non-condensable fractions, applicability as a fuel, and non-process elements in the biochar)
- Technical possibilities for large-scale pyrolysis combined with hot and also non-condensable fraction utilization in the lime kiln

More detailed design solutions and information on implementable large-scale process plants, including solutions, for example, for material handling, drying, and slow pyrolysis process types, as well as investment cost information from equipment and process unit suppliers will be needed.

11. Conclusions

For low carbon steelmaking a variety of technologies has been proposed in which the use of fossil carbon can be avoided or replaced by a renewable reducer material or respective energy source. In Finland the use of biomass based reductants will remain a major option for the existing technologies as well as for possible developments in the foreseeable future.

Substitution of powdered coal injection (PCI) coal in blast furnaces with biochar has the potential to reduce GHG emissions by about 20%. The biocarbon used as a reducing agent in steelmaking will serve as a value-added product for the forest sector while simultaneously reducing the climate impact of steel mills.

Softwood bark, black pellets made of such bark, and hydrolysis lignin were found to be applicable as sources for biocarbon production with a high enough carbon to oxygen ratio, although their calorific heating values, flow and transport characteristics, water uptake, ash content and ash chemical composition varied significantly. Respectively, the varying chemical and physical properties must be taken into account when designing the blast furnace biocoal injection system. The higher alkali and phosphorus content, particularly in bark-based biocarbon, is acceptable if less than 20% of PCI coal is replaced by biocarbon, while higher fractions of bio-based PCI coal from such sources will require further technological assessment.

The techno-economic analysis indicates that biocarbon made of softwood bark at an integrated pulp mill site will provide the best economic option for bio-based carbon production within the forest to metal value chain. Such process will also provide an ample source, at a potential 95 000 tonnes of biocarbon per year. The cost structure analysis coupled with the CO_2 emission trading scheme indicates that with EUA pricing of CO_2 emissions reaching 25 €/tonne, biochar replacement becomes economically viable if the PCI coal price exceeds 125 €/tonne.

A large-scale bioethanol plant integrated to a pulp and paper mill, producing biocarbon, shows also good potential with only ca. 10% higher production cost than bark to biocarbon scenario (above). In case the bioethanol plant is non-integrated and dried lignin raw material is transported to vicinity of a steel mill for biocarbon production the costs are around 40% higher than for the bioethanol plant integrated to pulp and paper mill scenario. These costs may be reduced by heat and/or fuel gas integration with the steel mill.

It was further concluded that transport of raw (undried) biomass to the steel manufacturing site from distributed sources would be an economically challenging alternative, yet potentially viable in north and north-west Finland where there is less competition from the energy sector for biomass sources.

The production of biocarbon from lignocellulosic side streams forms a key pillar of a sustainable industrial policy that will direct the use of available forest resources towards replacing carbon-intensive raw materials in the long term. The use of bio-carbon to replace fossil carbon as a reducing agent in metal production is directly comparable to a carbon sink, as the release of fossil-based carbon dioxide decreases proportionately. The extent of this substitutional effect is yet contingent on technological advancements and will eventually be determined by the changes occurring in the industrial raw material market that, in turn, will depend both on the hierarchy of needs between various biomass uses, as well as on the cost effect of the prices of both biomass and carbon trading.

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APPENDIX. Use of biochar in pyrometallurgical treatment of jarosite waste

As an alternative example of using a bio-reducer in metallurgy, the recycling/immobilization of jarosite waste formed as a by-product of Zinc production was conceptually evaluated.

1. Jarosite formation in zinc production

The majority (85%) of zinc production takes place using the roast-leach-electrowinning (RLE) process, which is also applied at the Boliden Kokkola plant in Finland. Having been adapted since ca. 1970, this approach is also known as the jarosite or goethite process, due to the iron-containing residue that is formed in the process. In *Figure 1*, the outline of the RLE process together with an optional pyrometallurgical residue treatment is schematically presented. The residue also holds some zinc, as well as typically lead, silver and other metals, some of which may bear significant economic value. For every tonne of zinc metal produced using the RLE process 0.5 tonnes of such residue is typically generated.

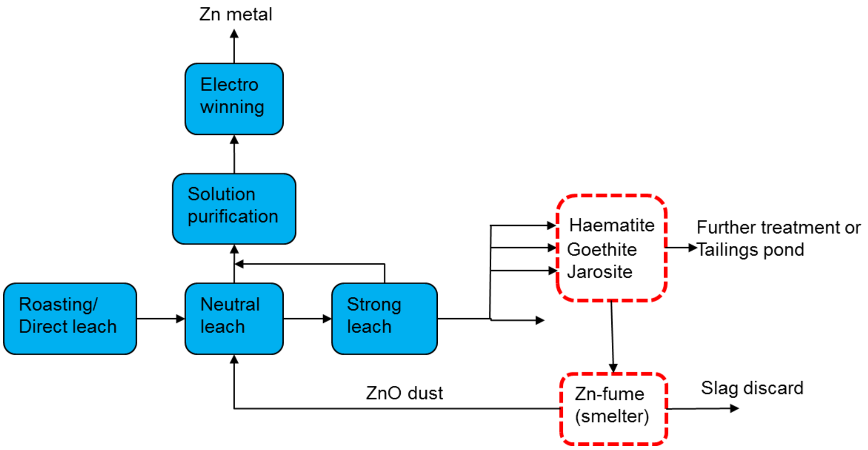
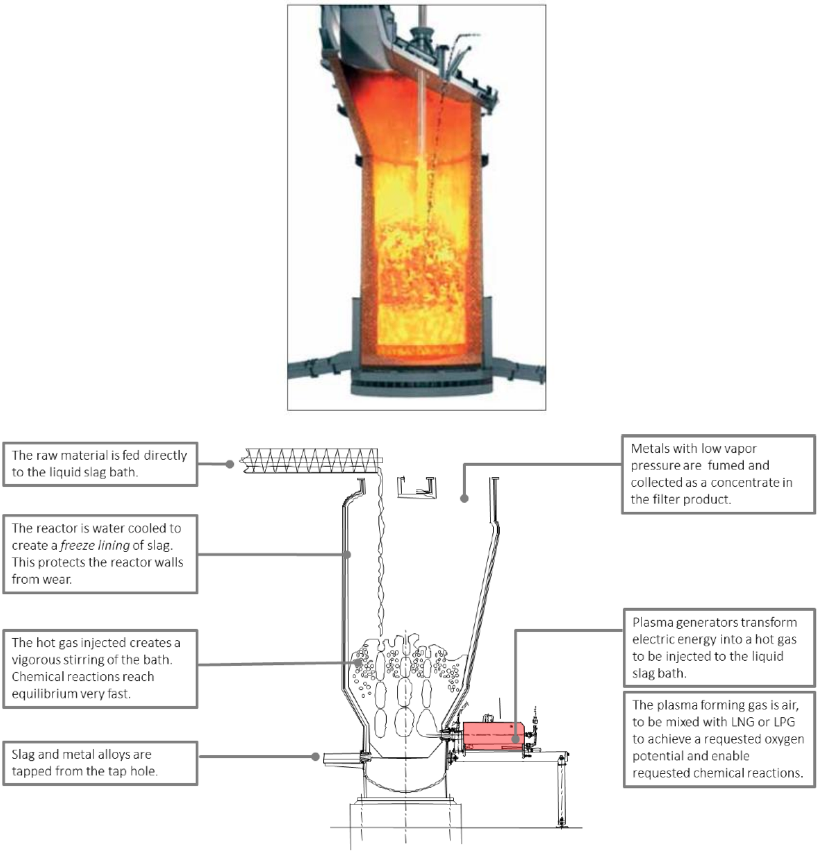


Figure 1. Zinc manufacturing process with pyrometallurgical treatment of jarosite/goethite waste (Hoang et al. 2009).

Traditionally, jarosite has been disposed of in problem waste facilities, or stored in on-site residue areas or tailings ponds. During over 40 years of operation, significant volumes of such metal containing deposits have been stockpiled in the vicinity of RLE facilities. As this practice is being restricted internationally, the zinc manufacturing industry is increasingly looking for options for enhanced waste treatment. The option most often sought to date is pyrometallurgical treatment, which consists of reductive smelting of the jarosite/goethite waste sulphates or oxides to produced zinc as vapour and iron as a component of slag discard (Nyberg 2017). The process then makes use of the volatility of zinc at temperatures exceeding 1300 °C, while the Zn-gas containing fume is then allowed to oxidise to ZnO (dust) which can then be led back to the leaching process. The target of the smelting is also to produce a slag in which harmful trace metals (such as lead or cadmium) are either immobilized or occur at such low levels that no environmental threat is posed. For the typical direct smelting and plasma smelting technologies, which both use coal powder as the reducing agent, it is viable to replace the fossil carbon with biochar.



The raw material is fed directly to the liquid slag bath.

The reactor is water cooled to create a *freeze lining* of slag. This protects the reactor walls from wear.

The hot gas injected creates a vigorous stirring of the bath. Chemical reactions reach equilibrium very fast.

Slag and metal alloys are tapped from the tap hole.

Metals with low vapor pressure are fumed and collected as a concentrate in the filter product.

Plasma generators transform electric energy into a hot gas to be injected to the liquid slag bath.

The plasma forming gas is air, to be mixed with LNG or LPG to achieve a requested oxygen potential and enable requested chemical reactions.

Figure 2. Schematic examples of zinc fuming furnaces. Left: Outotec Ausmelt furnace Right: Submerged Plasma Furnace (Heegaard & Swartling 2017).

2. Use of black pellet biochar in zinc fuming – ChemSheet model results

The smelting reduction of Zn-containing waste using biochar was evaluated using a simple ChemSheet thermodynamic simulation model. The smelting is typically performed in a direct smelting furnace using carbon as the reducing agent and energy source, fluxing the iron-containing oxide slag with a mixture of limestone and silica (Hoang et al. 2009). Carbon consumption has also been reduced by applying electric plasma heating of the smelting furnace (Verscheure et al. 2007). In what follows, carbon consumption in this reference plasma treatment was compared with the respective figures received for typical biochar and for black pellet material. The computational assessment was made using VTT's ChemSheet program using the FactSAGE GTTO database. The model used was validated by testing the model results against the experimental data published by Verscheure et al. (2007) and then replacing the carbonaceous reductant feed with that received from the black pellet analysis. The input amounts of jarosite and flux as specified by Verscheure et al. are given in *Table 1* as well as the figures for BP(500) reductant. These were varied in the calculations according to the respective analysis.

Table 1. Feed composition of jarosite. Only the compounds in bold are included in the calculation.

	[%]	[tonne/h]
Jarosite		**22.14**
$ZnFe_2O_4$	52.96	11.73
$ZnSO_4$	17.91	3.97
$PbSO_4$	9.53	2.11
SiO_2	4.97	1.10
$CaSO_4$	4.63	1.03
Cu_2O	2.19	0.485
Al_2O_3	1.2	0.265
Zn_2SiO_4	1.11	0.246
$MgCO_3$	0.44	0.097
ZnO	0.16	0.03
ZnS	0.09	0.0199
PbO	0.04	0.0088
$CaCO_3$	0.01	0.0022
H_2O	4.76	1.054
Total	**100**	**22.14**

	[%]	[tonne/h]
Flux		**4.99**
$CaCO_3$	54.29	2.71
SiO_2	0.57	0.028
$MgCO_3$	39.77	1.98
Al_2O_3	0.61	0.0304
H_2O	4.76	0.2375
Total	**100**	**4.99**
Reductant [values for BP(500)]		**2.64**
C	78.9	2.091
H	2.7	0.0716
O	6.6	0.1749
N	1.02	0.027
S	0.032	0.00085
SiO_2	2.51	0.0666
$CaCO_3$	6.99	0.185
$MgCO_3$	0.72	0.0191
H_2O	10.4	0.472
Total	**99.5**	**2.64**

Auxiliary fuel		
CH_4	Nm3/h	693.1
Air (total)	Nm3/h	7925.0
Plasma power*	kW	21.6*

**Power is given for the reference case (Verscheure et al., 2007), calculated for all others for comparison (see Figures A3-A4)*

All input streams, as listed in *Table 1*, were assumed to be at 25 °C and 1 bar pressure. The computation was made for 1 400 °C in order to compare the need for auxiliary heat as plasma power for the different reductant feeds. In *Figure 3*, the recovery of Zn as gaseous fume is depicted in all the calculated cases and the results compared with those published by Verscheure et al. (2007) are shown. It may be concluded that the need for additional power (plasma) remains practically unchanged when similar amounts of reductant on a mass basis are used; however, the introduction of biochar lowers the reducing power of the system and affects the

Zn yield. From *Figure 4* it is inferred that the reductant requirement on a mass basis is 10-30% higher if a Zinc yield matching the reference case should be targeted.

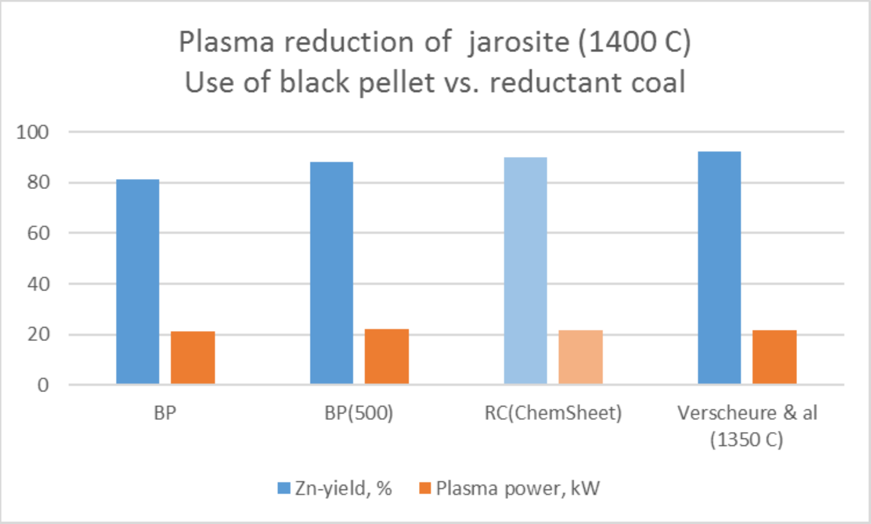


Figure 3. Zn yield and plasma power in jarosite smelting.

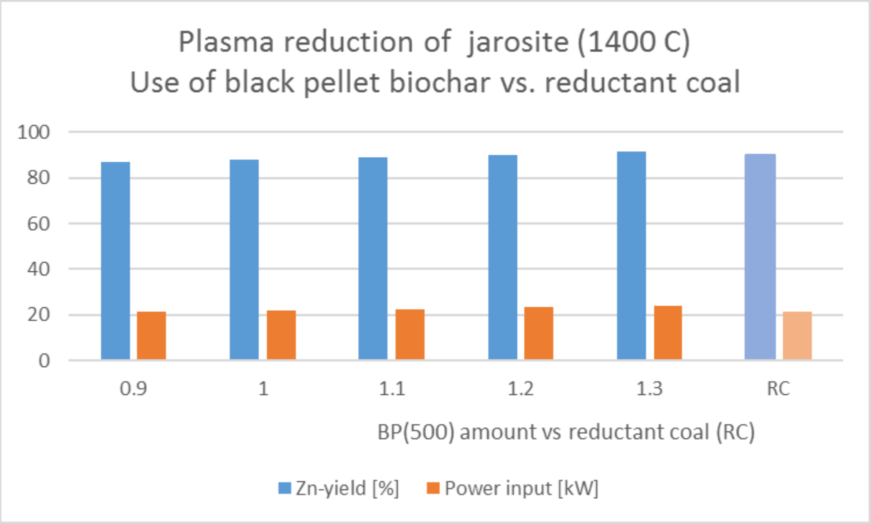


Figure 4. Zn yield and plasma power in jarosite smelting as a function of biochar feed.

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Title	**Replacing Coal Used in Steelmaking with Biocarbon from Forest Industry Side Streams**
Author(s)	Juha Hakala, Petteri Kangas, Karri Penttilä, Matias Alarotu, Martin Björnström & Pertti Koukkari
Abstract	Steelmaking is a highly carbon-intensive process due to its extensive use of coal as both an energy source and a reductant. The CO2 release of Finnish steel production plants exceeds 5 million tonnes/a, accounting for nearly 10% of Finland's CO2 impact. Approximately 80% of these emissions originate from the blast furnace ironmaking process, the production of which currently exceeds 2 million tonnes/a of raw steel. Blast furnaces will remain the major steel production route in Finland for the foreseeable future. In the last 10–15 years a remarkable research and development effort has been focused within the steel industry on improving the performance and energy efficiency of the blast furnace. While significant advances have been made, possibilities to gain further decreases in fuel or reductant consumption are limited due to the high level of maturity and efficiency of the current furnace technology. Thus, new technologies are needed to further reduce greenhouse gas (GHG) emissions associated with steel production. For this, the substitution of fossil carbon with biocarbon has been proposed. The first option is to use biocarbon as a component of the pulverized coal injection (PCI), which takes place through tuyeres in the lower part of the blast furnace. In this research, two kinds of side streams from the forest industry were tested and evaluated as raw materials for PCI biocarbon. Softwood bark from biorefineries (e.g. in the form of black pellets from steam explosion) or residual hydrolysis lignin from sawdust-based bioethanol production were used. Both raw materials were treated with slow pyrolysis and the physical, chemical and rheological properties of the biochar thus produced were compared with those of typical injection coal. Both materials appear to be viable PCI substitutes. Various scenarios for integrating pyrolysis to either the forest industry or steel industry site were examined and techno-economic analysis of the use of biocarbon as a substitute for fossil PCI coal was carried out for each case. It is inferred that there will be sufficient sources of both bark from biorefineries and hydrolysis lignin in Finland in the near future to replace substantial fractions of PCI coal. Use of biochar will decrease GHG emissions; however, the extent is dependent on the operational environment and the power source used to compensate the respective biomass-based electricity and district heat production. The economy of biochar usage as a PCI substitute is becoming more viable with rising CO2 emission costs.
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Tiivistelmä	Teräksen tuotannossa käytetään hiiltä sekä prosessin välttämättömänä lähtöaineena (pelkistimenä raakaraudan sulatuksessa) että energianlähteenä. Maamme terästeollisuuden hiilidioksidipäästöt ylittävät vuosittain viisi miljoonaa tonnia, mikä on lähes 10 % maamme kasvihuonekaasuista. Yli 80 % terästuotannon päästöistä syntyy masuuniprosesseissa, joilla nykyisin valmistetaan Suomessa 2,8 miljoonaa tonnia raakaterästä. Masuunien hiilen kulutus on tehostustoimien ansiosta jo lähellä teoreettista minimiä. Huomio on siten kiinnitetty mahdollisuuksiin korvata nykyisin käytössä oleva fossiilinen hiili biomassapohjaisella raaka-aineella. VTT:n ja Oulun Yliopiston yhteistyönä suoritetussa tutkimuksessa on vv. 2016-2019 selvitetty biomassan saatavuutta metsäteollisuuden sivuvirroista ja näin saadun raaka-aineen soveltuvuutta biohiilen valmistamiseen ja käyttöön fossiilisen hiilen korvaajana masuuneissa. Tulokset osoittavat, että kotimaisesta raaka-aineesta tuotettu biohiili soveltuu injektiohiilenä raakateräksen valmistukseen. Masuunien hiilentarve on n. 1,5 miljoonaa tonnia vuodessa, josta huomattava osa voidaan korvata metsäteollisuuden sivuvirroista pyrolyysitekniikalla valmistettavalla biohiilellä. Biohiilen valmistus luo sivuvirroista metsäsektorille lisäarvotuotteen, jonka hyödyntäminen metallien tuotannossa on hiilineutraalia. Biohiileen perustuvat kiertotalousratkaisut pienentävät siten teollisuuden ilmastokuormitusta sekä vähentävät tarvetta päästöoikeuksien ostamiseen. Tavoitteena on turvata maamme teollisuuden kilpailukyky hiilidioksidipäästöjen aiheuttamien kustannusten kasvaessa. Tutkimuksen yhteydessä on myös suoritettu ensimmäiset onnistuneet tehdaskoeajot. Tutkimusta ovat tukeneet SSAB Europe Oy (Raahe), St1 Oy, Valmet Technologies Oy ja Finnpulp Oy.
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Replacing Coal Used in Steelmaking with Biocarbon from Forest Industry Side Streams

In Finnish blast furnaces over 1.5 million tonnes of coal is used annually both as an energy source and as a necessary reductant to produce over 2.5 million tonnes of raw steel. The respective CO_2 released forms ca. 7 % of the greenhouse gas emissions in Finland. At sawmills and biorefineries, nearly 10 million m^3 of forest industry side streams are formed. A significant part of this could be processed via slow pyrolysis to a biocoal product to replace fossil carbon in steelmaking.

Alternative scenarios for utilizing the side streams of forest industry as sources for steelmaking biocarbon have been assessed. Three particular raw materials studied were hydrolysis lignin side-product from the bioethanol production as well as softwood bark and black pellets from cellulose producing biorefineries. Biocoal from these sources is apt to replace up to 20-30 % of blast furnace powdered coal injection (PCI). An annual reduction of 265 000 tonnes CO_2 equivalents is achieved by one biochar production plant, which would be integrated into a pulp mill.

Making biocoal from the side streams creates an added-value bioreducer product for the forest industry, while its usage provides a carbon neutral solution for steel manufacturing. The economic feasibility will depend on both emission trade cost and integration of the excess energy from pyrolysis and possibly available low grade heat at the biocarbon production site. The cost analysis coupled with the CO_2 emission trading scheme indicates that with European Emission Allowance (EUA) pricing of CO_2 emissions reaching 25 €/tonne, biochar replacement is looking to be an economically beneficial option, if the fossil PCI coal price exceeds 125 €/tonne.

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