Cost-effective Cultivation of Lodgepole Pine for Biorefinery Applications

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Doctoral Thesis
Swedish University of Agricultural Sciences
Umeå 2013

Acta Universitatis agriculturae Sueciae
2013:81

Cover: 29-year-old lodgepole pine stand in Hälsingland, September 2011.
(photo: I. Backlund)

ISSN 1652-6880
ISBN (print version) 978-91-576-7900-0
ISBN (electronic version) 978-91-576-7901-7

Print: Arkitektkopia AB, Umeå 2013

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Abstract

The overall objective was to evaluate the scope for the cost-effective cultivation of lodgepole pine (*Pinus contorta*, *PC*) stands in a way that would enable early biomass harvesting to supply raw material for biorefineries. Commercial direct seeded *PC* stands were shown to produce 200 m^3/ha of stemwood or 100 tons of d.w. biomass within 30 years despite one or two pre-commercial thinnings. Higher stand stem densities ($\geq$3000 st/ha) yielded even more biomass (ca. 300 m^3/ha) with only slight reductions in DBH (Study I). The effects of different silvicultural regimes on 20-year-old direct seeded *PC* stands were analyzed in a field experiment. A high biomass regime (no PCT) produced 144% more biomass and 134% more stem volume than the conventional regime (2200 st/ha). The diameter of the 1000 largest trees/ha did not differ between regimes. A regime with 4500 st/ha gave promising results in terms of both biomass and timber production. Importantly, producing large amounts of biomass early in the rotation period is compatible with a subsequent change of focus to emphasize pulp and timber production (II). To investigate the potential for using *PC* biomass in biorefineries to produce e.g. liquid biofuels, the chemical contents of wood samples from Scots- and lodgepole pine trials were compared. Heartwood had up to five times greater extractive contents than sapwood. 21 fatty and 10 resin acids were detected. It was estimated that ca. 150 kg of fatty acids and 1 ton resins/ha could be harvested from a mature boreal *PC* stand (III). The chemical compounds in the aboveground fractions of *PC* trees grown under a direct seeding-based regime were identified. The bark provided the highest extractive yields (16%) and the stemwood the lowest (1%). The extractive profiles of the needles differed strongly from the other fractions, being particularly rich in wax esters and fatty alcohols. It should be possible to harvest 2-3 tons of crude extractives/ha from a dense 30-year-old *PC* stand (IV). To estimate the commercial potential of different biorefinery products, a survey was performed. 95% of the respondents believed that the value of tree biomass will increase over the next ten years, mainly due to the replacement of oil-based products. Key product categories were: transportation fuels, special celluloses, materials and plastics, solid fuels and specialty chemicals. A strong correlation between the prices of electricity and wood fuel was identified, and electricity prices may play a key role in determining the future use of biomass (V).

Overall, there is considerable but currently unrealized potential for the cost-effective cultivation of lodgepole pine in directly seeded dense stands using short rotation periods to produce substantial quantities of biomass for biorefineries within only a few decades.

Keywords: biomass production, goal-oriented forestry, biobased products, Fennoscandia, chemical extraction, direct seeding, short rotation.

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Kostnadseffektiv odling av contortatall för användning i bioraffinaderier

Sammanfattning

Det övergripande målet med arbetet har varit att utvärdera möjligheten till kostnadseffektiv odling av contortatall *(Pinus contorta)* på ett sätt som skulle möjliggöra ett tidigt biomassuttag till bioraffinaderier. Kommersiellt sådda contortabestånd visade sig producera 200 m^3 stamved per hektar, eller ca 100 ton biomassa (torrsubstans) inom 30 år trots att en eller två röjningar genomförts. Högre stamantal (≥3000 st/ha) gav ännu mera biomassa (ca 300 m^3/ha) med endast en smärre reduktion i stamdiameter (Studie I). Effekterna av olika skogsskötselregimer analyserades i ett fältexperiment i ett 20-årigt sått contortabestånd. En regim för hög biomassaproduktion (ingen röjning) producerade 144% mer biomassa och 134% mer stamvolym än en konventionell regim (2200 st/ha). Diametern för de 1000 största träden per hektar skiljde sig inte åt mellan skötselregimerna. En regim med 4500 st/ha visade lovande resultat vad gäller både biomassa- och timmerproduktion. Tidig hög produktion av biomassa verkar vara förenlig med en senare omställning till produktion av massaved och timmer (II). För att undersöka potentialen att använda biomassa från tall i bioraffinaderier för att t.ex. producera fordonsbränslen, analyserades det kemiska innehållet i vedprover från både tall och contorta. 21 fettsyror och 10 hartssyror detekterades. Kärnveden innehöll upp till fem gånger mer extraktivämnen än splintveden. Uppskattningsvis 150 kg fettsyror och 1 ton hartssyror kan utvinnas per hektar från ett äldre borealt contortabestånd (III). I den fjärde studien identifierades kemiska föreningar i alla trädfraktioner ovan jord (stamved, bark, grenar, barr och kottar) från direktsådd contortatall. Barken visade sig innehålla högst halter extraktivämnen (16% av torrvikten) medan stamveden gav de lägsta halterna (1%). Barrens kemiska innehåll skiljde sig starkt från de andra fraktionerna, då de var särskilt rika på vaxestrar och fettsyraalkoholer. Utifrån dessa data borde det vara möjligt att utvinna 2-3 ton extraktivämnen per hektar från täta 30-åriga contortabestånd (IV). Slutligen utfördes en enkät för att bedöma den kommersiella potentialen för olika skogsbaserade bioraffinaderiprodukter. 95% av de tillfrågade i undersökningen tror att värdet på skogsbiomassa kommer att öka de kommande 10 åren, mestadels som ersättare av oljebaserade produkter. Viktiga produktgrupper inkluderar fordonsbränslen, specialcellulosa, plaster och andra biomaterial, fasta biobränslen samt specialkemikalier. Elpriset kan komma att spela en viktig roll för den framtida användningen av biomassa.

Slutsatsen av avhandlingsarbetet är att det finns en stor outnyttjad potential att odla contortatall i täta sådda bestånd i kortare rotationsperioder, för att kostnadseffektivt producera stora mängder biomassa med lämpliga kemiska egenskaper till bioraffinaderier.

Nyckelord: biomassaproduktion, målinriktat skogsbruk, biobaserade produkter, Fennoskandien, kemisk extraktion, direktsådd, kort omloppstid.

Dedication

To my beloved family, and to generations of forest workers who have sustained hard work and harsh winters to maintain Swedish forestry.

Jag vill hem till dalen vid Pajso,
till det gräsiga kärret vid So,
där skogarna murgrönsmörka
stå i ring kring mossig mo,
där starrgräs i ånga växer
vid källor som aldrig sina
och där växter väva i jorden
sina rötter silkesfina.
Dan Andersson

Contents

List of Publications

This thesis is based on the work contained in the following papers, which are referred to in the text using the Roman numerals shown below:

I Backlund, I. and Bergsten, U. (2012). Biomass production of dense direct-seeded lodgepole pine (*Pinus contorta*) at short rotation periods. *Silva Fennica* 46(4), 609–623.

II Ulvcrona, K.A., Karlsson, L., Backlund, I. and Bergsten, U. (2013). Comparison of silvicultural regimes of lodgepole pine (*Pinus contorta*) in Sweden 5 years after precommercial thinning. *Silva Fennica* 47(3), id 974.

III Arshadi, M., Backlund, I., Geladi, P. and Bergsten, U. (2013). Comparison of fatty and resin acid composition in boreal lodgepole pine and Scots pine for biorefinery applications. *Industrial Crops and Products* 49, 535–541.

IV Backlund, I., Arshadi, M., Hunt, A.J., McElroy, C.R., Attard, T.M. and Bergsten, U. Extractive profiles of different lodgepole pine (*Pinus contorta*) fractions grown under a direct seeding-based silvicultural regime. *(Manuscript).*

V Backlund, I., Karlsson, L., Mattsson, L. and Bergsten, U. Biorefinery product potentials using tree biomass - Effects of tree assortments and electricity prices. *(Submitted manuscript).*

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Ingegerd Backlund formulated the aims and issues addressed in all of these articles and manuscripts, in collaboration with the listed supervisors and co-authors.

The author's contributions to these papers were as follows:

I The author was primarily responsible for selecting the lodgepole pine stands, forest inventories and measurements, conducting the data analyses, and writing the paper.

II The author contributed to the forest inventories and measurements, performed data analyses, the making of tables and figures and writing the paper.

III The author conducted the variance analyses, participated in the production of tables and figures, and wrote the paper together with the co-authors.

IV The author was primarily responsible for planning the study and selecting the sampled trees, tree sampling, performing chemical analyses, conducting variance analyses and writing the paper.

V The author planned and executed the questionnaire study together with Lars Karlsson, analysed the responses, and wrote the paper in collaboration with the co-authors.

1 Introduction

1.1 A challenge and an opportunity

Oil is a limited resource that can only be extracted economically on a large scale in a few countries. Moreover, its use as a fuel and a chemical feedstock over the course of the last century has caused the release of large quantities of carbon dioxide into the atmosphere. This is one of the main reasons for the climate change that the world is currently experiencing (Malcolm *et al*., 2001; European Commission, 2008; Allen *et al*., 2010; Schoene & Bernier, 2012). Global warming causes floods, droughts, storms and other weather-related natural catastrophes. These phenomena in turn cause losses of agricultural land, conflicts over resources, diseases, migrations and losses of various species, and reductions in habitat diversity. The transport sector accounts for the bulk of all oil consumption in many countries (including Sweden), followed by heating and then industrial use (McCormick *et al*., 2006).

The development of economical methods for the large-scale production of fuels, chemicals and materials from lignocellulosic non-food biomass rather than oil is likely to represent an essential step towards the establishment of a sustainable society, for two reasons. First, it will enable the replacement of CO_2-releasing fossil fuels with carbon-neutral biomass-derived alternatives. Second, increasing the amount of biomass that is grown will increase the rate of carbon sequestration from the atmosphere; carbon is sequestered in growing trees and in long-lived wood products such as timber-framed houses and furniture (Hynynen *et al*., 2005). In addition to the urgent need to reduce CO_2 emissions and increase carbon sequestration, it is also important to reduce the quantity of resources consumed by modern societies and to find sustainable alternatives to widely used non-renewable raw materials and energy sources (Clark & Deswarte, 2008). Forest biomass is a very abundant raw material that could potentially replace a number of oil products, especially in countries that have extensive forest cover such as Sweden and Finland. The production of lignocellulosic biomass on marginal non-food producing land can also contribute significantly to the social and economic development of rural communities (Charlton *et al.*, 2009).

The demand for bio-based fuels and chemicals has been stimulated by lobbying, the strong value proposition offered by certain green technologies, and public policy measures that increase the cost of fossil fuels relative to biofuels (Wright, 2006; Söderholm & Lundmark, 2009; Ulmanen *et al.*, 2009; Wiesenthal *et al.*, 2009; Collantes, 2010). In 2008, the EU introduced the 20-20-20 targets, which call for a 20% reduction in EU-wide greenhouse gas emissions and for sustainable sources to account for at least 20% of Europe’s energy requirements by 2020 (European Parliament, 2009). In the US, the Department of Energy has collaborated with the Department of Agriculture and the American paper industry to invest several hundred million dollars into biorefinery projects whose main aim is to develop alternative fuels (Collantes, 2010). The growing demand for biomass from biorefineries and bioenergy facilities may be impossible to meet with current supply levels (Conrad *et al.*, 2011; Näyhä & Pesonen, 2012).

Large quantities of biomass will be required to compensate for the expected reductions in the availability of energy derived from oil, coal, natural gas, and nuclear power, which are driven by environmental and security concerns. Germany has made pioneering advances in promoting the use of biomass, notably through the 'Energy Transformation' ('Energiewende') project that was introduced in 2011. This ambitious initiative has several key goals: to eliminate all nuclear power generation in Germany within 10 years and to fully replace the country’s nuclear capability with renewable energy resources; to achieve a 40% reduction in greenhouse gas emissions by 2020 and an 80% reduction by 2050 while ensuring that renewables supply 80% of Germany's energy requirements by 2050; and to achieve a 20% reduction in the country’s energy consumption by 2020 and a 50% reduction by 2050 (Agora Energiewende, 2013).

Forests have long been managed for different purposes, mainly the production of massive wood products, pulp and fibres. Forest managers can manipulate the growth of the trees to achieve their desired outcomes in many ways. These include selecting the seeds or seedlings that get planted, choosing the soil preparation techniques that are applied and using specific planting methods, performing pre-commercial and commercial thinnings to achieve desirable stand stem densities, applying fertilizer and disease control measures, and selecting the method of harvesting. The production of biomass from tree species such as the lodgepole pine for bioenergy and biorefineries has not previously been prioritized in forest management, but it is becoming increasingly important due to the gradual shift away from fossil fuels to renewable feedstocks. This presents both a large challenge and an important opportunity for foresters to expand their product ranges and increase their incomes, for the chemical industry to increase its usage of sustainable raw materials, and for increased cooperation between the two sectors.

1.2 Lodgepole pine

In this work, "lodgepole pine" refers to the latifolia subspecies (*Pinus contorta var. latifolia),* which grows in western North America along the Rocky Mountains, from northern New Mexico in the USA to the Yukon Territory in Canada (Hagner, 1983; Despain 2001). *Latifolia* is the northern inland form of the species and was introduced on a large scale in Sweden during the mid-20th century (Elfving *et al*., 2001). It specializes in establishing itself rapidly in recently burned woodlands: the seeds of older trees are enclosed in serotinous cones that require a relatively high temperature to open (Engelmark *et al*., 2001). These cones can persist in the environment for up to 40 years until a fire provides the high temperatures required for their opening. This enables dense lodgepole pine stands to grow rapidly in the aftermath of a fire (Hagner, 1983; Despain, 2001). The trees exhibit fast initial growth, fast root development and high initial survival rates as long as the site in which they are growing is not too shady (Norgren & Elfving, 1994; Coates, 2000; Elfving *et al*., 2001). This reduces the duration of the period during which the tree is vulnerable due to its small size. Compared to other boreal conifers, lodgepole pine is less sensitive to competition from other plants and poor habitat conditions during its juvenile phase (Dermer, 2007).

Lodgepole pine produces approximately 36% more stem volume than Scots pine grown under the same conditions in northern Sweden (Elfving *et al*., 2001). This is due to a number of factors including an earlier start of growth in spring and a lower required heat sum for the initiation of shoot elongation (Elfving *et al*., 2001; Fedorkov, 2010). In southern Sweden, lodgepole pine has fewer advantages (Liziniewicz *et al*., 2012) because the soils are generally more fertile and the climate is milder. In traditional planting-based forest regimes, the optimum rotation length for lodgepole pine is 10-15 years shorter than that for Scots pine (Elfving *et al*., 2001). The leaf area index (LAI) and stem increment values for lodgepole pines peak at 40-45 years of age, following canopy closure (Long & Smith, 1992).

The needles of the lodgepole pine are longer and heavier than those of Scots pine (Norgren & Elfving, 1994). Moreover, they have a greater surface area (and therefore absorb more light) and a lower nitrogen content, giving them a greater productivity per unit of nitrogen compared to Scots pine needles (ibid.). Together, these factors mean that lodgepole pine accumulates biomass more rapidly than Scots pine. Even though a stand of lodgepole pine generates more stem biomass than one of Scots pine in absolute terms, the stems account for a smaller proportion of the total biomass in planted lodgepole pine than in Scots pine (Norgren, 1996). That is to say, branches and needles account for a greater proportion of the total biomass in lodgepole pine. Lodgepole pine is therefore an attractive species for short rotation, whole-tree biomass production in Fennoscandia.

1.2.1 Lodgepole pine in Sweden

Lodgepole pine was introduced in Sweden via a series of small plantations that were established in the 1920s (Elfving *et al.*, 2001). Due to its rapid growth, hardiness, and ability to grow in many different climates and on many types of soil, its performance in Sweden was examined more extensively in the 1960s (Hagner, 1983). At the time, a lack of spruce- and pine timber was expected by the beginning of the 21st century, and lodgepole pine was considered to have the potential to fill this gap, especially as a source of pulpwood. It was found that seeds from northern British Columbia and the Yukon, the northern boundaries of the species' natural range in North America, were most tolerant of the Swedish climate (ibid.). Trees from these regions exhibit the greatest levels of growth during the early parts of the summer and tend to be better prepared for the early onset of winter conditions, particularly in terms of their ability to survive damage caused by climatic factors and pathogenic fungi (Hagner, 2005).

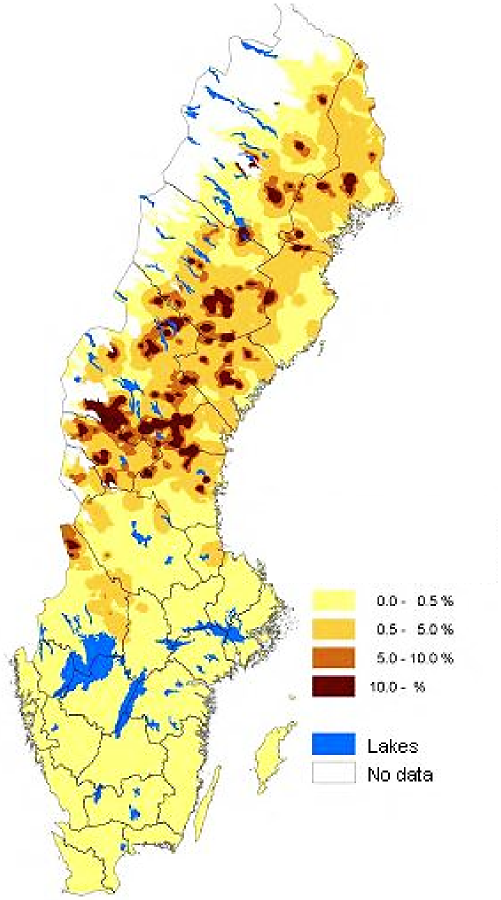

Figure 1. The distribution of lodgepole pine as a proportion of the total productive woodland in Sweden (Swedish National Forest Inventory, 2009).

In northern Sweden, Scots pine benefits from being grown at more southerly latitudes compared to where the seeds were collected. Conversely, lodgepole pine benefits from a northerly transfer of 2-5^{o} compared to the latitudes at which it is found in Canada. This is because the climate in these more northerly parts of Sweden is very similar to that in the slightly more southerly parts of British Columbia (Hagner & Fahlroth, 1974; Elfving *et al.*, 2001). Consequently, sites planted with lodgepole pine in northern Sweden tend to have higher latitudes but lower elevations than the sites from which the seeds were collected in Canada. Lodgepole pine was planted quite extensively between the end of the 1970s and the late 1980s (Hagner, 2005). The level of planting peaked in 1984-85, when almost 40 000 hectares were converted into lodgepole pine plantations annually in Sweden (Swedish Forest Agency, 2012).

In 1988 the planting of the species decreased dramatically because of problems with unstable paper pot-seedlings and the fungus *Gremmeniella abietina* (Elfving *et al.*, 2001; Hagner, 2005).

In recent years there has been renewed interest in lodgepole pine due to its high biomass production and the potential it offers for inexpensively regenerating stable stands by direct seeding. The total area of lodgepole pine in Sweden amounts to ca. 600 000 ha, corresponding to around 2% of the country's total woodlands or 30.2 million cubic meters of wood (Swedish Forest Agency, 2012). In 2011, 7100 hectares of Swedish land were regenerated with lodgepole pine (ibid.).

However, lodgepole pine is an exotic species in Europe and therefore may introduce new diseases or cause landscape fragmentation (Karlman, 2001; Knight *et al.*, 2001). Consequently, Sweden's forestry guidelines state that it should mainly be used where domestic species do not regenerate satisfactorily (Skogsvårdslag 1979:429) and that it should be monitored in order to detect and prevent potential adverse ecological effects (Engelmark *et al.*, 2001). Because of these restrictions, lodgepole pine is mainly planted in the northern part of the country at latitudes above 60^{o}N (Fig. 1). Western Sweden is an exception in that lodgepole pine can be planted at latitudes above 50^{o}30'N in this region (Skogsvårdslag 1979:429). The Swedish Forest Agency has further stated that no more than 14 000 ha of lodgepole pine should be regenerated annually. Changes to these rules that would enable lodgepole pine to be planted more extensively are currently being considered (Swedish Forest Agency, 2009).

Many studies have been conducted to determine the effects of lodgepole pine on Swedish ecosystems (Swedish Forest Agency, 1992; Andersson *et al.*, 1999; Engelmark *et al.*, 2001; Karlman, 2001; Knight *et al.*, 2001; Sjöberg & Danell, 2001; Nilsson *et al.*, 2008). The species has a wide canopy that causes a greater shading effect than Scots pine, and produces more needle litter. Together, these factors mean that lodgepole pine stands have a more homogeneous understory flora (Nilsson *et al.*, 2008). However, it seems that lodgepole pine ecosystems can accommodate co-management regimes that target the production of both timber and NTFPs (non-timber forest products) such as herb and shrub species, e.g. berries (Clason *et al.*, 2008). An Environmental Impact Analysis (EIA) of lodgepole pine forestry in Sweden concluded that a balanced lodgepole pine usage pattern in which the species is grown on land selected with due consideration of social and biological factors should not have negative effects on the country's biodiversity (Andersson *et al.*, 1999).

Northern Sweden is also an important location for reindeer husbandry, and the different sectors that use the forest need to cooperate with one-another in order to avoid causing excessive harm (Sandström & Widmark, 2007; Roturier, 2009; Kivinen *et al.*, 2010). Direct-seeded lodgepole pine forestry with biomass outtakes can have both positive and negative implications for reindeer husbandry compared to traditional lodgepole pine or Scots pine forestry. Direct seeding with gentle soil preparation may affect the ground less than traditional scarification and planting. Lichens provide up to 80% of all reindeer forage in winter (Berg *et al.*, 2008), so it is important to minimize the impact of forestry on lichen-rich land. However, lodgepole pine produces long and robust branches, especially when grown in sparse stands, and dense lodgepole pine stands may be difficult for reindeer to travel through while also harboring a less varied undergrowth (Swedish Sami Association, 2011). Whole-tree biomass harvesting (e.g. by corridor thinning) might produce stands that are easier for reindeer to move through than those created by traditional pre-commercial thinning or thinning because the corridors are regularly spaced and located in close proximity to one-another within the stand, and comparatively few tree residues are left behind after the harvest.

1.3 Lodgepole pine as a biomaterial

1.3.1 Physical properties

Lodgepole pine was primarily introduced in Sweden as a pulpwood species, but in a few years larger volumes of timber will also be accessible. Compared to Scots pine of the same age grown under the same silvicultural regime, the wood of the lodgepole pine is slightly less dense with longer fibres and a greater proportion of heartwood (Ståhl & Persson, 1988; Persson, 1993). Moreover, because the lodgepole pine grows more rapidly, it will have a greater stem diameter at any given age than an equivalent Scots pine. Lodgepole pine also has a thinner bark and more ductile branches than Scots pine (Swedish Forest Agency, 1992).

According to Sable *et al.* (2012) lodgepole pine gives slightly higher pulp yields than Scots pine, and provides pulp handsheets with higher burst strengths. Small diameter lodgepole pine timber is also suitable for the production of structural composite lumber (SCL) materials such as steampressed scrim lumber (SPSL) composites. Lodgepole pine SPSL has high modulus of elasticity (MOE) but low modulus of rupture (MOR) values (Linton *et al.*, 2010).

The Swedish forest company SCA holds large areas of land that have been regenerated with lodgepole pine since the 1970s and onwards. In recent years, they have explored the viability of sawing lodgepole pine for timber production and have tested various surface treatments and methods of drying the wood (Andersson, 2013). Most of these experiments were conducted using the most abundant type of lodgepole pine timber produced in Sweden, i.e. small dimension timber from commercial thinnings with a high content of juvenile wood. It was concluded that lodgepole pine timber is most likely to be useful for panels and simpler construction timbers because it is weaker than Scots pine timber. The current volume of lodgepole pine timber produced in Sweden is insufficient for large scale commercial use. However, in ca. 10 years' time, commercial thinnings will provide sufficient volume to support sawing on a larger scale.

The wood fibres from the living crown of the tree tend to be very short and the density of the wood is low. Wood of this sort is known as juvenile wood (Briggs & Smith, 1986). As the trees grow, the juvenile wood gets progressively more distant from the green crown and is transformed, first into transition wood and then into stronger mature wood. On average, lodgepole pines only begin producing mature wood after reaching about 30 years of age (Mansfield *et al.*, 2007). However, trees that are subject to significant competition in dense stands will start producing it at an earlier stage because such conditions favor the loss of lower branches and crown lift (ibid.). Juvenile wood contains more lignin and extractives than mature wood and may therefore be more useful as a raw material for biorefineries (Hatton & Hunt, 1993). Because of its shorter fibre lengths, paper made from juvenile wood is smoother but weaker than that made from mature wood (Hatton, 1997). It would be possible to produce wood pulps with tuned properties for specific purposes by using blends of juvenile and mature wood in appropriate proportions, but that would require additional sorting of the pulpwood. If the aim is to maximize biomass production in lodgepole pine stands, they should be managed in a way that emphasizes stem volume growth rather than wood density, which is important when producing saw timber (Wang *et al.*, 1999). At present, lodgepole pine is mainly grown as a pulpwood species. However, it could also be cultivated to produce a combination of biomass and saw timber.

1.3.2 Chemical properties

Wood consists of cellulose, hemicelluloses, lignin, extractives and ashes (inorganic substances). Extractives such as fatty and resin acids, waxes, sterols, terpenes and other phenolic compounds are nonstructural constituents of wood (Hillis, 1987; Ekeberg *et al.*, 2006). They are most abundant in the external heartwood and in damaged parts of the tree because they confer resistance to insects, fungi and rot. Hardwoods contain more extractives than softwoods, and pines contain more extractives than spruces (Heinze & Liebert, 2001).

Lodgepole pine is generally believed to be richer in extractives than Scots pines (Sjöström, 1993; Koch, 1996) but some contradictory results have been presented (Sable *et al.*, 2012). Lodgepole pine is appreciably richer in condensed tannins and total phenolics than Scots pine (Stolter *et al.*, 2009). In particular, its bark is very rich in tannins (Mila & Scalbert, 1995). Ruminants often avoid plants that are rich in tannins (Foley & Moore, 2005), which may explain why the species is less heavily browsed by moose than is Scots pine. Lodgepole pine also has lower nitrogen content than Scots pine and higher content of lignin (Stolter *et al.*, 2009). In addition, it has high concentrations of flavonoids, waxes, fatty acids, resin acids, phytosterol, terpenes and antibacterial stilbenes (Hergert, 1956; Rowe & Scroggins, 1964; Hadley & Smith, 1989; Willför *et al.*, 2003; Välimaa *et al.*, 2007).

The heartwood forms the innermost part of the stem. It can be distinguished from the outer sapwood by its darker colour (for an illustration, see Fig. 8). Compared to sapwood, heartwood is more resistant to decay and less biologically active. The heartwood of pines normally contains more extractives than the sapwood (Hillis, 1972; Campbell *et al.*, 1990; Uusitalo, 2004; Eriksson *et al.*, 2012). In lodgepole pines, mature heartwood starts to form at the base of the stem at an age of 20-21 years, and expands by two-thirds of a year ring annually. According to “the heartwood age square root law”, the best predictor of heartwood content is the cambium age (Gjerdrum, 2003). The highest concentrations of extractives are therefore found at stump level and decrease progressively with height up to a certain point (which occurs around 30% of the way up the stem). Beyond this point the extractive concentration does not change significantly with height (Koch, 1996; Eriksson *et al.*, 2012).

1.4 The uses of tree biomass

1.4.1 Traditional products and assortments

In modern Swedish forestry, pine and spruce stemwood is harvested and divided into timber, pulpwood and fuelwood based on its dimensions and quality. The European pulp and paper market is quite unstable and is experiencing growing competition from other continents. European pine timber is also subject to competition from other tree species and continents, but more and more people are discovering the advantages of wood as a construction material and are using it to produce things such as environmentally-friendly houses (CO_2 sinks). Multi-storey wood houses are becoming more common as a “greener” alternative to concrete buildings. Bioenergy assortments including densified wood fuels (pellets and briquettes), wood chips and various logging residues and industrial byproducts are used in the generation of heat and electricity.

The extraction of wood residues and the use of solid biofuels have increased significantly in recent years (Ericsson & Nilsson, 2004). The demand for biomass-derived thermal fuel is increasing steadily around the world, especially in Asia and Latin America (Wright, 2006), but also in regions like the southern U.S. (Conrad *et al.*, 2010). In Sweden, the extraction of wood residues for the production of bioenergy has become a significant part of the economy (Eriksson & Nilsson, 2004). According to the Swedish Bioenergy Association, Svebio, bioenergy is the largest energy source (meaning all forms of energy, not just electricity) in Sweden, accounting for 130.8 TWh or 32.4% of the total domestic energy consumption in 2012 (403 TWh). For comparative purposes, oil accounts for 108 TWh (26.7%), hydro power for 71 TWh (17.6%), and nuclear power for 55.9 TWh or 13.8% of the total (Swedish Bioenergy Association, 2013). In addition, bioenergy accounts for 11% (16 TWh) of Sweden’s total electricity production, which was 150 TWh on average between 2011 and 2013 (Swedish Energy Trade Association, 2013). Bioenergy is therefore responsible for a large proportion of Sweden’s heat production, notably as a fuel for district heating, and has replaced oil in heating- and cogeneration plants to a large extent. Hydro power and nuclear power are the country’s main electricity sources and oil remains the main energy source for transportation.

1.4.2 The biorefinery concept and related products

A biorefinery integrates various biomass conversion processes to produce fuels, chemicals, materials, heat and power from biomass (Demirbas, 2009). Essentially, a biorefinery adapts concepts and techniques for petroleum refining and uses them to convert biomass into valuable chemicals. The whole tree and its various constituents (cellulose, hemicelluloses, lignin, and extractives) can be used to create a wide range of different products. The suitability of a given tree or assortment of wood for this purpose is more dependent on the chemical properties of the biomass than the mechanical properties of the wood. Ideally, biomass for biorefineries would be regarded as a distinct assortment that could complement the existing timber, pulpwood and fuelwood (bioenergy) assortments. Tree biomass is suitable both for producing heat and electricity and for the manufacture of biorefinery products such as chemicals, fuels, plastics and fibres. Extractives are known to cause problems in pulp- and papermaking (Farrell *et al.*, 1997; Sun & Tomkinson, 2001) as well as in pellet production, storage and transportation (Arshadi & Gref, 2005; Arshadi *et al.*, 2009) even though they increase the wood's energy content (Filbakk *et al.*, 2011; Eriksson *et al.*, 2012). Thus, there may be multiple advantages to their isolation and use in the production of valuable compounds. The economic value of the chemical industry is comparable to that of the fuel industry, although the former uses far fewer resources (Clark & Deswarte, 2008; FitzPatrick *et al.*, 2010).

The idea of producing refined products from biomass is not new; indeed, it has been used on several occasions in history, especially during times of war or when oil was scarce for other reasons. Many biorefineries were originally sulfite pulp mills, such as the Borregaard plant in Norway (established in 1889) and the Domsjö plant in Sweden (established in 1903). The Nippon Paper Chemicals biorefinery in Japan and the Lenzing plant in Austria also started out as sulfite pulp mills (Larsson & Ståhl, 2009). However, a range of industrial facilities could be converted into biorefineries, including saw mills, heating plants, pulp mills, and chemical plants that were originally established to process petroleum-derived materials. Products that have long been possible to make from lignocellulose include ethanol (which has many applications, including as a biofuel and a solvent), thickening agents and viscose. These days, viscose is regarded as an environmentally friendly alternative to chemically-intensive cotton and petroleum-based polyesters (Larsson & Ståhl, 2009).

Biofuels are an important category of biorefinery products that can be divided in three groups: solid (e.g. wood residues and pellets), liquid (e.g. ethanol and biodiesel) and gaseous (e.g. biogas and hydrogen). Biofuels can also be separated according to the processes involved in their production from biomass, which may include biological and chemical processes as well as thermochemical and physical upgrading processes (Arshadi & Sellstedt, 2008; Egnell, 2009). Tall oil is a pine-derived byproduct of the sulfate pulping process that is rich in fats and resins (Hopkins & Hüner, 2004; Altiparmak *et al.*, 2007; Ramos *et al.*, 2009). It can be used to manufacture oils, soaps, resins and FAME (fatty acid methyl esters), which are used to produce a biodiesel that has a high energy content and favorable thermal properties (Ramos *et al.*, 2009).

Most biorefineries primarily produce bulk products such as special forms of cellulose, pulp, or biofuels. However, fine chemicals, food additives, cosmetics and health-promoting agents are also viable products. New textiles and plastics made from bio-based polymer precursors that could potentially replace oil-based polymers are also under development. The ongoing development of nanotechnology will make it possible to create biomass-derived materials that are simultaneously strong, light and environmentally friendly. Other potential biorefinery products include lignin-based vanillins, yeasts and lignosulfonates that are used as cost-effective dispersing agents in concrete, dyes and asphalt (Assarsson & Blomqvist, 2005). In addition, phytosterol is a common extractive from wood that can be used to reduce the cholesterol content of margarine and other foods, while terpenes are used as solvents and in paints (ibid.).

Many types of biomass, including waste materials generated by the agricultural and food industries, are currently being studied to determine their potential for conversion into useful substances in biorefineries (Amidon & Liu, 2009; Demirbas, 2011). It is therefore important to evaluate the advantages of using forest biomass as a biorefinery feedstock compared to biomass from other sources. The separation and isolation of important chemicals from biomass must be done using modern process technologies that minimize the use of substances harmful to human health and the environment. Historically, chemical extraction technologies have been heavily reliant on resource-consuming and environmentally harmful solvents and techniques. The concepts of green chemistry were developed to address this deficiency by emphasizing the use of renewable feedstocks such as forest biomass and natural (non-organic) solvents, e.g. water and carbon dioxide in conjunction with green technologies such as supercritical fluid extraction (SFE), microwave processing and other clean synthesis methods to isolate the desired products (Clark *et al*., 2006; Arshadi *et al*., 2012).

1.5 Cost-effective goal-oriented management regimes

Young stands must be carefully tended in order to ensure the sustainable development of forest resources in terms of growth, density, structure and profitability (Mitchell, 1992). However, many silvicultural measures such as planting and pre-commercial thinning are expensive and time-consuming. The predominant goal of practical forest management has been to optimize the production of saw timber and pulpwood. However, it is currently held that forests should be managed to satisfy a wider range of goals, including traditional timber production, nature conservation, the provision of recreational facilities, and the production of biomass for biorefineries. The most important goal is to diversify forestry as this is seen as the only way to make it environmentally, socially, and economically sustainable. No single solution is universally applicable, and so a variety of solutions are needed.

Direct seeding makes it possible to establish stable and dense lodgepole pine stands at low cost by reducing the risk of root and stem deformation and enabling the use of mechanized seeding (Rosvall, 1994; Wennström *et al*., 1999). Direct seeding imitates natural regeneration and increases the stability of the resulting trees (Rosvall, 1994). The use of a close initial stand spacing significantly improves wood quality parameters, giving reduced microfibril angles while increasing the modulus of elasticity (MOE), fibre length, latewood percentage and cell wall thickness relative to wide-spaced trees (Middleton *et al*., 1995; Persson *et al*., 1995; Erikson *et al*., 2000; Lasserre *et al*., 2009).

However, dense stands are problematic in traditional forestry because they require extensive labor-intensive pre-commercial thinning in order to avoid growth stagnation (Johnstone, 1981; Pettersson *et al*., 2012). Precommercial thinning might be avoided if one instead performs schematic (e.g. corridor) thinning using forest machines to harvest biomass (Bergström *et al*., 2010; Karlsson *et al*., 2013). This makes it possible to harvest the discarded young trees for bioenergy production or for use in biorefineries rather than leaving them in the forest to rot. However, the optimum corridor harvest procedures, i.e. the corridor width that will minimize snow and wind damage to the remaining trees, remains to be determined (Valinger *et al.*, 1993; Rosvall, 1994; Bergström *et al*., 2010; Teste & Lieffers, 2011).

The established codes of practice dictate that lodgepole pine stands should be pre-commercially thinned to a density of 1300-2500 stems per hectare depending on site fertility (Pettersson *et al*., 2012). However, the advantages of thinning only to a density of around 3000 stems of sown lodgepole pine per hectare are becoming increasingly apparent (Normark, 2011). In particular, the overall stem volume and biomass production increase with the number of stems per hectare up to a stand density of 4 000-5 000 stems per hectare (Sjolte-Jørgensen, 1967; Harms & Langdon, 1976; Pettersson, 1993; Liziniewicz *et al*., 2012).

By choosing an appropriate regeneration method, stand density, harvest age and method, it is possible to create a management regime that will achieve specific goals. These may involve maximizing timber or pulpwood production, biomass growth, or some combination of the three.

1.6 Lodgepole pine forestry for new product ranges

Lodgepole pine might be an attractive source of biomass for biorefineries in boreal countries because it is a pioneer species that exhibits rapid juvenile growth and produces more biomass than Scots pine, especially of branches and needles (Norgren, 1996; Elfving *et al.*, 2001; Gardmo, 2007). Biomass is a valuable commodity due to its importance in the switch from fossil to renewable energy sources. Therefore, it may be more profitable to perform initial biomass/biofuel harvests in lodgepole pine stands instead of conventional pulpwood thinnings (Kero, 2007). For all existing lodgepole pine stands, the value of the total biomass should be compared to that of the stem volume alone. Unfortunately, there are few biomass functions for Fennoscandian-grown lodgepole pine stands. Ulvcrona (2011) and Elfving (2013a) are developing new biomass functions, both local ones and more general ones, for different types of planted and sown lodgepole pine. The distribution of biomass between tree fractions differs between dense and sparse stands, and the chemical composition also varies with age and diameter (Koch, 1996).

There is a need to develop more extensive links between the forestry and chemical industries. At present, forest companies tend to optimize their holdings to maximize the production of timber and pulpwood, while biorefineries have little control over the properties of their feedstocks. A closer collaboration would enable biorefineries to obtain assortments that are more suited to their needs and give foresters more access to this new market, enabling them to produce a wider range of profitable assortments.

1.7 Objectives

This thesis aims to unify several different perspectives on the use of lodgepole pine in biorefineries. The overall objective of the work presented herein was to evaluate the potential for cost-effective cultivation of lodgepole pine (*Pinus contorta*) in Fennoscandia and to determine the optimal uses of lodgepole pine biomass in bioenergy and biorefinery applications. The specific goals were to:

1. quantify the stem volume and biomass production of direct seeded lodgepole pine stands grown under different site conditions with different stem densities in mid-northern Sweden, at an early age that would permit extensive harvesting of biomass (I).

2. compare the impact of different silvicultural regimes on lodgepole pine stands, 19-20 years after their direct seeding and five years after a pre-commercial thinning. Key variables of interest in this comparison were stem volume and diameter, biomass production, and damage frequency (II).

3. compare the fatty- and resin acid contents of the stemwood of mature lodgepole pine and Scots pine grown at different sites in northern Sweden, to determine the potential for the large scale isolation of pine stemwood extractives for use in biorefineries (III).

4. identify and quantify the chemical compounds present in the stemwood, bark, branch wood, needles and cones of 30-year-old lodgepole pine trees grown under a direct-seeding based regime and to consider the potential industrial applications of each fraction (IV).

5. estimate the potential of different biorefinery products from tree biomass and the raw material requirements for the most promising product areas, and to analyze the connection between electricity prices and the prices of different tree assortments (V).

2 Materials and Methods

2.1 Study sites

Three sets of study sites in northern Sweden were used.

The biomass measuments in Study I and the chemical analyses in Study IV focused on a group of 30-year-old direct seeded lodgepole pine stands located in mid-northern Sweden (latitude 61.8-62.1 ^{o}N; Fig. 2). These stands are located on land owned by Holmen Skog and are among the oldest commercially direct seeded lodgepole pine stands in Sweden. Eight stands located in Härjedalen county close to the Scandinavian mountain range (altitude 400-610 m.a.s.l.), and another eight in Hälsingland county close to the Gulf of Bothnia (altitude 230-400 m.a.s.l.) were examined in Study I. The 16 stands were divided into four site index (SI) groups based on the dominant height of Scots pine at 100 years of age (Hägglund & Lundmark, 1977) as estimated by the landowner Holmen skog AB. The site indices used to delimit the different groups were 16 and 20 m (Härjedalen), and 22 and 26 m (Hälsingland). The lower site index values for Härjedalen reflect the harsher weather conditions and lower nutrient availability at this site. The field layer vegetation at the sites ranged from reindeer lichen (*Cladonia rangiferina*), heather (*Calluna vulgaris*), black crowberry (*Empetrum nigrum*), lingonberries (*Vaccinium vitis-idaea*) and bilberries (*Vaccinium myrtillus*), to grasses (*Poaceae* species a.k.a. *Gramineae*), and woodland geraniums (*Geranium sylvaticum*). In Study IV, four trees were felled for biomass measurements and chemical extractions. Two trees were taken from one of the mountainous Härjedalen-stands and two from one of the lower Hälsingland stands.

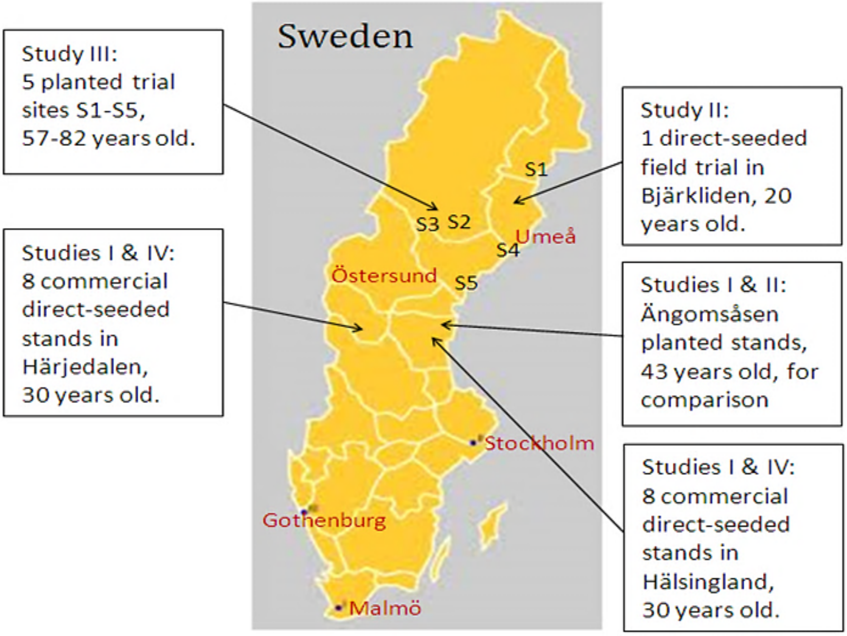

Figure 2. Map showing the study sites (Study I-IV).

The chemical analyses reported in Study III focused on five old trial sites that had been planted with both lodgepole pine and Scots pine, all of which are located in Northern Sweden (latitude 63.0-65.4 ^{o}N, altitude 20-420 m.a.s.l.; Fig. 2). Some of the stands were mixed Scots pine/lodgepole pine stands while others had the two species planted in separate neighboring plots. The stands were between 57 and 82 years old and were therefore much older than the directly seeded stands considered in studies I, II and IV.

Study II focused on trees growing at a site in Bjärkliden outside Norsjö in Västerbotten (latitude 64.5 °N, altitude 310-340 m.a.s.l.; Fig. 2). A regime comparison trial was established at this site in September 2006, 14 vegetation periods after it had been directly seeded in July of 1993. The study focused on lodgepole pine stands in a commercial forest owned by Holmen Skog. The dominant field layer at the site is bilberry (*Vaccinium myrtillus*) and the site index is 20. The experimental site is located on a south-southwest facing slope. The stands at this site were the youngest considered in this work, being only 20 years old.

An older set of planted lodgepole pine stands at Ängomsåsen (lat. 62.4 °N, alt. 105 m.a.s.l.) 10 km SW of Sundsvall, Sweden was used in the stem volume comparisons presented in Studies I and II (Fig. 2). These stands were originally planted by the forest company SCA in 1970 as part of a spacing trial and were assigned to SLU in 1983 to secure further inventory (Elfving, 2006). Five different spacings were considered (1.1 m; 1.6 m; 2.0 m; 2.85 m and 4.0 m) and the site has been studied on four different occasions over a period of 30 years (1983, 1992, 1997 and 2006) (ibid.). The soil at the site is fertile and grass is dominant in the field layer, as is the case for the more productive stands examined in Hälsingland. The planted stands at Ängomsåsen are situated 30-50 km from the inventoried and direct seeded stands in Hälsingland that were examined in Study I.

2.2 Stem volume and biomass measurements (Papers I & II)

Studies I and II both examined the stem volume and biomass production of lodgepole pine stands. For Study I, eight circle plots with areas of 100 m^2 each were laid out in each stand using ArcGIS. Five stands were shaped in such a way that eight such circle plots could not be accommodated and so only seven, seven, three, six and six plots were defined for these stands, respectively (the stand borders were not precisely defined in the register data and so the GIS polygons did not match up perfectly with the recorded stand borders in some cases). The stand where only three circle plots could be measured had a long and narrow shape and was situated next to a road (circle plots could not be positioned on the road bank). In total, 117 circle plots were defined. Within each circle plot, all trees whose height was $\geq$1.3 m were subjected to diameter-at-breast-height (1.3m) measurements. About 20% of the trees in each circle plot were also randomly selected for height and crown length measurement. Measurements were performed in the autumn of 2010 and the summer of 2011.

Height curves were then constructed for each circle plot using Näslund’s (1936) equation. The volume (on bark) of each tree with a stem diameter of more than 50 mm was calculated using Eriksson’s (1973) equation, while Andersson’s (1954) equation for the volume (on bark) of small pines in northern Sweden was used for all trees with a diameter of $\leq$ 50 mm. Elfving’s (2013a) biomass function (the equation for biomass above stump level) for lodgepole pine was used to calculate the biomass of each stand considered in Study I.

Four trees, two from Härjedalen and two from Hälsingland were cut for biomass measurements in September 2011 as described by Ulvcrona (2011). The measured biomass data were compared to the calculated biomasses reported by Elfving (2013a) for each of the four trees. Biomass samples from these trees were also used to determine the chemical contents of different tree fractions in Study IV. One small tree with a DBH of 9 cm and one large tree with a DBH of 14 cm were cut down from each stand. In all cases, the trees were chosen to be representative of the stand in terms of tree dimensions and crown shape, and had sustained no visible damage. The DBH, height and crown length of each tree were measured, and the crowns were divided into four strata of equal length according to the biomass measurement schedule proposed by Ulvcrona (2011). A sample branch was collected from each stratum in different directions (a branch that would have pointed in the northerly direction from stratum 1, one that would have pointed east from stratum 2, one pointing to the west from stratum 3 and one pointing south from stratum 4) along with six discs cut from the stem at various positions (the base; breast height, i.e. 1.3 m; and at 30%, 55%, 70%, and 85% of the tree's total height). These and the remaining parts of the trees were weighed in the field to determine their fresh weights. The sample branches and discs were then frozen and their dry weights were measured after drying at 85 °C for 48 hours.

In Study II, field measurements were conducted at Bjärkliden towards the end of the autumn in 2011. The DBH (1.3 m height) was recorded for all trees within the net plots. The heights of selected trees (five of the tallest trees and an additional 20-30 sample trees per plot representing all DBH-classes) were measured at the same time. Height curves and stem volumes were calculated as in Study I (Näslund, 1936; Andersson, 1954; Eriksson, 1973). Mean DBH values were calculated by weighting the mean diameter against the basal area and are referred to as Dgv values. Local biomass functions were then constructed for lodgepole pine after destructive biomass harvesting of 29 sample trees using a method reported by Ulvcrona (2011). Biomass functions were constructed for the stem above stump including bark, and for the total tree including stem, bark, branches, foliage and dead branches. During the inventory conducted in 2011, 24 different types of damage were recorded, based on the position at which the tree had sustained damage and the severity of the damage. Trees that had sustained the most severe types of damage were divided into two groups: trees that were laying or leaning significantly (but were still alive) constituted the first group, while trees that had died or had broken stems (either below the crown or within the crown but not a broken top shoot) constituted the second group.

2.3 Comparison of different silvicultural regimes (Paper II)

A field experiment had been established in September of 2006 to study different lodgepole pine management regimes in a 14-year-old stand in Bjärkliden, Västerbotten. The studied regimes differed mainly in terms of the spacing of the trees, the thinning method applied, and the fertilization scheme used. The trial involved two blocks, each with seven 400 m^2 plots and two 700 m^2 plots, with net dimensions of 20 x 20 m and 20 x 35 m, respectively. Each plot was surrounded by a 5 m buffer zone. Pre-commercial thinning (PCT) of a specified number of stems for each treatment was performed in July 2007 (year 15) on the gross plots using a motor-manual brush saw. The treatments applied to the 400 m^2 plots were as follows:

1. Conventional regime - PCT to 2200 stems/ha (only one replicate per block since this represents a conventional stem density)
2. High biomass regime - no PCT (two replicates in each block)
3. Large dimension regime - PCT to 1700 stems/ha (two replicates per block)
4. Combined regime aiming for both high biomass and timber trees - PCT to 4500 stems/ha (two replicates per block).

Corridor thinning was performed in June 2012 in the two larger plots in each block to achieve a total corridor area of approximately 70% of the total plot area. The thinning was performed motor-manually in order to avoid causing machine-related damage. The corridor treatments are more thoroughly described in Karlsson (2013).

2.4 Chemical analyses (Papers III & IV)

In Study III, the stemwood of 60 trees (30 lodgepole pines and 30 Scots pines) aged 57-82 years was sampled using a 5 mm increment borer at a point 1.3 m above ground level. Only dominant healthy and undamaged trees were considered. The border between the sapwood and heartwood was marked on the freshly cut cores in the field. Prior to analysis, the heartwood and sapwood fractions were separated and the samples from the two cores for each tree were pooled to form one heartwood and one sapwood sample. The fatty- and resin acids were isolated by Soxhlet extraction using a mixture of petroleum ether and acetone (90:10 v/v) as the solvent for 1 hour (12 cycles). The extracts were then analysed by GC-MS. An internal standard (heptadecanoic acid) was added to enable the quantitative analysis of fatty acids and resin acids. The analyses were performed at the Swedish University of Agricultural Sciences in Umeå, Sweden.

In Study IV, fewer trees were sampled but every above-ground fraction of the studied lodgepole pine trees was analyzed. One smaller tree (DBH=9 cm) and one larger tree (DBH=14 cm), by the age of 29-30 years, were cut down by chainsaw from each stand in September 2011 (giving a total of four sampled trees). Stem discs were sawn from the top and base of each tree and the bark was separated and combined to give one bark sample per tree. Branches with needles and cones were retrieved from the whole crown. Cones were only present on the trees from the mountainous region. In total, 22 samples representing six distinct fractions were obtained from the four trees (stem top, stem base, bark, branches, needles and cones). The samples were dried at 105 ^{o}C for 16 hours, milled using a Retsch knife mill (1 mm sieve) and stored in sealed plastic bags pending chemical analysis. They were then extracted thoroughly for 4 hours in a Soxhlet extraction apparatus with hexane as the solvent. The extracts were weighed carefully and the extractive yield was calculated as a percentage of the original dry-weight sample mass. The extracts were then analysed by GC-MS and the lipid components were quantified based on their response factors (Rf), in conjunction with internal standard calibration. Fatty acids, fatty alcohols, fatty aldehydes, alkanes, sterols and wax esters were quantified by generating seven-point linear calibration graphs using octacosanoic acid, decanol, dodecanal, hentriacontane, stigmasterol and stearyl palmitate as external standards. The analyses were performed at The Green Chemistry Centre of Excellence at the University of York, United Kingdom.

An extra two discs were cut from each sample tree in Study IV, one from the top of the stem and one from the base, for heartwood analysis. Each disc was dyed with a 50/50 blend of saturated sulphanilic acid ($C_6H_7NO_3S$) and 10% sodium nitrite ($NaNO_2$) as described by Cummins (1972). This dye gives the heartwood a darker red color than the sapwood, making it possible to determine the amount of heartwood in each disc by measuring the length of the heartwood (mm) along eight radial axes and summing the areas of the eight wedges defined by two adjacent axes and the boundary between the heartwood and the sapwood.

2.5 Questionnaire study and price analysis (Paper V)

In 2011, a questionnaire about tree products and the potential expansion of biorefinery businesses was sent out to 102 individuals working in industrial organizations, businesses, and academia (Study V). To the best of our knowledge, all of the targeted individuals worked on issues relating to wood products. Most of them were resident in Sweden (ca. 70%) but some lived in other countries. A reminder containing a second copy of the questionnaire was sent out two months later. The questionnaire had two parts with 16 questions in total. Three questions concerned the commercial potential of tree biomass and biorefinery products in general. The following 13 questions asked the respondents to select lignocellulosic products that they considered to have reasonable commercial potential and to estimate their product development requirements and the quantities of raw materials and electricity required for their large-scale production. In Study V, an electricity- and wood raw material price analysis was performed as well, by monitoring the prices between 2000 and 2011. The correlation between them was tested by linear regression and Pearson correlation values. For more details on the price analysis, see Karlsson (2013).

2.6 Statistical analyses

Statistical calculations were performed using Analysis of variance in Minitab 15 (Minitab Inc., USA). To check the validity of the assumption of constant variance, plots of residuals against fitted values were studied. A significance threshold of 0.05 (corresponding to a 95% confidence interval) was used in all studies when testing p-values to determine whether the null hypothesis could be rejected. Differences between samples were analysed using Tukey´s test.

In Study III and IV, multivariate tools such as Principal Component Analysis (PCA) were used (Jackson, 1991; Beebe *et al.*, 1998; Brereton, 2003). These statistical tools facilitate the identification of important trends in large data sets. PCA results are typically visualized in score plots, which are used to analyse clustering, outliers and gradients of objects (e.g. individual samples); and loading plots, which are used to study the corresponding patterns in the variables (e.g. chemical substances). Correlations between observations and variables can be identified by inspecting both the score plots and the loading plots. All multivariate analyses were conducted using SIMCA (Umetrics, Sweden) and the PLS Toolbox (Eigenvector, USA) for Matlab (The Math Works, USA).

3 Results and Discussion

3.1 New silvicultural regimes for the production of biorefinery-oriented assortments

3.1.1 Stem volume and biomass production of lodgepole pine (I)

The 30-year-old stands in Härjedalen had a mean stemwood production of 71 m^3/ha while those in Hälsingland averaged 154 m^3/ha (Table 1). Mean levels of almost 200 m^3/ha were achieved at the best sites, rising to about 300 m^3/ha for the best circle-plots (≥3000 stems/ha) even though one or two pre-commercial thinnings had been performed at the sites. There was a positive correlation between stem density and stem volume. Dry weight biomass ranged from 38 tons/ha on average in Härjedalen, to 78 tons/ha on average in Hälsingland. 100 tons/ha of d.w. biomass were achieved at the best sites, rising to about 140 tons/ha for the best circle-plots.

There were significant differences between regions and site index groups with respect to both stem volume and dry weight biomass. The stems, including bark, accounted for around 70% of the total aboveground biomass of the sampled trees, with living branches and needles representing approximately 10% of the total each. The dry weight biomass of the needles was roughly equal to that of the living branches.

Table 1. Mean stem volumes and dry weight biomass values for the four site index groups and for each region as a whole, 29-30 years after direct seeding (Study I). All of the observed differences between the two regions and between site index groups within the regions were significant ($p \leq 0.05$).

Parameter	Region	Site index group	Site index	29-30 years after sowing
Stem volume (m^3 ha^{-1})	Härjedalen	1	14-16	38.81
	Härjedalen	2	20	108.56
	Hälsingland	3	22-24	131.33
	Hälsingland	4	26	180.95
	Härjedalen		Average	71.30
	Hälsingland		Average	154.10
D.W. biomass (t ha^{-1})	Härjedalen	1	14-16	25.31
	Härjedalen	2	20	51.80
	Hälsingland	3	22-24	64.73
	Hälsingland	4	26	92.55
	Härjedalen		Average	37.86
	Hälsingland		Average	77.71

The trees had mean diameters of 8-16 cm and mean heights of 6-13.5 meters, depending on site fertility. Thus, especially the lodgepole pines in the more fertile stands had achieved substantial diameter and height growth given that they were only 30 years old and were growing in northern Sweden. The denser stands (≥3000 stems/ha) had only slightly lower stem diameters than the sparser stands, indicating that it may be favorable to aim for a stem density of about 4000 stems per hectare. This is consistent with the results of previous studies on lodgepole pine (Varmola *et al.*, 2000; Liziniewicz *et al.*, 2012), which showed that higher stem densities generally yield greater quantities of biomass. Our results suggest that even denser stands (around 4000 stems/ha) will produce good quality biomass for biorefineries as well as pulpwood and timber trees. To achieve equivalent stem densities by planting would be very expensive. The high biomass production means that a partial or complete biomass harvest is possible within a very short time period.

The denser stands (≥3000 stems/ha) yielded results comparable to those observed for planted lodgepole pine stands (2500 stems/ha), which attained stem volumes of 350 m^3/ha at a dominant height of 18 meters. Interestingly, denser direct seeded stands reach a similar stem volume as conventional planted stands at the same dominant height, even though the diameter might be somewhat lower. Almost all of the direct-seeded stands had undergone one or two pre-commercial thinnings before our study was conducted, and were managed under a traditional silvicultural regime that emphasized pulp production. It seems reasonable to suggest that biomass yields of 200 m^3 (or 100 tons) per hectare could be achieved using current methods within 30 years of direct seeding with lodgepole pine, with approx. 70 tons of this being stemwood along with 10 tons of needles and 10 tons of branches.

The predicted values obtained using Elfving's (2013a) functions were in good agreement with the experimental data for the four sampled trees. The DBH:green weight ratios for the mid-Swedish stands were comparable to those for stands in British Columbia, Canada (Fig. 3), although the larger Canadian trees were somewhat heavier (Koch, 1996). Similar increases in weight may occur in the Swedish stands as they continue to grow.

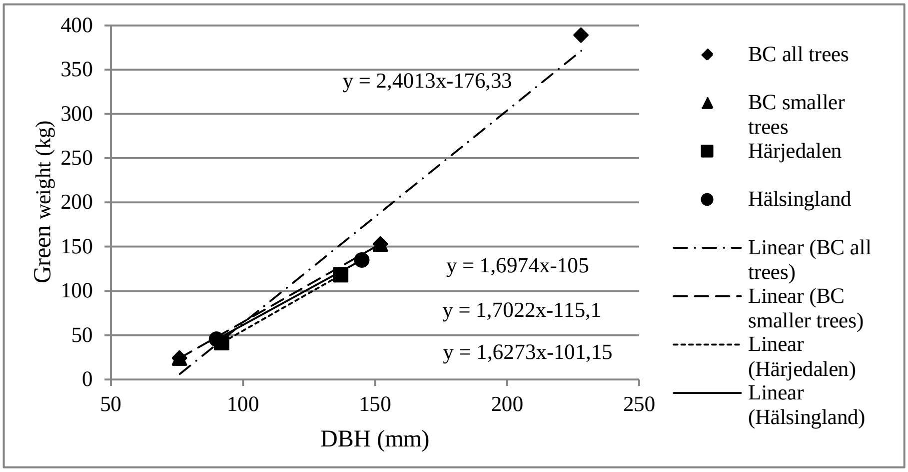

Figure 3. Biomass (green weight, kg) plotted as a function of DBH (Diameter Breast Height, mm) in sampled trees from Härjedalen and Hälsingland, Sweden (Study I & IV); and British Columbia, Canada (Koch, 1996).

The mountain stands considered in Study I were generally healthier and less damaged than those at lower altitude. Some of the lower sites had a high frequency of boulders, which may have been why they were chosen for direct seeding with lodgepole pine rather than planting with Scots pine 30 years ago. Direct seeding with lodgepole pine may have been chosen at the mountain sites due to their harsh climate and environment. However, the trees at the mountain site did not sustain any more snow damage than those at the lower site.

3.1.2 Goal-oriented lodgepole pine forestry (II)

The four regimes (Conventional, High biomass production, Large dimension trees and Combined) were evaluated 19-20 years after direct seeding. The High biomass regime produced 144% more biomass and 134% more stem volume than the Conventional regime, and 157% more biomass and 143% more stem volume than the Large dimension regime (Table 2). The biomass production of the High biomass regime was significantly greater than that for all other regimes, and the stem volumes of the High biomass and Combined regimes were significantly higher than those for the Conventional and Large dimension regimes. Interestingly, however, the mean diameters of the 1000 and 2000 largest trees per hectare did not differ significantly between the four regimes, so all of the regimes could potentially yield harvestable timber trees in the future. Lindgren & Sullivan (2013) performed a similar study on planted Canadian lodgepole pine, albeit with wider spacings (250-2000 st/ha), and found that the mean DBH and volume growth increments per tree were not affected by stand stem density.

Table 2. Per-regime mean stem volumes, biomass production, numbers of stems per hectare, dominant heights, mean diameters (Dgv), and mean diameters for the 1000 and 2000 largest lodgepole pine trees per hectare for each treatment considered in Study II. Means followed by different superscripted letters differ at the $p<0.05$ level according to Tukey´s multiple comparison test (no test was performed for the number of stems).

Regime	Number of stems/ha	Dgv (cm)	Dgv 1000 largest trees/ha (cm)	Dgv 2000 largest trees/ha (cm)	Stem volume (m^3/ha)	Biomass (ton/ha)	Dominant height (m)
Conventional	2 150	8.0^b	8.9^a	8.0^a	31.5^b	21.6^c	7.2^a
High biomass	15 331	6.2^c	8.8^a	8.1^a	73.9^a	52.8^a	7.6^a
Large dimension	1 663	8.9^a	9.6^a	-	30.4^b	20.6^c	7.5^a
Combined	4 481	7.7^b	9.6^a	8.7^a	63.0^a	39.5^b	7.7^a

The lowest damage levels (in terms of relative tree numbers and basal area) with respect to laying and leaning trees were observed under the unthinned High biomass regime (2%); the highest levels occurred under the Large dimension regime (10%), where the spacings between trees were wider. The greatest number of dead trees was observed under the High biomass regime, and most of those dead trees had a diameter at breast height (DBH) of less than 5 cm, i.e. some self-thinning occurred in this case. At a corridor width of 0.7 m, 27 tons of biomass and 38 m^3 of stem wood were extracted per hectare from the larger plots; the corresponding values for a corridor width of 1.4 m were 17% and 18% higher, respectively. See Karlsson (2013) for more details on the corridor thinning that was conducted. According to measurements taken in 2008 and 2011, the High biomass regime provided a comparable or even somewhat better level of stem volume development compared to conventionally planted lodgepole pine stands in Ängomsåsen, Sundsvall, with a stem density of 2500 stems (Fig. 4) (Elfving, 2006; Elfving, 2013b).

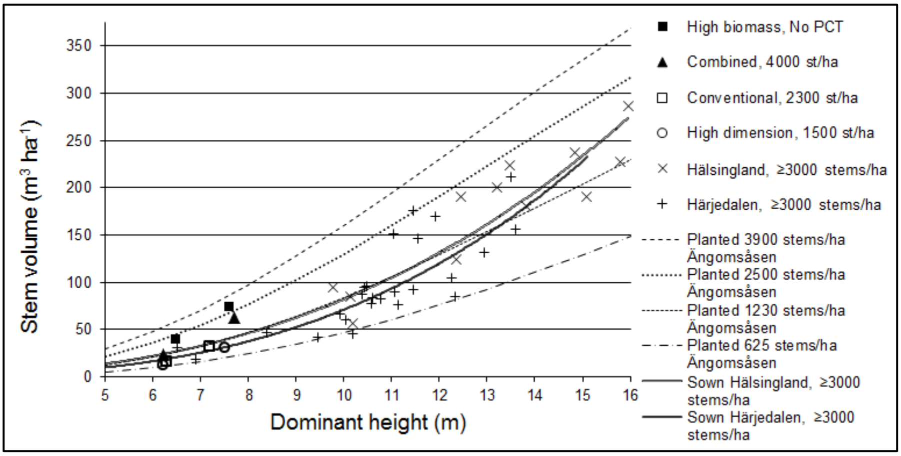

Figure 4. Stem volume development (m^3/ha) with respect to dominant height (m) for the different Bjärkliden regimes (Study II; squares, triangles and circles) compared to dense sown stands in Härjedalen and Hälsingland (Study I; crosses and solid lines) and planted stands at Ängomsåsen, Sundsvall (Elfving, 2006; Elfving, 2013b; dotted lines).

The High biomass regime will probably achieve the same volume development as the densely planted stands (3900 stems/ha) in a few years. The volume production under the Combined treatment did not differ much from the Conventional and High dimension regimes in 2008. However, this regime has since produced substantial volume growth and is currently not far behind the High biomass regime in terms of stem volume development.

Each regime seems to meet its intended purpose, but if the goal is an early biomass harvest combined with a later timber harvest, the optimal stem density before the first biomass harvest seems to be about 4000 stems per hectare after precommercial thinning (PCT). Alternatively, corridor thinning could be performed once the stand reaches 20 years of age with no previous PCT, giving a residual stand density of 4000-5000 trees per hectare with the trees arranged in clusters and allowing for the extraction of 30 tons of biomass. A total biomass harvest when the stand is this age or slightly older (c.f. Study I) might also be an attractive option. It will be necessary to study stands that have undergone corridor harvesting more extensively in order to determine the viability of the remaining trees. However, these results clearly show that lodgepole pine stands can be cultivated and managed to fulfill different goals, which may range from regeneration to high volume harvesting.

3.1.3 The chemical composition of different tree fractions (III & IV)

Thirty-one different fatty- and resin acids (21 fatty- and 10 resin acids) were identified in the extracted stemwood samples in Study III (Table 3). The highest observed concentration of any individual acid was 14 mg/g. Mature lodgepole pine (57-82 years) from northern Sweden was found to contain less fatty- and resin acids (0.2-2.6%) than mature Scots pine (0.2-4.1%) per unit of dry weight. This is consistent with the results of Sable *et al.* (2012) for Scots pine and lodgepole pine in Latvia, but contradicts the findings of Sjöström (1993) and Koch (1996).

There were significant differences ($p \leq 0.05$) between the two wood tissue types (heartwood and sapwood) for both species and with respect to all chemical components (i.e. fatty acids, resin acids and total extractive content). The amount of heartwood is thus the most important determinant of the extractive content of pine stemwood. It may therefore be necessary to determine the proportion of heartwood in trees of different ages and different stem diameters, and to separate the heartwood from the sapwood if the goal is to maximize the industrial utility of each tree.

Table 3. Extracted fatty and resin acids (Study III) showing the name, number of measurements with nonzero concentrations, and maximum concentrations for each acid. Entries 1-21 are fatty acids, 22-31 are resin acids.

Number	Name	Number above zero	Max conc. (mg/g)
1	Hexadecanoic acid	120	0.54
2	Heptadecanoic acid	118	0.16
3	Linolenic acid	92	0.32
4	9,12-Octadecadienoic acid	113	0.89
5	Oleic acid	120	1.26
6	Nonanoic acid	40 *	0.06
7	Linolenic acid, anteiso	73 *	0.64
8	Octadecanoic acid	57 *	1.43
9	Octanoic acid	19 **	0.04
10	dodecanoic acid	10 **	0.02
11	Tetradecanoic acid	37 **	0.07
12	Pentadecanoic acid	26 **	0.06
13	Heptadecanoic acid, anteiso	7 **	0.34
14	Heptadecanoic acid, anteiso	30 **	0.42
15	(E)-9-Octadecenoic acid	32 **	0.21
16	trans-9-Octadecenoic acid, anteiso	12 **	0.50
17	11-cis-Octadecenoic acid	19 **	0.08
18	Eicosanoic acid	28 **	0.15
19	Docosanoic acid	25 **	0.91
20	Docosanoic acid, anteiso	24 **	0.06
21	Tricosanoic acid	5 **	0.08
22	Pimaric acid	117	2.63
23	Pimaric acid, anteiso	89	0.59
24	Isopimaric acid	119	2.75
25	Isopimaric acid, anteiso	80	3.03
26	Dehydroabietic acid	121	13.67
27	Abietic acid	110	7.59
28	7-Oxodehydroabietic acid	106	3.25
29	Pimaric acid, anteiso	54 *	2.87
30	Dehydroabietic acid, anteiso	42 *	2.06
31	Isopimaric acid, anteiso	20 **	0.79

* Fewer than 80 measurements with nonzero conc.

** Fewer than 40 measurements with nonzero conc.

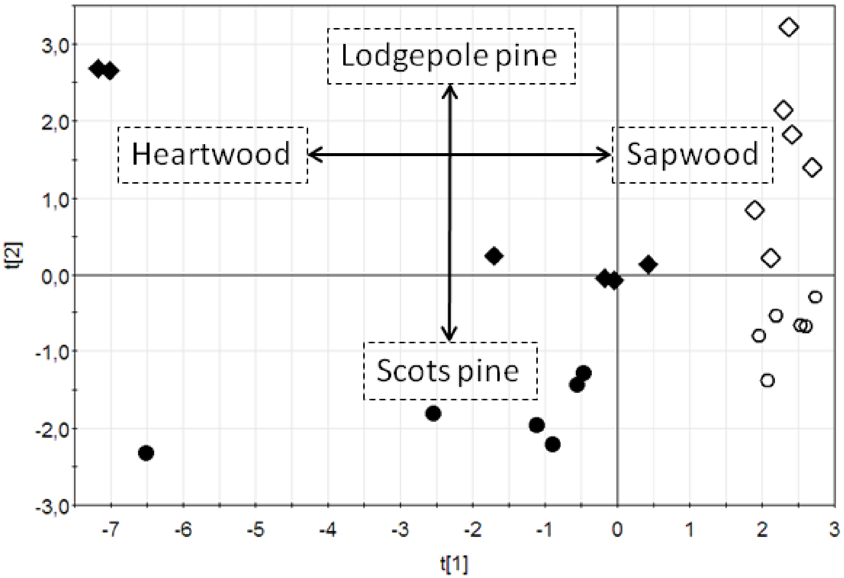

Figure 5. PCA score plot for fatty and resin acid concentrations in the heartwood and sapwood of Scots and lodgepole pines from site S2 (Study III). Circles indicate data for Scots pine, diamonds for lodgepole pine, open symbols for sapwood, and filled symbols for heartwood.

In general, the concentration of resin acids was higher than that of fatty acids. For lodgepole pine, the resin- and fatty acid concentrations were 3.4 and 1.2 times greater in the heartwood than in the sapwood, respectively. The corresponding factors in Scots pine were 5.0 and 2.5. Thus, the acids were more evenly distributed between wood types in lodgepole pine than in Scots pine. Visual inspection of the score and loading plots showed that the resin acids were mainly associated with the heartwood while the fatty acids were more strongly associated with the sapwood (Fig. 5; loading plot not shown). Both the two species and the two wood types are clearly separated in the score plot (Fig. 5). Lodgepole pine produces more biomass and has a higher growth rate than Scots pine (Elfving *et al.*, 2001). Therefore, on the stand level, lodgepole pine is a better option for general fatty- and resin acid extraction because of this species' more even distribution of extractives across the different wood types. A mature stand of cultivated lodgepole pine could provide at least 300 m^3 per ha of stemwood, corresponding to about 150 tons (d.w.) of biomass containing approx. 150 kg of fatty acids and 1 ton of resin acids per hectare.

While Study III investigated the fatty- and resin acid composition of the stemwood, Study IV focused on the extractive content and composition of all of the aboveground fractions from lodgepole pine: stemwood from the top, stemwood from the base, bark, branches, needles and cones. The bark was found to have the highest extractive content (16% by mass on average), and the stemwood had the lowest (1% on average) (Table 4).

Table 4. Extractive yields (percent of d.w.) for the different fractions after Soxhlet extraction with hexane (Study IV).

Fraction	Tree 1, Mount. Small	Tree 2, Mount. Large	Tree 3, Lower Small	Tree 4, Lower Large	Average Yield*	Dry matter** (Average, %)
Bark	14.49	19.96	13.41	17.68	16.4 ± 3.0[a]	42.4 ± 1.8
Branches	7.16	9.89	6.20	6.49	7.4 ± 1.7[b]	52.1 ± 1.0
Needles	6.07	9.47	4.14	5.50	6.3 ± 2.3[b]	45.3 ± 1.0
Stem at base	0.99	0.31	3.46	1.59	1.6 ± 1.4[c]	44.6 ± 1.9
Stem at top	0.77	1.35	0.46	0.75	0.8 ± 0.4[c]	36.3 ± 2.9
Cones	1.55	1.77	-	-	1.7 ± 0.2	68.2 ± 3.8

*Yields are quoted as means ± one standard deviation.

**The dry mass of the pooled sample as a percentage of its original fresh mass.

[a,b,c] Yield values with different superscripted suffixes differ significantly at the 0.05 probability level according to Tukey’s multiple comparison test (cones were excluded from this test because no cones were obtained from trees 3 and 4).

These results were expected because the bark serves as a barrier against intruders and diseases and normally protects the stem, so the tree benefits from having large quantities of extractives there. There were significant differences between the six fractions, on average over trees. This is consistent with the results of Study III, which showed that the extractive contents of the heartwood and sapwood differed significantly. Sample discs were taken both from the top and the base of the tree, because the base contains more heartwood than the top (Fig. 8). The base fraction can therefore be compared to the heartwood in Study III, while the top fraction corresponds to the sapwood fraction. As in Study III, there were no significant differences between single trees, sites (i.e. climates) or tree sizes in Study IV. Tree size seemed to be a better predictor of extractive content than site: larger trees tend to have more extractives per unit dry weight.

Principal component analysis (PCA) revealed that the different fractions had distinct extractive compositions (Fig. 6). In the PCA score plot, the needle samples were located below those for the other fractions, indicating that their extractive composition differed substantially from those of the other fractions. The compounds primarily found in needles included wax esters and fatty alcohols such as n-heptacosanol and n-nonacosanol. The top stemwood from the larger trees (located on the right hand side of the score plot, Fig. 6) were separated from those from the smaller trees (on the left of the plot). This may reflect the formation of heartwood in the stem tops of the larger trees.

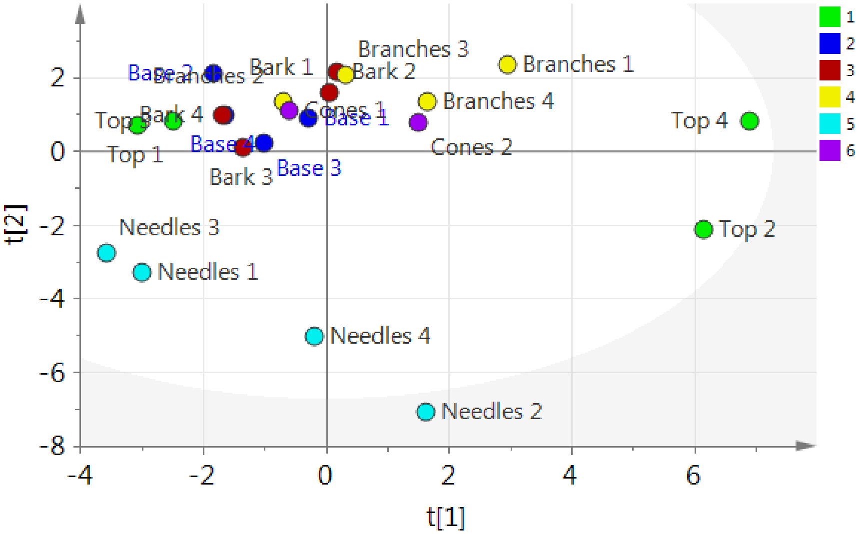

Figure 6. Score plot for all observations showing the differences between the fractions in terms of their extractive contents (Study IV).

When the needle fraction was excluded from the analysis, a distinct separation between the stem fractions and the other fractions was observed. The bark and the branches could also be distinguished. The diterpenes pimaral and epimanool and the fatty acids hexadecanoic acid and oleic acid were primarily associated with the stem wood. The total extractive content of the base stemwood was higher, but there was a greater degree of variation in the upper parts, which are closer to the canopy where many important chemical processes such as photosynthesis occur. The bark fraction was rich in diterpenes and ketones, but the outer bark also seemed to contain abundant wax esters. The branch fraction had the second highest extractive concentration after the bark (Table 4), and contained diterpenes, ketones, fatty acids and wax esters. The cones were unique in that they had a very strong pine scent, which is consistent with their high contents of aromatic compounds such as 1-methoxy-4-[1-(4-methoxyphenyl)vinyl]benzene and diterpenes such as cryptopinone.

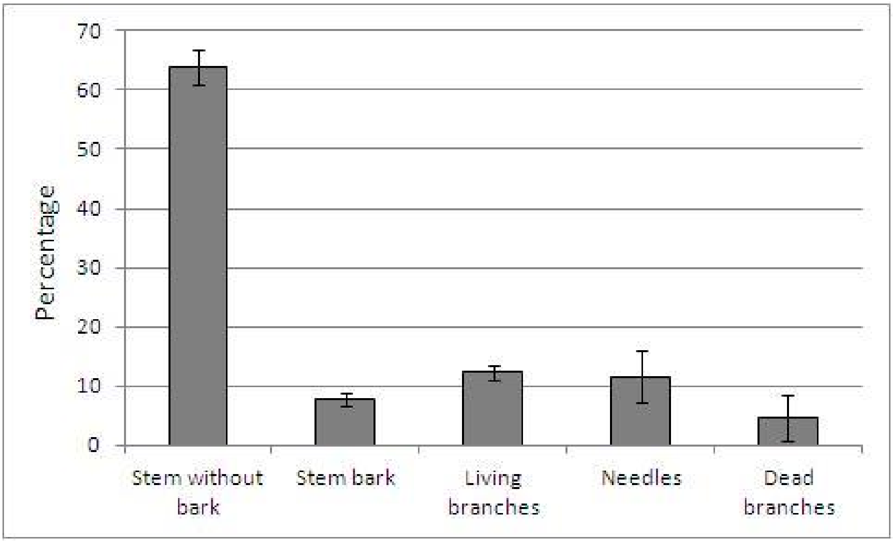

Figure 7. Partitioning of the different tree fractions (as a percentage of the dry weight of the total aboveground biomass) for the four sample trees considered in Studies I & IV. The vertical bars indicate the average values for all four trees; the staples span a range corresponding to one standard deviation (1 STD) in each case.

Based on the average biomass production for the stands from which the sampled trees were taken, the partitioning of the trees' biomass across the different fractions (Fig. 7) and the yields of the different fractions, a 30-year-old lodgepole pine stand in the lower area of Hälsingland could produce about 950 kg of stem extractives, 990 kg of bark extractives, 660 kg of branch extractives and 370 kg of needle extractives per hectare. A mountainous stand in Härjedalen of the same age could produce about 270 kg of stem extractives, 775 kg of bark extractives, 580 kg of branch extractives and 600 kg of needle extractives per hectare. Focusing on a specific compound group, the mountainous stand produced about 6.5 kg of crude needle wax per hectare while the lower stand in Hälsingland gave 3.0 kg needle wax per hectare. While not all extractives are economically valuable for chemical production, and the number of sampled trees was small, these results provide a useful indication of the chemical feedstock supplies that can be obtained from lodgepole pine stands today, using the studied management regime.

There are more environmentally friendly solvents than petroleum ether, acetone and hexane, and more modern extraction techniques than using Soxhlet technology. Examples include supercritical fluid extraction (SFE) with carbon dioxide or water, and microwave extraction. These methods are less harmful to the environment than conventional techniques, require less solvent and allow for efficient solvent recycling. Traditional extraction techniques were used in this work because they are efficient, well proven and inexpensive. However, identical biomass samples to those used in Study IV were subjected to various supercritical extractions and microwave extractions at the Green Chemistry Centre of Excellence in York, to compare the performance of different green and conventional extraction methods for pine biomass. These results will be presented in papers that are beyond the scope of this thesis. It should be noted that if the industrial-scale extraction of pine biomass is being contemplated, green techniques should be used in preference to traditional methods where possible.

Figure 8. The distribution of heartwood in stemwood samples. The figures show stem base and top discs from four 30-year-old trees (Studies I & IV) that have been dyed with a reagent that stains heartwood dark orange and sapwood pale orange. Samples from tree No 1 are shown on the top left, No 2 on the top right, No 3 on the bottom left, and No 4 on the bottom right. Heartwood is present at the center of all the base discs and there is a small amount of it in the middle of the top disc from tree No 2 (top right).

The stand's stem density is not the only important factor in determining the biomass partitioning within the trees. The lower stand in Hälsingland had 2338 stems per hectare, with living branches accounting for ca. 11% of the total biomass, dead branches for 7%, and needles for 8%. The mountainous stand in

Härjedalen had 3663 stems per hectare because it had undergone only one pre-commercial thinning whereas the lower stand had experienced two. In the mountainous stand, branches accounted for ca. 13% of the total biomass, needles for 15% and dead branches for only 1.5%. The lower site thus had a higher total branch share but 40% of those branches were dead (compared to 12% in the mountainous stand). Moreover, the remaining live branches at the lower site had fewer needles than those in the mountainous stand. The mountainous trees had a crown limit of ca. 2 meters above ground whereas trees from the lower site had crown limits of about 5 meters. A higher stem density would theoretically lead to losses of lower branches and encourage crown lift (Mansfield *et al.*, 2007), but it has to be considered that the lower site had a better nutrient supply and was more favorable for growth overall, which may encourage the self-pruning of lower branches. The mountainous site has a shorter growing season and a lower temperature sum, so trees grown there may increase their needle production to compensate.

3.1.4 Economy and product potentials (V)

Twenty-four individuals completed the questionnaire about the commercial potential of biorefinery products. Overall, 67% of the respondents worked at major businesses, representing enterprises such as Borregaard, Nippon Paper, Domsjö, Chemrec, SCA, Akzo Nobel, Preem, Lenzing, Swerea and Metso Paper. Business organizations and academia accounted for ca. 17% of the respondents each.

The responses indicate that heat and electricity are considered to have the greatest potential returns on investment over both five- and ten-year periods, followed by solid wood products, bioenergy assortments and textiles (Fig. 9). Fuels and chemicals are believed to have good investment potentials in ten years' time, while foods, cosmetics and health-promoting agents were assigned lower potential returns. All of the biorefinery product groups were assigned higher potential returns on investment over ten years than five, with the exception of pulp and paper, bioenergy assortments and cosmetics. In the case of pulp and paper, the potential returns in ten years were considered to be lower than those that could be achieved in five. The respondents predict a promising future for biorefinery products, especially as substitutes or complements for oil-based products. However, they also predict that problems are likely to occur due to the lack of suitable raw materials, in keeping with the results of Conrad *et al.* (2011) and Näyhä & Pesonen (2012). Accordingly, 55% of the respondents believe that the value of wood biomass will increase strongly over the next ten years. However, 41% believe it will increase only marginally.

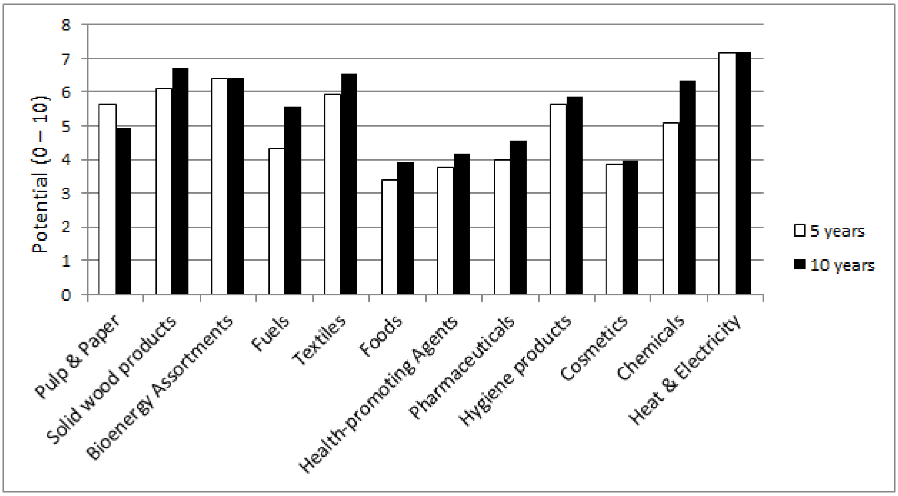

Figure 9. Mean investment potentials of wood-based biorefinery product groups within five and ten years, respectively, as listed (0 – 10) by the respondents. 0 = no potential; 10 = great potential (Study V).

Stemwood is assumed to have the greatest level of underutilized potential, followed by branches, stumps and bark (Fig. 10). Needles, roots and knots were given lower values. These answers clearly reflect the structure of the forest industry and its current logistical capabilities.

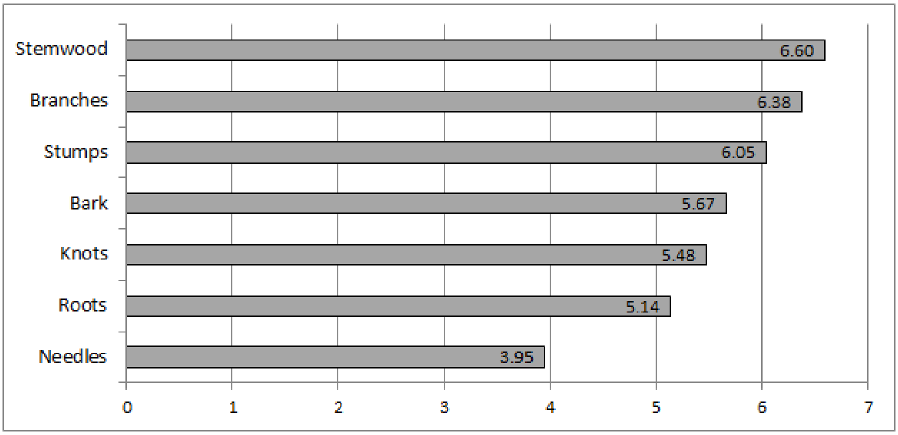

Figure 10. Mean values of unutilized potential in different parts of trees, as listed 0 – 10 by the respondents. 0 = no potential; 10 = great potential (Study V).

The respondents were also asked to project the market potentials for one or more products of their own choosing. Products from five categories were chosen: transportation fuels, special forms of cellulose, materials and plastics, solid fuels, and specialty chemicals. Solid fuels were considered to have the greatest potential, but the majority of the products mentioned by the respondents could be categorized as specialty chemicals. Thus, while the respondents had considered a wide range of wood-derived chemical products, they had the greatest confidence in more traditional solid wood products. There was a consensus that most of the new chemical products could be readily integrated into existing production chains. Biomaterials and clothing were judged to have bright futures, but there was considerable skepticism regarding the potential of wood-based ethanol because other more effective or environmentally friendly biofuels are being developed.

Most respondents stated that stemwood is required for the manufacture of their chosen product but did not mention any specific part of the stemwood. The respondents that identified specific chemicals as required inputs (45%) mentioned cellulose, hemicelluloses, lignin and various extractives (fatty acids, resin acids and phytosterols). Overall, it was generally agreed that it would be necessary to separate the main constituents of the harvested wood to enable the manufacture of the chosen products but the preliminary isolation of specific chemicals would not be required (although it might become desirable in the future).

The respondents identified the main opportunities and threats to wood-based biorefineries:

Opportunities

1. Increased demand for green products
2. Higher oil and energy prices
3. Increased use of policy instruments
4. Accessible raw material
5. Research and technical progress
6. Rural economic growth

Threats

1. High investment costs
2. An uncertain political environment
3. Competition for raw materials
4. Ecological risks

In terms of the mass or volume of raw material required relative to the amount of product generated, wood for heating, construction wood and pellets had values of almost 1:1, whereas tall oil and tall oil diesel had much higher values (Fig. 11). For fatty acid extraction to be economically viable, the value added by extracting the acids must exceed that obtainable by simply burning them to recover their stored energy.

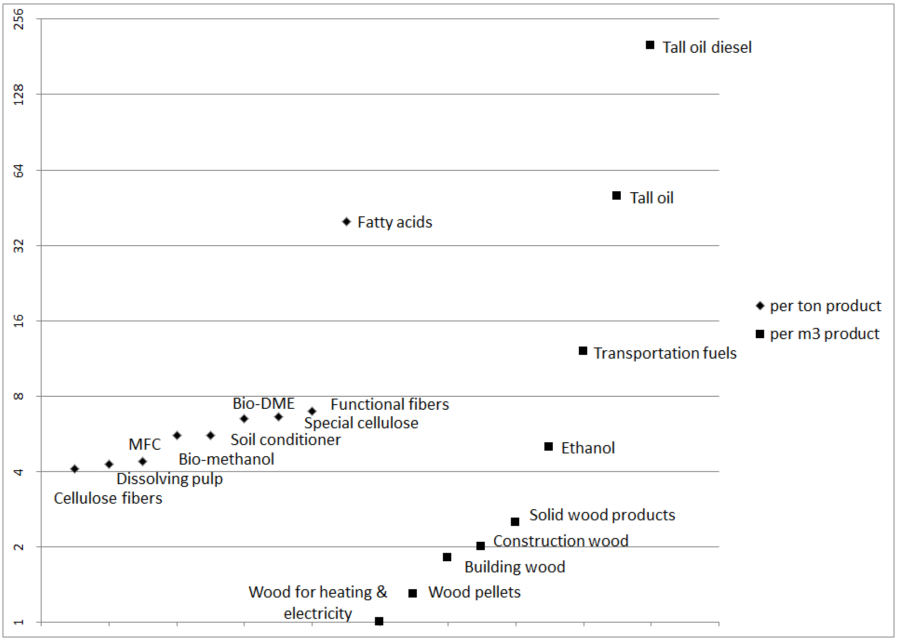

Figure 11. Quantity (m^3) of raw material (i.e. wood) required for the production of 1 ton or 1 m^3 of the various chosen products (Study V). Note the logarithmic scale on the vertical axis.

The respondents stated that the prices of their chosen products would be marginally (52%) or very strongly (39%) affected by electricity prices. Higher energy prices can increase production costs but can also make bio-based products more competitive with oil products. Combined bioenergy-biorefinery plants may become more profitable when energy prices are high, so the issue is rather complex. Several respondents pointed out that the unit price for woody biomass would rise if the energy price rises, because this would provide a greater incentive for using wood to generate bioenergy. In contrast to popular opinion, the supply of raw material is as sensitive to rising energy prices as are the various manufacturing processes, if not more so. The respondents agreed that higher oil prices would benefit their bio-based products and that the production of wood is less energy-dependent than that of other building materials such as concrete. In addition, wood-based materials are better thermal insulators than their alternatives, which tends to reduce energy costs as they become more widely used.

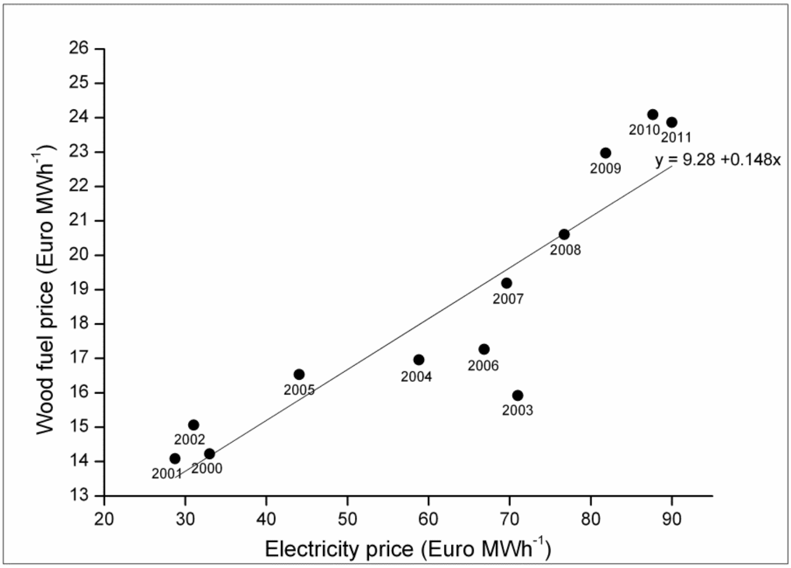

Figure 12. The relationship between Swedish electricity prices and wood fuel prices for Swedish district heating and industry between 2000 and 2011 (Study V).

Swedish electricity prices were shown to correlate with the price of wood fuel between 2000 and 2011 (Pearson correlation coefficient = 0.91; Fig. 12). Electricity prices may thus be an important driver of the total value of biomass, maybe via the oil price since bioenergy can replace oil in CHPs. It has to be emphasized that the reverse causal connection is not likely to exist because around 75% of the electricity in the Nordic countries is traded on the Nord Pool Elspot market, where its price is determined by the balance between supply and demand. Key factors include macroeconomic variables, cold weather and the capacity of the available hydro- and nuclear plants (Swedish Energy Agency, 2012). Biomass-fired power plants have relatively little impact on the electricity price even though it strongly affects their profitability.

Higher electricity prices are expected to encourage energy efficiency and a general move towards renewable-based technologies (International Energy Agency, 2008), so rising energy prices may provide strong incentives for investment into wood-based biorefineries. The electricity prices within OECD countries are expected to increase by 15% between 2011 and 2035 (International Energy Agency, 2012). If the relationship identified in our work continues to hold, Swedish wood fuel prices will therefore increase by around 10% during this period. The price of electricity may therefore play a decisive role in determining whether forest biomass is mainly used to generate bioenergy or as a source of valuable chemical compounds in the future. The capacity to pay for processed wood chips has already reached the level of pulpwood (Fig. 13).

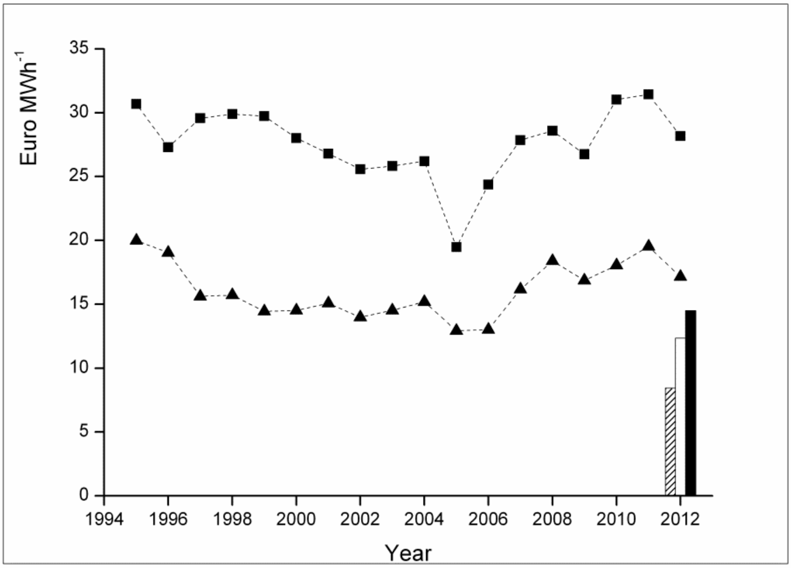

Figure 13. Yearly price change of pulpwood (triangles) and timber (squares) in Sweden between 1995 and 2012 (Study V). The vertical bars indicate the current prices of woodchips (black = energy wood; white = whole tree; filled = tops, branches and needles.

However, the traditional pulpwood and saw timber industries are needed to maintain the profitability of the forest industry sector and thereby enable the cost-effective removal of biomass assortments. This study shows that several products from forest biomass have considerable potential for future exploitation, especially heat/bioenergy and electricity, solid wood products and textiles. Experts with various backgrounds have great confidence in traditional lignocellulosic products and see biorefinery products such as textiles and specialty chemicals as having the potential for further development. Higher electricity prices will likely force wood fuel prices upwards and hence also the value of tree biomass. Political incentives might be required to spur further investment into biorefineries.

3.2 Sustainable forest management in a biobased economy

> "Management and use of forests and forest land should be performed in such a way that biodiversity, productivity, regrowing capacity and vitality are preserved. Management should make sure forests are able – now and in the future – to fulfill important environmental, economical and social functions on local, national as well as global levels without harming other ecosystems."
>
> (Swedish Forest Agency, 2013)

Sustainable forest management entails many things including both ensuring the preservation of forests and enabling their exploitation, as indicated by the above quote from the Swedish Forest Agency. The Swedish Environmental Protection Agency has drawn up 16 environmental quality objectives that describe the quality and state in which the environment must be maintained to establish long-term sustainability (Swedish Environmental Protection Agency, 2013). Almost all of these objectives relate to either silviculture, or reducing the use of oil-based products and other chemicals in favour of bio-based products and energy. The goals are referred to by the following names: Reduced Climate Impact, Clean Air, Natural Acidification Only, A Non-Toxic Environment, A Protective Ozone Layer, Zero Eutrophication, Flourishing Lakes and Streams, Good-Quality Groundwater, Flourishing Coastal Areas and Archipelagos, Thriving Wetlands, Sustainable Forests, A Good Built Environment and A Rich Diversity of Plant and Animal Life. In some cases, one goal may contradict another, and working towards them all in a cohesive way will not be straightforward. It will probably therefore be necessary to introduce greater diversity in the way forests are managed in order to fulfil all of the goals. Some forests will continue to be managed according to traditional European silvicultural principles while others will be managed according to the principles of continuous cover forestry, the requirements of nature conservation, to promote community or social forestry, or to enable shorter rotation forestry for biomass production and the manufacture of bio-based products. The development of optimized forest management regimes for bioenergy production and the supply of biorefineries remains an important objective, not least because of the positive impact such regimes could have in terms of mitigating climate change.

Trees grow rather slowly under Nordic conditions. By cultivating lodgepole pine in the way it grows naturally, i.e. from seeds in dense stands with rapid juvenile development, we can exploit its ability to rapidly accumulate biomass under harsh conditions. The extractives in its wood, which have traditionally been regarded as problematic by-products in the pulping industry, can instead become important sources of added value. For this to happen, cooperation between forest industries, chemical industries, policy makers, economists and other stakeholders will be required. As stated in Paper IV, the forest industries' production processes will require several adaptations to facilitate the large-scale production of biomass production for biorefineries from Fennoscandian conifer stands. These include:

1. An increased use of silvicultural tools such as the direct seeding of dense stands in conjunction with one or several biomass harvests instead of traditional pre-commercial and commercial thinnings.

2. The determination of reference chemical profiles for various tree fractions at different stand ages and for different tree sizes.

3. The development of sorting systems for the isolation of assortments with certain chemical contents.

4. The introduction of processes for extracting valuable compounds, either separately or using methods that can be integrated with existing activities such as pulping.

5. The introduction of methods for the further separation and exploitation of biomass-derived feedstocks at chemical plants.

If these changes are made, it may be possible to manage stands "by fraction", meaning that each tree fraction would be used in an appropriate way as the stand grows rather than regarding the stemwood as the main product and the other assortments as by-products. Determining the properties of the harvested biomass in advance will facilitate its use in the production of valuable products at biorefineries. The adoption of integrated planning processes based on a bottom-up approach in forest management is expected to facilitate forest planning of this sort and also to encourage local empowerment within forest organizations (Nilsson, 2013). This in turn will promote the use of local knowledge and improve the company’s competitive position by enabling local forest managers to select suitable stands for biomass harvesting.

3.2.1 Guidelines for lodgepole pine biomass production

The data suggest that lodgepole pine stands in Fennoscandia could readily be managed in ways that would yield good biomass production at low cost (Study I & II). To optimize the harvesting of lodgepole pine, it will be necessary to determine how its chemical content changes over time, how the distribution of biomass between tree fractions varies as the trees age, and how stand stem density affects biomass allocation as well as the properties and chemical composition of the wood. Genetic engineering and breeding programs can also be used to manipulate tree properties, but approaches of this sort are beyond the scope of this thesis. Silviculture is largely about making choices concerning matters such as which tree species to plant, what management practices to use, and what harvesting technologies and priorities should be adopted. There are a few important variables that must be considered when managing lodgepole pine for biomass production:

1. Dense versus sparse stands

Young dense lodgepole pine stands (3000-4500 stems per hectare) produce more biomass than corresponding sparse stands (Study I & II), with only slight reductions in stem diameter (Study I). It may therefore be optimal to aim for a stem density of about 4000 stems per hectare for biomass production. Pine is traditionally grown at approx. 2000 stems per hectare in order to allow for increases in stem diameter and to reduce self-thinning (Pettersson *et al.*, 2012). This method is appropriate if conventional timber production is the main goal. However, new approaches are evolving in which a post-thinning density of around 3000 stems per hectare is targeted in order to exploit the high biomass production of lodgepole pine (Normark, 2011; Liziniewicz *et al.*, 2012). The benefits of thinning in Scots pine stands has also been questioned: Nilsson, *et al.* (2010) evaluated a range of thinning treatments in Scots pine stands and showed that all of them reduced the total gross stem volume production compared to that for unthinned control plots. Conversely, in Norway spruce stands, only very heavy thinning produced such comparatively poor results. Lodgepole pine accumulates biomass even more rapidly than Scots pine and thus may benefit even more from being grown in dense stands.

2. Young versus old stands

The chemical composition of lodgepole pine wood varies with age and diameter (Koch, 1996). Heartwood formation begins once the trees reach around 20 years of age (Gjerdrum, 2003). All of the 30-year-old trees examined in Study IV contained heartwood at the base of their stems, and heartwood was also starting to form at the tops of the larger trees. Heartwood contains more extractives than sapwood (Studies III & IV). The biomass partitioning of young and old trees also differs, with more being allocated to stems and needles in older and larger trees, and to branches in younger trees (Vanninen, 2004). Young lodgepole pine grows very rapidly (Norgren, 1996; Elfving *et al.*, 2001), and there are only few older lodgepole pine stands in Fennoscandia that can be studied (two that were planted in 1928 were sampled in Study III). The stemwood of the 30-year-old trees extracted in Study IV had a crude extractive content of 0.3-3.5% (d.w), whereas the 57-82 year old trees examined in Study III had fatty- and resin acid contents of 0.2-2.6% (d.w.). Because the latter study focused on fats and resins exclusively, it is reasonable to assume that the actual extractive content of the older trees is substantially higher than the quoted value. It is also important to consider the balance between biomass growth, extractive content and alternative uses for wood. Older trees might produce timber with desirable properties (Andersson, 2013), meaning that their stemwood is best used to produce massive wood products. According to the results of Studies I, II and IV, lodgepole pine stands aged between 15-40 years are probably the best options for whole-tree biomass harvesting for biorefineries.

3. Direct seeding versus planting

Direct seeding facilitates the establishment of dense, stable lodgepole pine stands (Rosvall, 1994; Wennström *et al.*, 1999). Direct seeding is particularly useful for lodgepole pine cultivation because of this species' rapid initial growth, which reduces the period during which its seedlings are vulnerable to browsing and competition from grasses, etc. (Dermer, 2007). Planting is the predominant regeneration method used in Fennoscandia because it is considered reliable, the planted seedlings have considerable advantages over competing vegetation, and the seedlings can be planted in precisely defined locations (Hallsby, 2013). However, planting is still mainly done by hand, which is expensive and time-consuming. Direct seeding can be done mechanically and is usually significantly cheaper than planting: its only major costs are those associated with soil preparation and buying the seeds (Wennström, 2001; Bergsten & Sahlén, 2008). Microsite preparation using a mixture of orchard and stand seeds can be a particularly effective technique because stand seeds increase stand density while orchard seeds yield more rapid growth (Wennström, 2001).

4. Lodgepole pine versus Scots pine

Scots pine has the advantage of being a native pine species in Fennoscandia. However, Scots pine seedlings have significant moose browsing problems and produce around 36% less stemwood than an equivalent lodgepole pine stand over a given period of time (Elfving *et al.*, 2001). Scots pine might produce somewhat stronger wood (Andersson, 2013), while lodgepole pine is a better option for fatty- and resin acid extraction because of its more even distribution of different extractives across its various types of wood (Study III) and its more rapid growth. Lodgepole pine also allocates more biomass to branches and needles than Scots pine (Norgren, 1996).

5. Added value of chemical extraction compared to traditional products

Lodgepole pine was primarily introduced to Europe as a pulpwood species (Hagner, 1983; Elfving *et al.*, 2001). The value of wood biomass is considered to increase over the next ten years, and biorefinery products are believed to have a promising future (Study V). A typical 30-year-old lodgepole pine stand in the lower parts of northern Sweden will produce 93 tons of dry weight biomass per hectare, of which 3 tons will be extractives (3.2%; Study IV). A corresponding stand in the Scandinavian mountains will produce 52 tons of d.w. biomass per hectare, of which 2.2 tons will be extractives (4.3%; Study IV). Of course, the cellulose, hemicelluloses and lignin in the wood are also useful in biorefineries. Products such as tall oil biodiesel, resins, natural waxes and phytosterols can be produced from lodgepole pine (Studies III and IV). The exact added value that can be obtained by the chemical extraction of biomass is uncertain and heavily dependent on the prices of different wood assortments, which are in turn sensitive to energy prices (Study V). The costs of whole-tree harvesting are particularly uncertain (Kero, 2007), which brings us to the next subject.

6. New harvest technologies compared to traditional technologies

By corridor thinning of 70% of the total stand area at two different corridor widths, it was possible to collect 27-32 tons of biomass (or 38-45 m^3 stemwood per hectare) from a dense 20-year-old lodgepole pine stand in which the mean diameter of the 1000 largest trees per hectare was around 8 cm (Study II). This gives the forest owner a source of income at an early stage in the rotation period and avoids the need for a costly pre-commercial thinning. The health, growth and yield of the remaining trees left behind after corridor thinning will have to be investigated in more detail over longer periods of time, but these results show that it is possible to produce and harvest large amounts of biomass and stem volume early in the rotation period without necessarily preventing pulp and timber production using the remaining trees. These harvesting technologies are under ongoing development; for an overview of recent work in this area, see the publications by Bergström *et al.* (2010) and Karlsson (2013).

7. When should biomass be harvested from lodgepole pine stands?

As discussed in Paper II, several different management regimes can be used in lodgepole pine stands. It may not be optimal to use aggressive thinning and focus exclusively on timber trees because this approach sacrifices a lot of potential income and causes a lot of damage to the remaining trees (around 10% of the trees showed signs of damage). While, this regime did produce somewhat greater stem diameters than the alternatives, it was inferior to the combined regime (which is intended to favor both biomass production and the growth of timber trees) in terms of stem volume development. Two viable options for harvesting lodgepole pine stands were identified. The first involves an early final felling at around 30 years of age (c.f. Study I), to produce around 100 tons of dry weight biomass per hectare. The second entails corridor thinning at age 20 to produce ca. 30 tons of biomass, with the remaining trees being allowed to continue growing for use in pulpwood or timber production (Fig. 14). Current Swedish forestry laws permit the total biomass harvesting of 30-year-old lodgepole pine stands (Skogsvårdslag 1979:429) as discussed in Study II, although this was not appreciated by the authors when writing Study I. Whole-tree harvesting presents a risk of removing nutrients and acid-buffering capacity from the soil (Swedish Forest Agency, 2013). If large amounts of needles and branches are removed, nutrients should be returned, e.g. by ash recycling or fertilization, and some quantity of tree residues should be left in the forest to provide nutrients for decomposers and other organisms (Egnell *et al.*, 1998; Swedish Forest Agency, 2008).

3.2.2 Possible regime routes for biomass outtakes to biorefineries

There are a number of potential ways of managing and harvesting lodgepole pine stands, and of uses for the resulting assortments and products (Fig. 14). Instead of the traditional routine of planting, pre-commercial and commercial thinnings, and final harvesting (illustrated on the left hand side of Fig. 14), it may be profitable to regenerate stands by means of cost-effective direct seeding and then perform biomass harvest(s) to supply biorefineries (illustrated on the right hand side of Fig. 14). The remaining trees could then be managed according to the principles of traditional silviculture and grown for the purposes of pulpwood and timber production (Study II).

After direct seeding, it may be desirable to perform a cost-effective early PCT (at a tree height of ca. 1 m) in order to control the stand's growth and facilitate future crop tree harvesting (Fig. 14). However, high stand stem densities are required in order to sustain rapid biomass production during the early stages of the rotation period (c.f. Study I & II). It may be appropriate to perform an initial biomass harvest when the stand is 15-25 years old (Study II) in cases where there are no plans to perform complete biomass harvesting once the stand reaches 30-40 years of age (i.e. in cases where short rotation forestry is not intended; Study I). In young stands, it may be advantageous to harvest whole trees to supply biorefineries and to use the leftover material for bioenergy production (c.f. Studies IV & V). Heartwood does not start to develop until the stand is between 20 years of age (at the base) and 30 years of age (at the top; see Fig 8), so the stemwood of young trees has a comparatively low extractive content. Nevertheless, good yields of extractives can be obtained from their needles, bark and branches. Needles regenerate throughout the rotation period, but their composition seems to change when heartwood forms at the top of the tree (c.f. Study IV). Needles can provide waxes, fatty alcohols and phytosterols for use in cosmetics and bioactive food additives, while bark can provide terpenes and ketones for flavoring agents, fragrances, pharmaceuticals or polymer precursors (Study IV). Lignocellulose can be used to produce useful goods such as plastics, clothing and dispersing agents.

Biomass for biorefineries represents a new assortment that can be produced in both old and new stands and which complements timber, pulpwood and fuelwood (Fig. 14). Fats and resins can be extracted from trees of intermediate size prior to pulping (Study III), and further useful compounds can be obtained from needles and bark. Cones start to develop after a few decades, and can be separated to extract their aromatics. However, it is important to recall that the profitability of these techniques remains to be determined, and that they may require the development of new methods of separation. By adopting silvicultural regimes that involve appropriate methods of regeneration and carefully chosen stand stem densities and fertilization schemes, it is possible to influence the trees' allocation of biomass to increase the yields of the most valuable fractions.

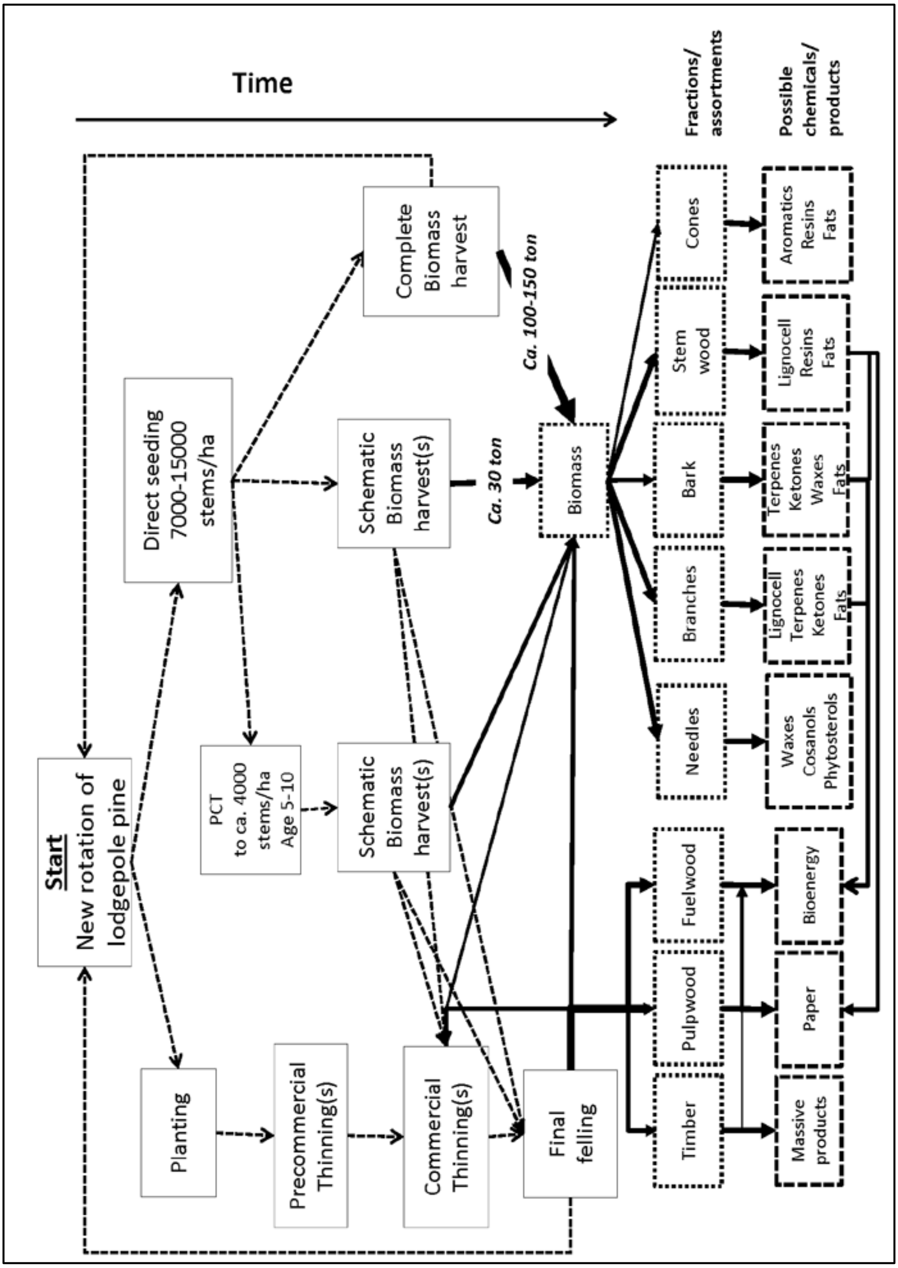

Figure 14. Lodgepole pine management regimes (dotted arrows) and specific silvicultural measures (solid boxes) with corresponding product streams (solid arrows with different weights), assortments and products (dotted boxes). The traditional regime involving planting is shown on the left, while direct seeding regimes with periodic biomass harvests are shown on the right.

4 Conclusions and Future Perspectives

- There is considerable scope for the further development of regimes for short(er) rotation lodgepole pine forestry that would yield substantial quantities of inexpensive biomass for biorefineries within a few decades.

- Around 200 m^3 of stem wood per hectare, or 100 tons of dry weight biomass, can be obtained within 30 years of direct seeding with current management methods. Stemwood accounts for approximately 65 tons of this, along with 8 tons of bark, 12 tons of needles and 12 tons of branches. Higher stand stem densities (≥3000 stems/ha) yield more biomass (up to 300 m^3) with only slight reductions in diameter at breast height.

- By using higher stem densities (ca. 4000 st/ha), more biomass and higher stem volumes can be obtained for early harvesting without sacrificing the diameter of future crop trees. Stands can be cost-effectively managed by direct seeding and schematic harvest. The harvesting of large quantities of biomass at an early stage in the rotation period does not preclude the subsequent conversion of the stand to focus on pulp and timber production.

- The amount of heartwood and the partitioning of biomass between different tree fractions are the most important predictive factors to consider when estimating the extractive content of pines.

- Lodgepole pine is a good source of extractable fatty- and resin acids because the extractives are fairly evenly distributed between the different wood types and because of its high biomass production. In a mature lodgepole pine stand with 150 tons of d.w.

biomass per hectare, ca. 150 kg of fatty acids and 1 ton of resins can be extracted from the stemwood.

- In 30-year-old direct seeded lodgepole pine stands, the bark was found to have the highest extractive content (16%) and the stemwood the lowest (1%). The extractive composition of the needles differed substantially from that of all other tree fractions. A large variety of extractives could be identified in lodgepole pine, including various fatty- and resin acids, waxes and aromatics.

- Using current management regimes, 2-3 tons of crude extractives per hectare can be obtained from 30-year-old lodgepole pine stands. The precise extractive yield depends on the site fertility, tree partitioning and tree size. The extractives can be used to produce products such as biodiesel, glue, bioactive food additives, cosmetics, and polymer precursors.

- The value of woody biomass is believed to increase over the next ten years, and bioenergy assortments and textiles are considered to have the highest investment potentials. A wide range of materials, fuels and specialty chemicals can be produced from tree biomass.

- The prices of electricity and wood fuel were found to be strongly correlated. Electricity prices within OECD countries are expected to increase by 15% between 2011 and 2035, with wood fuel prices increasing by roughly 10% during the same period.

- Large-scale commercial biorefinery production seems to be viable in the near future, but it may be necessary to define new wood assortments that specify both the fibre properties and the chemical properties (e.g. extractive content) of the wood. Political support may be required to promote further investment into lignocellulosic biorefineries. Biomass can be used to generate energy and as a raw material to supply biorefineries, so the added value provided by the biorefineries may become an essential contributor to its economic worth.

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Acknowledgements

When I was six years old, in 1988, I attended my father Lennart's dissertation at the Institute of Technology, University of Linköping. I could never imagine then, that I would be in the position of my own dissertation 25 years later. You have been a great inspiration to me, Dad, and you have also helped me practically through this work by being my field assistant in the Härjedalen and Hälsingland forests during many summer weeks, accompanied by thousands of mosquitos! Similarly, I want to thank my wise and dear mum Alice for inexhaustible support and interest in my work!

Two of the most important persons during my PhD studies have been my two supervisors. My principal supervisor Professor Urban Bergsten has contributed with large knowledge and long experience in silviculture, thinking beyond the current boundaries of forestry and always offering quick response to emails and manuscripts. My assistant supervisor Dr Mehrdad Arshadi, with deep knowledge in chemistry and biorefining, has been a great support and a swift soul to juggle ideas with. You complement each other very well Urban and Mehrdad, and I hope to be able to cooperate with you sometime in the future too. Thank you!

Some other colleagues have also been important to facilitate this thesis. Lars Karlsson started his PhD studies at the same time as me and will finish at the same time too! You have been my foremost coffee break- and lunch companion in Umeå, and have been my sounding board many times. Kristina Ulvcrona, it is always enlightening and refreshing to work with you! The lunch breaks in the Bjärkliden forest with you and your two dogs are unforgettable. Many thanks to my other article co-authors: Leif Mattsson, Paul Geladi, Andy Hunt, Rob McElroy and Tom Attard. Thanks also to Björn Elfving, Tommy Mörling, Carina Jonsson, Sören Holm, Holmen Skog AB and SCA for assistance and interest in my work. The Kempe foundation and the Brattås

foundation are thanked for supporting this project financially, while Sees Editing Ltd and Arkitektkopia AB in Umeå are thanked for editing and printing services.

My regards and wishes for many new interesting research results and fruitful cooperations, go to all the colleagues and friends at the three different units where I have executed my work: the brand new Department of Forest Biomaterials and Technology at SLU in Umeå, where I spent the last year of my PhD; the Department of Forest Ecology and Management also at SLU in Umeå, where I started my PhD studies in 2009 and belonged until 2012, and the Green Chemistry Centre of Excellence at the University of York, UK, where I performed the chemical analyses 2011-2012 and also learned a lot both about green extraction technologies and the British way of life.

I have performed a great deal of this work away from Umeå, as I live in Jämtland, in the north-western part of Sweden. There have also been numerous travels to England, France, Canada, Ethiopia and Spain. My dear love Jonte has supported me through these years and convinced me that I will make it! Thank you for being such a kind, loving, fun and competent man! Our building projects have offered me natural breaks from the academic world, and I look forward to different kinds of projects to come! Doris the cat has accompanied me at my home office during many days, and added a feeling of coziness to my work. She is actually lying on a sheepskin by my feet right now. Finally, I want to sincerely thank my brother Erik, my sister Anna and her husband Cory, my mother-in-law Lotten and the rest of my family, friends and relatives for support and love.