Afforestation of Former Arable Land in North-Western Europe

Nitrate Leaching, Carbon Sequestration and Water Recharge

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Photo front: Afforested oak stand at Vestskoven, Denmark (Lars Vesterdal, Jan 1999)

Abstract

Rosenqvist, L. 2007. Afforestation of arable land in north-western Europe – nitrate leaching, carbon sequestration and water recharge, Doctor's dissertation. ISSN: 1652-6880, ISBN 978-91-576-7301-5.

Afforestation of former arable land may serve many purposes and provide many benefits, including carbon sequestration, reduced nitrate pollution of water bodies, restoration of biodiversity and improved recreational possibilities. However, afforestation may also have adverse environmental effects such as reduction of water recharge to groundwater and surface water. This study evaluates the implications of a land use change from agriculture to forestry for N, C and water cycles, based on six afforestation chronosequences (species and age) in Sweden, Denmark and the Netherlands. Results showed that estimated losses by nitrate leaching varied considerably among these six chronosequences, but were at least 3-5 times lower than the reported nitrate leaching from arable land in the three countries. Compared with nutrient-poor sandy soils, nutrient-rich clayey soils appeared more vulnerable to disturbance of the N cycle and to increased N deposition, possibly leading to N saturation and enhanced nitrate leaching. Over approximately 35 years following afforestation, nitrate leaching from the root zone was generally higher below common oak than below Norway spruce. Simulated water recharge revealed a general pattern of decline during the first 30-40 years, followed by a recovery in older stands. Oak stands older than 20 years showed a 60-90 mm higher water recharge than adjacent stands of Norway spruce of similar age. The aboveground and belowground biomass accounted for about two thirds of the increase in ecosystem C, and the remaining one third was sequestered in the O horizon and the Ap horizon. The effect of time since afforestation on soil solution DOC (and DON) was partly masked by spatial variability in soil properties that influence DOM adsorption in the mineral soil. Concentrations of DOC in the A horizon at the sites were primarily related to the oxalate-extractable amounts of Al and Fe in the same horizon. It is evident that planning and planting new forests on arable land in an environmentally sound way requires a holistic and long-term perspective on the subsequent changes.

Keywords: afforestation, arable land, chronosequence, water recharge, interception, nitrate leaching, carbon sequestration, dissolved organic carbon, dissolved organic nitrogen, Norway spruce, common oak

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Appendix

Papers I-V

The present thesis is based on the following papers, which are referred to in the text by their Roman numerals:

I. Rosenqvist L., Hansen K., Vesterdal L., Denier van der Gon H., van der Salm C., Bleeker A. & Johansson M.-B. 2007. Nitrogen deposition and nitrate leaching following afforestation: experiences from oak and Norway spruce chronosequences in Denmark, Sweden and the Netherlands. In: Heil G., Muys B. & Hansen K. (Eds.). Environmental Effects of Afforestation in North-Western Europe - From Field Observations to Decision Support. Springer, *Plant and vegetation*, Vol 1. ISBN-10: 1-4020-4567-0, ISBN-13: 978-1-4020-4567-7, 79-108.

II. Hansen, K., Rosenqvist, L., Vesterdal, L. & Gundersen, P. 2007. Nitrate leaching from three afforestation chronosequences on former arable land in Denmark. *Global change biology* (Submitted).

III. Rosenqvist, L., Hansen, K., Vesterdal, L. & van der Salm, C. 2007. Water balance in chronosequences of forest stands (ages and species) following afforestation of arable land in Denmark and southern Sweden. (Manuscript).

IV. Vesterdal L., Rosenqvist L., van der Salm C., Hansen K., Groenenberg B.-J. & Johansson M.-B. 2007. Carbon sequestration in soil and biomass following afforestation: experiences from oak and Norway spruce chronosequences in Denmark, Sweden and the Netherlands. In: Heil G., Muys B. & Hansen K. (Eds.). Environmental Effects of Afforestation in North-Western Europe - From Field Observations to Decision Support. Springer, *Plant and vegetation*, Vol 1. ISBN-10: 1-4020-4567-0, ISBN-13: 978-1-4020-4567-7, 19-52.

V. Rosenqvist, L., Berggren Kleja, D. & Johansson, M.-B. (2007) Concentrations and fluxes of dissolved organic carbon and nitrogen in a Norway spruce chronosequence on former arable land in Sweden. (Manuscript).

Introduction

General background

In recent decades, afforestation of modern agricultural land has been widespread in western Europe. Apart from improving the economic structure of agricultural systems, the incentives for this change in land use include protection of groundwater reserves, reduction of atmospheric carbon dioxide (CO_2) by increasing the stock of carbon (C) in the growing forest, restoration of biodiversity and a demand for more recreational areas (Heil, Muys & Hansen, 2007). However, afforestation may also lead to adverse negative environmental effects such as reduction of water recharge to groundwater and surface waters (Bosch & Hewlett, 1982; Bastrup-Birk & Gundersen, 2004; Farley, Jobbágy & Jackson, 2005), higher solubility of heavy metals (Andersen *et al.*, 2002), soil acidification (Alriksson & Olsson, 1995; Ritter, Vesterdal & Gundersen, 2003) and nutrient deficiency (Jug *et al.*, 1999).

Today, abandonment of marginal farmland is making large areas available for alternative land uses in EU countries. Much of this land is suitable for afforestation. Due to European Common Agricultural Policy (CAP) reforms, afforestation of arable land is expected to increase in Europe in the coming decades (Rabbinge & van Diepen, 2000). However, many EU countries have been planting forest on former arable land for quite a long time. In Sweden, the agricultural area has decreased by more than 900,000 ha during the past half century (Anon., 2004), and a substantial part of this abandoned cultivated land has become forest, either through plantation or by natural succession. Since 1960, more than 530,000 ha of farmland have been converted to forest as part of afforestation programmes funded by the Swedish government (Johansson, 1995b). The most commonly planted tree species has been Norway spruce (*Picea abies* (L.) Karst.), but since the 1990s, the planting of deciduous species has been encouraged by government subsidies (Helles & Lindahl, 1996). In Denmark, a government resolution from 1989 calls for a doubling of the forested area within the next 100 years. This resolution aims at a 400,000 ha increase in the forested area. Today, 28,000 ha of forest have been planted since 1990 and the number of hectares of new forest planted on arable land is increasing year by year (Figure 1).

This thesis focuses on various environmental aspects of farmland afforestation, namely its influence on nitrogen (N) deposition (Paper I), nitrate leaching (Papers I and II), the water balance (Paper III), C sequestration in biomass and soils (Paper IV) and fluxes and retention of dissolved organic matter (Paper V). To make qualified decisions on the afforestation of former agricultural areas in Europe from an environmental viewpoint, more knowledge of these aspects is needed. The majority of this work was carried out within the framework of the EU-project AFFOREST (Heil, Muys & Hansen, 2007; http://www.sl.kvl.dk/afforest). Data were also obtained from an earlier EU-project, MEMO.

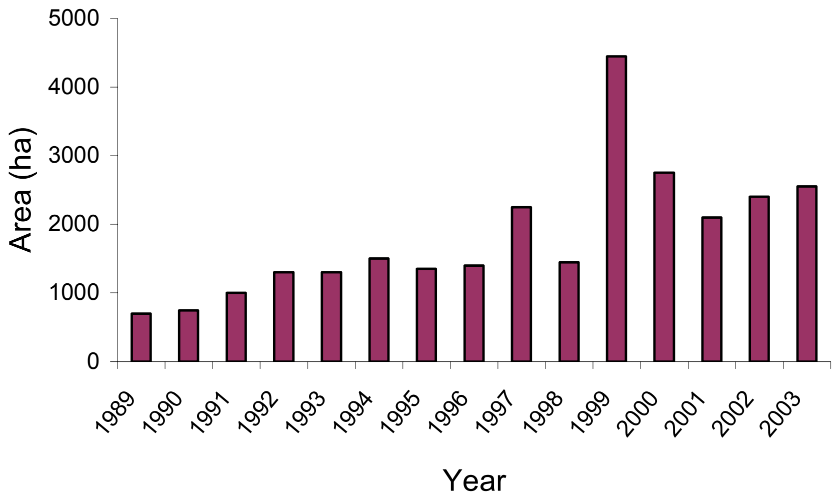

Figure 1. Number of hectares afforested in Denmark since 1989. Afforestation is mainly performed on former arable land. Anon. (2006).

Implications of farmland afforestation for water quality and quantity

Changes in water use and water recharge after afforestation

It is widely accepted that forests use more water than shorter types of vegetation (Bosch & Hewlett, 1982; Calder, 1990; Kirby, Newson & Gilman, 1991; Whitehead & Robinson, 1993). In wet temperate climates, the higher water use by trees is mainly due to the interception of rainwater by their aerodynamically rougher canopies compared with shorter vegetation (Calder, 1990; Heal *et al*., 2004). The impact of afforestation or deforestation on water recharge has been addressed in a number of papers (Bosch & Hewlett, 1982; Sahin & Hall, 1996; Scott *et al*., 2000; Bastrup-Birk & Gundersen, 2004; Brown *et al*., 2005; Farley *et al*., 2005). These authors report that large-scale conversion from low vegetation (such as arable crops or grasses) to mature forest reduces water recharge by 25-45% mainly caused by increased interception. The reduction in water recharge differs strongly from site to site due to differences in climate, site characteristics and field layer vegetation. The effect of a vegetation change on water recharge is larger when coniferous trees are planted than when deciduous forest is raised (Sahin & Hall, 1996; Bastrup-Birk & Gundersen, 2004). The relationship between the increase in forest canopy and the water recharge further depends on the age of the forest. The maximum run-off reduction following grassland afforestation has been observed 10-20 years after planting (Farley, Jobbágy & Jackson, 2005).

Nitrate leaching from afforested farmland

In Europe, intensified management of farmland and long-term fertilisation has altered N budgets substantially during the past 50 years. Modern arable soils are generally characterised by high soil N contents, low soil C/N ratios (around 10-15) and high mineralisation and nitrification rates. In regions dominated by agricultural activities, surface water and shallow groundwater often have nitrate concentrations exceeding the drinking water standard of 11.3 mg N l^{-1} (50 mg l^{-1} NO_3^-) (Egmont, Bresser & Bouwman, 2002). In contrast to agricultural systems, old existing forests are characterised by a more tight N cycle and the rate of net nitrification is also rather low. Although N deposition has increased considerably over the past 50 years in Europe (*e.g*. Gundersen, Schmidt & Raulund-Rasmussen, 2006), most forest waters have low nitrate concentrations compared with other land uses (*e.g*. Callesen *et al*., 1999; Thornton *et al*., 2000; Kristensen *et al*., 2004). Thus, afforestation of arable land is seen as a strategy to improve water quality (*e.g*. Iversen *et al*., 1998).

Leaching losses of N from ecosystems mainly occur as dissolved N in seepage water. Dissolved N may be in the form of nitrate, ammonium or dissolved organic nitrogen (DON). Nitrate is highly mobile in the soil profile and is easily lost from the system by leaching. Nitrate is therefore the most relevant N compound for water quality and is addressed as such in specific EC directives (91/676/EEC and 98/83/EEC). Ammonium adsorbs to the soil cation exchange complex and is less mobile in the soil profile. Ammonium usually contributes less than 5% to the total dissolved N concentration, except in extremely ammonium-rich soils (Diese, Matzner & Gundersen, 1998). In forest ecosystems in areas with a naturally low N input, DON is often found to be the dominant N form in soil solution (Andersen & Gundersen, 2000) and runoff (Hedin, Armesto & Johnson, 1995). However, concentrations of DON in seepage water under forests are observed to be low (Andersen & Gundersen, 2000).

A shift in land use from agriculture to forestry induces major changes in the N cycle, including inputs, internal cycling and losses. Nitrate leaching from forests is at risk when the ecosystem becomes saturated with N, *i.e*. when the availability of inorganic N exceeds the demand from plants and microorganisms (Aber *et al*., 1989; Gundersen, 1991). The termination of fertiliser use after afforestation may lead to substantially reduced nitrate leaching, as concluded from simulations of land use change in the Netherlands (Rijtema & de Vries, 1994) and from simulations of afforestation in a Danish catchment (Bastrup-Birk & Gundersen, 2004). At three sites in Denmark, concentrations of nitrate in seepage water 10 years after afforestation were considerably lower than those measured immediately after afforestation (Hansen & Vesterdal, 1999).

Nevertheless, nitrate leaching from forest plantations on former arable land may still be higher than that from old forests. In Denmark, higher concentrations of nitrate in soil water below the root zone (0.7-1.0 m depth) were observed in recently (<10 years) afforested arable land compared with old forest ecosystems (Callesen *et al*., 1999). Moreover, Jussy, Colin-Belgrand & Ranger (2000) found leaching of 22-39 kg ha^{-1} yr^{-1} from forests planted 20-60 years ago on previous arable soils. Compared with old forests, plantations on former arable land have higher N status as a legacy of former fertilisation, which supports continued high mineralisation and nitrification in the mineral soil (Jussy, 1998; Compton *et al*., 1998). Even former arable soils afforested 50-100 years ago have N-cycling characteristics differing significantly from those of nearby soils with unbroken forest cover (Compton *et al*., 1998; Jussy *et al*., 2002; Koerner *et al*., 1997). This land use legacy makes forests on former cultivated land susceptible to N saturation. Decisive factors for nitrate leaching under the new forests are the local N deposition level, the quality of the soil (N status) and the choice of tree species.

Carbon sequestration in the afforested ecosystem

Stock changes in tree biomass and soil

Concern over climate change from elevated CO_2 has increased interest in afforestation of arable land, since the Kyoto protocol (article 3.3.) allows for accounting of the biological sinks arising from afforestation as emission reductions. In a European perspective, afforestation may provide the greatest potential for C sequestration in agricultural soils (Powlson *et al*., 1998). The conversion from agricultural land to forest implies a shift from a shorter to a longer circulation time of C, as annual crops are replaced by long-lived perennial trees, and labile soil organic matter (SOM) pools are replaced by pools with longer residence times. The aggrading tree biomass is the dominant sink for the C sequestered following afforestation. On former farmland sites, the potential for rapid growth is great (Johansson, 1995b). Afforestation of agricultural land is also generally regarded as an effective means to increase soil C stocks. A review (Post & Kwon, 2000) found that the average rate of soil C sequestration (forest floor + mineral soil) was 0.34 Mg ha^{-1} yr^{-1} (range 0-3 Mg ha^{-1} yr^{-1}) across different climatic zones. An analysis of 29 studies on afforestation of arable cropland showed that C stocks increased by on average 18% over a variable number of years (Guo & Gifford, 2002). However, studies with unchanged total soil C stocks have also been published (*e.g*. Vesterdal, Ritter & Gundersen, 2002). A few studies have included concurrent measurements of soil and biomass C sequestration, and these studies suggest that about 25% of the total C could be sequestered in the soil (Richter *et al*., 1999; Hooker & Compton, 2003; Thuille & Schulze, 2006).

Whether soil is a source or a sink of C depends on the balance between litter production and decomposition of the litter produced. Decomposition rate of organic matter is expected to decrease following afforestation as a result of changed temperature and moisture conditions, the introduction of litter types of lower quality compared with agricultural soils, and the enhanced physical protection of SOM against microbial mineralisation resulting from cessation of frequent soil cultivation (Vesterdal, Ritter & Gundersen, 2002). Soil C stocks eventually reach saturation as a new putative equilibrium between organic matter gains and losses is attained. The storage capacity and rate of C sequestration depend on various factors such as the climate, soil type, tree species used for afforestation, current forestry practices, pre-afforestation management and land use history (Post & Kwon, 2000).

A change in land use from agriculture to forestry involves a reorganisation of soil organic matter. Afforestation with coniferous species in temperate climates generally results in substantial C sequestration in the forest floor (O horizon) gradually accumulating on top of the previously cultivated mineral soil (*e.g*. Post &

Kwon, 2000). Mineral soils, on the other hand, have been shown to gain C, experience no change in C, or even lose C following afforestation (Guo & Gifford, 2002; Vesterdal, Ritter & Gundersen, 2002). A commonly observed pattern is enhanced C content in the uppermost soil layer, and declining C content in the lower parts of the former plough layer (A-horizon) (*e.g*. Jug *et al*., 1999; Richter *et al*., 1999; Vesterdal, Ritter & Gundersen, 2002). Some long-term studies have also reported increasing C content in lower parts of the plough layer and below (Robertson & Vitousek, 1981; Leth & Breunig-Madsen, 1992). A recent review showed that C stores in the upper 30 cm of the mineral soil increased by on average 0.14 Mg ha^{-1} yr^{-1} following afforestation of arable land (Paul *et al*., 2002). Thus, the contribution of mineral soil to ecosystem C sequestration is uncertain, but small compared with C sequestration in tree biomass and forest floor.

Role of dissolved organic carbon

In forests, major sources of C in the mineral soil are root litter and dissolved organic carbon (DOC) leached from the O-horizon. Pedoturbation (mixing of surface SOM into deeper soils by soil fauna) may be important for C allocation during the first decades after afforestation. In temperate forest ecosystems, fluxes of DOC from the O horizon into the mineral soil have been estimated to be 100-400 kg C ha^{-1} yr^{-1} (Michalzik *et al*., 2001). Concentrations and fluxes of DOC in mineral subsoil horizons are usually very low, indicating strong retention in the mineral soil. According to current knowledge, physico-chemical processes (sorption/adsorption) rather than biological processes (mineralisation) are responsible for the efficient removal of DOC from solution in the mineral soil (Kalbitz *et al*., 2000; Kaiser & Guggenberger, 2000; Kalbitz *et al*., 2005). Despite being a small proportion of the total amount of soil organic carbon (SOC), DOC may contribute significantly to C storage in mineral soils. Typically, 40-370 kg ha^{-1} of DOC leaving the O horizon are retained annually in the mineral soil under forests (Currie *et al*., 1996; Guggenberger & Kaiser, 2003). Once retained in the mineral soil, the residence time for sorbed organic matter increases considerably compared with corresponding dissolved organic matter (DOM) in solution (Kalbitz *et al*., 2005).

A change in land use from agriculture to forestry affects many of the driving variables for soil processes and thereby also factors influencing DOM dynamics (Paper V). However, not many studies have directly described the effect of afforestation of arable land on soil solution DOC. Quideau & Bockheim (1996) found that pine afforestation of a former prairie site induced a significant increase in DOC concentration in the mineral soil. Likewise, the mineral soil of an afforested Sitka spruce stand exhibited significantly higher DOC concentrations than in an adjacent grassland site (Hughes, Reynolds & Roberts, 1990). Moreover, Karltun *et al*. (2005) studied the influence of afforestation on DOC dynamics in the Ap horizon by means of ^{14}C analysis and found that the proportion of DOC originating from the forest stand gradually increased with time after afforestation, but that a substantial part must still have originated from SOM formed before afforestation.

Objectives

The general objective of this work was to evaluate the implications of a land use change from agriculture to forestry for N, C and water cycles. The region under focus was north-western Europe, where field studies were carried out along forest chronosequences on former arable land in Denmark, Sweden and the Netherlands.

Specific objectives were to:

- Assess the effect of afforestation of arable land on N deposition (Paper I) and on nitrate leaching from the root zone (Papers I and II) in forests of changing age/height.
- Estimate the effect of afforestation on the water balance, especially on water recharge below the root zone, during stand development from young to mature forest (Paper III).
- Quantify C sequestration over time in living biomass and in the soil following afforestation of agricultural soils (Paper IV).
- Quantify the effect of forest conversion from agriculture on concentrations and fluxes of DOC and DON along an ecosystem profile, and assess soil factors controlling the retention of DOM in the mineral soil (Paper V).
- Evaluate the above-mentioned environmental performance parameters (the first three objectives) for two contrasting tree species and on contrasting soil types.

Materials and Methods

The study approach

We used a chronosequence approach to determine changes in various ecosystem components following afforestation of arable land. Here, the term chronosequence is used for a series of arable and afforested sites differing mainly in time since land use conversion. All sites within a chronosequence are located on similar soils within the same region experiencing the same climate and deposition regime. The use of a chronosequence approach implies that time is substituted for space.

Study sites

This thesis presents data from six chronosequences, of which two were differently aged oak stands (*Quercus robur* L.) and four were differently aged Norway spruce stands (*Picea abies* (L.) Karst.) (Figure 2). In Sweden, one chronosequence of Norway spruce was studied on sandy soil in the province of Halland, east of Halmstad. In Denmark, one oak chronosequence and one Norway spruce chronosequence were studied on a clay-rich and nutrient-rich soil at Vestskoven, close to Copenhagen. Just outside Vestskoven, a 200-year-old afforested oak stand at Ledøje Plantage was included for comparison. Another Norway spruce chronosequence in a contrasting environment was established on sandy, nutrient-poor soil at Gejlvang, west of Vejle in southern Jutland (Figure 3 & 4). In the Netherlands, one chronosequence of oak and one chronosequence of Norway spruce were studied on similar sandy soils close to Sellingen. Selected site characteristics are provided in Table 1. Detailed site descriptions are given in the Papers I-V.

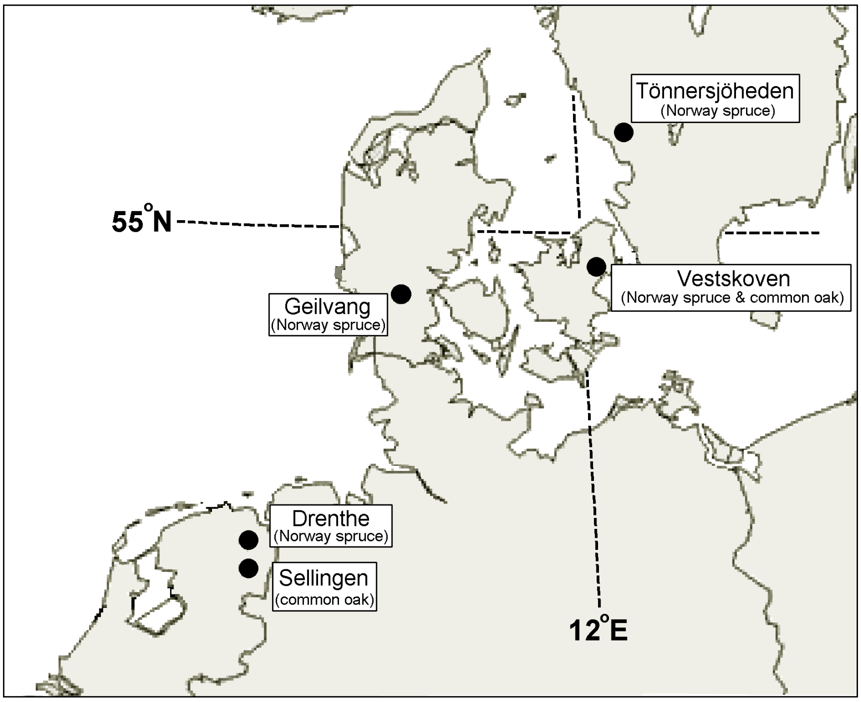

Figure 2. Locations of the six afforestation chronosequences in Sweden (Tönnersjöheden), Denmark (Vestskoven and Gejlvang) and the Netherlands (Sellingen and Drenthe).

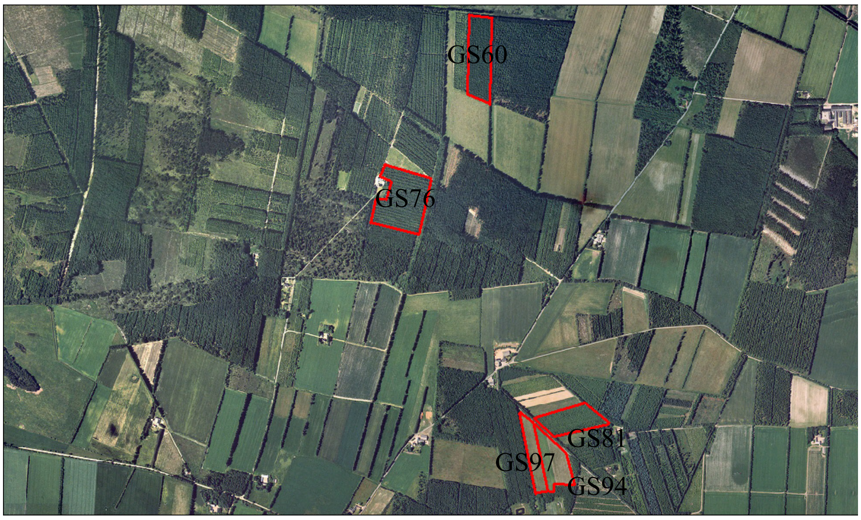

Figure 3. Overview of the Gejlvang chronosequence near Vejle in Denmark. Danmarks Digitale Ortofoto 1999 © Cowi A/S.

Table 1. *Selected site characteristics for the six investigated forest chronosequences in Sweden, Denmark and the Netherlands*

Country	Sweden	Denmark	Denmark	Denmark	The Netherlands	The Netherlands
Chronosequence	Tönnersjöheden	Vestskoven	Vestskoven	Gejlvang	Sellingen	Drenthe
Tree species	Norway spruce	Norway spruce	Oak	Norway spruce	Oak	Norway spruce
Closest city	Halmstad	Copenhagen	Copenhagen	Vejle	Groningen	Assen
Year of planting	1983	1997	1993	1997	1998	1997
	1972	1990	1988	1994	1995	1994
	1937	1988	1977	1981	1991	1990
	1928	1973	1979	1976	1984	
	1910	1969	1970	1960		
			~1800[1)]			
Soil type (USDA/FAO)	Arenosol	Hapludalf	Hapludalf	Durorthod/Quartzipsamment	Gleyic Podzol	Gleyic Podzol
Soil texture	Sandy	Sandy loamy	Sandy loamy	Sandy	Sandy	Sandy
Mean precipitation (mm yr^{-1})	800-1050	640	640	900	745	745-830
Mean temperature (ºC yr^{-1})	6.1-7.3	7.7	7.7	7.7	9	9

[1)] The 200-year-old mixed deciduous stand at Ledøje Plantage was not considered as part of the chronosequence, but was included for comparison.

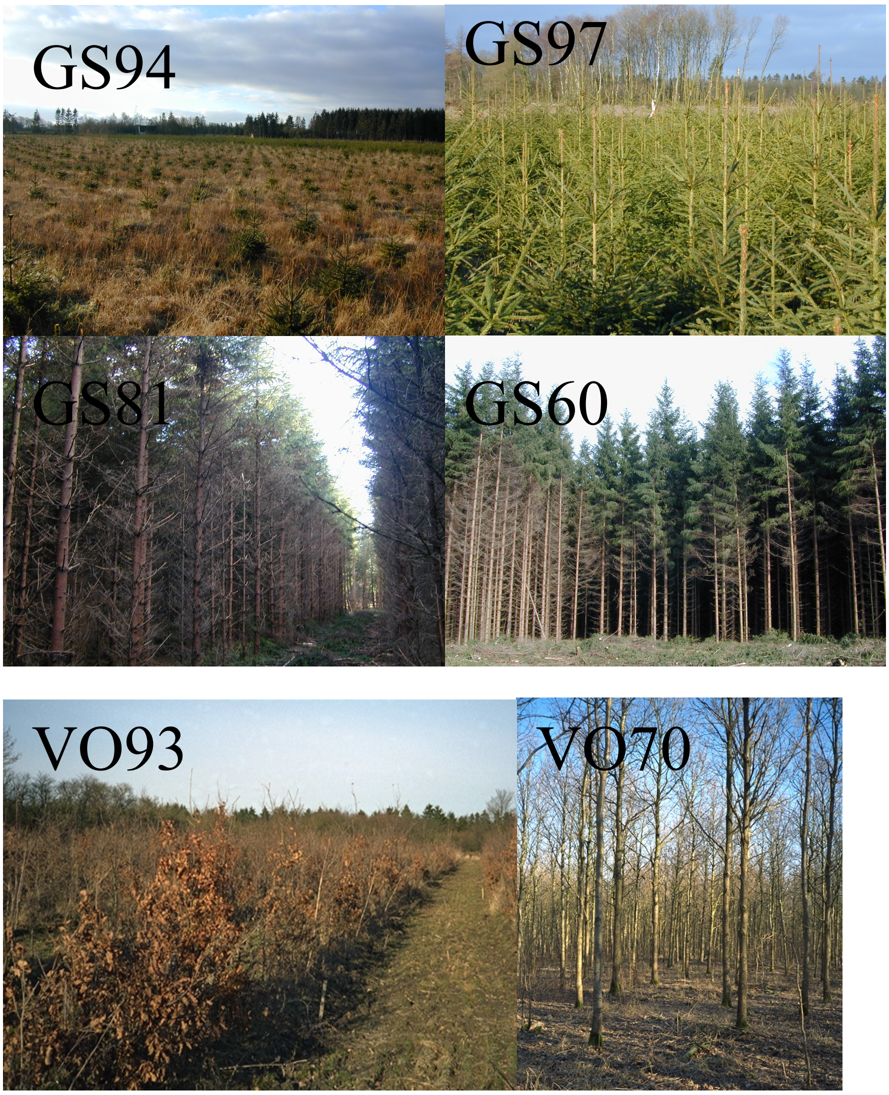

Figure 4. Stands from the Gejlvang Norway spruce chronosequence (GS94, GS97, GS81 & GS60) and from the Vestskoven oak chronosequence (VO93, VO70). Photos: Karin Hansen and Lars Vesterdal. Abbreviations: G=Gejlvang, V=Vestskoven, S=Norway spruce, O=common oak, and the year of planting.

Sampling scheme

Bulk precipitation, throughfall and soil solution (Papers I-III and V)

Bulk precipitation, throughfall and soil solution were sampled monthly for approximately two years in the Swedish and Danish chronosequences and somewhat shorter in the Dutch chronosequences (Table 2). Sampling of bulk precipitation and throughfall was conducted using polyethylene funnels. Bulk precipitation was collected in open areas adjacent to the forest stands using collectors similar to those used for throughfall.

Table 2. *Sampling periods for bulk precipitation, throughfall, soil water and soil water content in the chronosequences in Sweden (SE), Denmark (DK) and the Netherlands (NL).*

Country	Location	Species	From	To
SE	Tönnersjöheden	Norway spruce	Apr-01	June-03
DK	Vestskoven[1)]	Oak & Norway spruce	Dec-00	Dec-02
	Ledøje[2)]	Mixed deciduous	Dec-00	Dec-02
	Gejlvang	Norway spruce	Apr-01	Mar-03
NL	Sellingen	Oak	Apr-01	Dec-02
	Drenthe [3)]	Norway spruce	Apr-02	Mar-03

[1)] At Vestskoven, additional soil water sampling was carried out 1999-2001 in one subplot per stand (for details see Paper II).
[2)] Bulk precipitation was not measured at Ledøje.
[3)] Only soil water is dealt with at Drenthe.

The mineral soil solution was sampled by suction cup lysimeters. Monitoring periods and sampling intervals were the same as those for collection of precipitation and throughfall (Table 2). In Denmark and the Netherlands, concentrations measured at 0.9 m depth represent concentrations in soil water leaving the root zone. In Sweden, the corresponding depth was 0.6 m. In Sweden, soil leachate from the O horizon was collected during one year (May 2002-June 2003) in two mature stands (planted 1910 and 1937).

Soil water content (Paper III)

Soil moisture was measured using TDR (Time Domain Reflectometry) equipment. Sampling periods for each country are shown in Table 2. In Sweden and Denmark, the TDR probes were located within fixed depth intervals of 0-0.2 m, 0-0.5 m, and for Denmark only, within 0-0.9 m. Soil water contents were recorded monthly in all forest stands. Information on the measurements of soil water content in the Dutch chronosequences is available in van der Salm *et al*. (2006).

Soil and litterfall (Papers I, II & IV)

The soil sampling designs and methods for estimation of C and N stocks are summarised in Table 2.2 in Paper IV. Soil samples were taken from the forest floor (*i.e*. O horizon: Oi, Oe and Oa when present) and the mineral soil down to the base of the plough layer (*i.e*. approximately 0-25 cm). Detailed descriptions of the sampling and analysis of soils in Denmark may be found in Vesterdal, Ritter & Gundersen (2002) and Ritter, Vesterdal & Gundersen, 2003, as well as in Paper

IV. With regard to soil sampling and analysis in Sweden and the Netherlands, see Denier van der Gon *et al*. (2004) and Paper IV.

Total litterfall was measured in selected chronosequence stands using slightly different sampling designs and equipment (section 2.4 in Paper IV). Sampling periods extended over two years in Denmark and over one year in Sweden and the Netherlands.

Biometrics (Paper IV)

Forest mensuration parameters were determined in order to determine C stocks in aboveground biomass. In selected chronosequence stands, diameter at breast height, tree height and stem number were measured and standing volumes of commercial-grade wood were calculated according to commonly applied methods in the three countries (Paper IV). Stand mensuration was performed during June 2001 in Denmark, September 2002 in Sweden and October 2002 in the Netherlands.

Leaf area index (Paper III)

In Sweden and the Netherlands, leaf area index (LAI) was measured with a LI-COR LAI-2000 Plant Canopy analyser. For Norway spruce stands at Tönnersjöheden, the measured values were multiplied by a correction factor of 1.6 according to Gower & Norman (1991) to obtain the actual LAI. In Norway spruce stands at Vestskoven, Denmark, fisheye photos were taken during the autumn of 2003 and converted to LAI by Hemiphot (ter Steege, 1996). In order to attain a suitable correction factor, LAI was also measured (March 2005) in one spruce stand (planted 1969) using LICOR LAI 2000. In the oak stands at Vestskoven, LAI was estimated using the METAFORE metamodel developed for the EU-project AFFOREST (van Deursen *et al*., 2007). Leaf area index in Norway spruce stands at Gejlvang was estimated from the relationship between LAI and stand age in the Norway spruce chronosequences at Vestskoven and Tönnersjöheden.

Calculations

Deposition and soil solution (Papers I-III and V)

The bulk and throughfall deposition of various N forms (NO_3^-, NH_4^+ and DON), DOC, and additional cations (Ca^{2+}, Mg^{2+}, K^+, Na^+, Al^{3+} and H^+) and anions (Cl^- and SO_4^{2-}) were calculated by multiplying the amount of precipitation by the concentrations of these elements in the appropriate fraction.

Total deposition of inorganic N (NO_3-N + NH_4-N) to the forest ecosystem was estimated using a canopy exchange model, the extended Ulrich model (Draaijers & Erisman, 1995; Draaijers *et al*., 1998; de Vries *et al*., 2001). This model allows for discrimination between canopy exchange and atmospheric deposition using long-term throughfall and precipitation fluxes. Detailed information on the canopy exchange model and its application to the forest chronosequences are found in Paper I.

In Sweden and Denmark, the leaching of nitrate from the root zone (at 0.6 and 0.9 m depth, respectively) was calculated from measured monthly concentrations times estimated monthly soil water fluxes at corresponding depths. Annual fluxes were calculated by summation of monthly fluxes. In the Netherlands, the soil solution was sampled discontinuously. Here, leaching of nitrate was calculated using the concentration, linearly interpolated on a daily basis between sampling occasions, multiplied by the estimated daily flow of soil water at 0.9 m depth.

The vertical flow of soil water was simulated using the dynamic simulation model SWAP (van Dam *et al*., 1997). The water transport module of SWAP is based on the well-known Richards equation (Richards, 1931). The potential evapotranspiration is calculated with the Penmann-Monteith equation (Monteith, 1965). Input to the model consists of daily meteorological data, abiotic characteristics of the site and vegetation-dependent parameters. The SWAP model was parameterised using measured data on rainfall, throughfall and soil water content at various depths. Calculated Cl balances were constructed as an additional possibility for validation of the simulated hydrological fluxes. Detailed information on the soil hydrological model and its parameterisation is presented in Paper III.

Carbon and nitrogen stocks in soil and litterfall (Papers I, II & IV)

Forest floor C and N contents were calculated by multiplying C and N concentrations by forest floor mass. For the mineral soil C and N contents, the ≥ 2mm fraction was neglected (McNabb, Cromack & Fredriksen, 1986; Homann *et al*., 1995) and SOC and N contents in [Mg ha^{-1}] for the soil layers to 25 cm depth were calculated according to equations given in Paper IV. The litterfall C and N fluxes were quantified by multiplying C and N concentrations by annual litterfall mass.

Carbon stocks in biomass (Paper IV)

In Sweden, biomass dry weight in whole tree and tree compartments was estimated according to functions presented by Marklund (1988), where the diameter at breast height and tree height are used as independent variables. In Denmark and the Netherlands, calculated tree volumes were transformed into biomass using country-specific expansion factors for oak and Norway spruce (section 2.3 in Paper IV). In all three countries, dry mass was converted to C stores by multiplying by 0.5 (IPCC, 2003).

Statistics (Papers I, II & IV)

Simple linear regression was used to explore relationships between stand age and C and N dynamics (C and N contents and C/N ratios) in soil, biomass and litterfall after afforestation. The influence of tree species in Denmark was tested by analysis of covariance (Vesterdal, Ritter & Gundersen, 2002; Ritter, Vesterdal & Gundersen, 2003). No transformations were necessary to fulfil the requirements regarding normally distributed residuals and homogeneity of variances. All statistical tests were carried out using the procedure GLM in SAS (SAS Institute, 1993). The 200-year-old mixed deciduous stand at Ledøje (LM1800) was not included in regressions, but was included in figures for comparison.

Results and Discussion

Nitrate leaching from afforested farmland (Papers I and II)

Effects of plantation age, soil type and N deposition

All available knowledge indicates that a change in land use from agriculture to forestry leads to decreased nitrate leaching to groundwater and surface water. This is mainly a result of ceased fertilisation and less frequent soil disturbance in the new forest (Paper II). Measured nitrate concentrations in soil solution just below the root zone in the six chronosequences in Sweden, the Netherlands and Denmark (Figure 4.5a in Paper I) were mostly markedly lower than the average nitrate concentrations for arable soils reported in the three countries (Johnsson & Mårtensson, 2002; Fraters *et al*., 2004; Grant *et al*., 2004). Average concentrations beneath several oak stands (>20 years) and one old spruce stand at Vestskoven approached the drinking water standard (50 mg NO_3^- l^{-1}) but were still considerably lower than current concentrations beneath arable soils.

Estimated losses by nitrate leaching varied considerably among the chronosequence sites (Figure 4.5b in Paper I) but were at least 3-5 times less than the nitrate leaching from arable land reported in the three countries (Rijtema & de Vries, 1994; Johnsson & Mårtensson, 2002; Grant *et al*., 2004). Highest leaching losses were found in the oak chronosequence at Sellingen in the Netherlands, where nitrate leaching reached a level of 26 kg ha^{-1} yr^{-1} in the 8-year-old stand. In the rich soil at Vestskoven, nitrate leaching was also high, with maximum average levels of about 13 kg N ha^{-1} yr^{-1} in the oldest spruce stand (planted in 1969) (Figure 4 in Paper II).

Our results showed that plantation age strongly affected nitrate concentrations and nitrate fluxes in seepage water from the root zone. Gundersen *et al*. (2003) presented a hypothesis for the development of soil water nitrate concentration below the root zone after afforestation on former arable land (Figure 1 in Paper II) where they hypothesised that the cessation of agricultural activities, including N fertilisation, would lead to an immediate reduction in nitrate concentration in soil water leaving the root zone. This was supported by findings in the youngest spruce stands at Vestskoven (Figures 2 & 3 in Paper II). The majority of stands showed low or negligible average nitrate concentrations and annual nitrate leaching rates between 5 and 20 years of age. This is in line with the stated hypothesis and may indicate high uptake rates of N during the stand expansion phase. In this period, trees have a high demand for N to satisfy the growth of N-rich tissues such as leaves, twigs and roots (Miller & Miller, 1988; Richter *et al*., 1999; Bastrup-Birk & Gundersen, 2004). On the other hand, concentrations and leaching rates of nitrate were consistently high beneath the young oak stands at Sellingen (age range 8-18 years) (Figure 4.5 in Paper I). Obviously, factors other than the biological N uptake, such as the net N mineralisation rate and external N input, are more important for the regulation of nitrate export from these sites (van der Salm *et al*., 2006).

After canopy closure, when the trees had matured and no longer had the same need for N and when N deposition had increased because of larger leaf area and higher filter capacity of taller trees, nitrate concentrations and nitrate leaching increased again for several stands (Paper II). In particular, all stands older than 20 years at Vestskoven (except the 29 year-old spruce stand, planted in 1973) showed elevated nitrate concentrations and leaching higher than 5 kg N ha^{-1} yr^{-1} (Figures 3 & 4 in Paper II). However, a tendency for decreasing nitrate leaching after afforestation was observed along the oak chronosequence at Sellingen. Here, annual nitrate leaching fluxes appeared to decrease from 16-26 kg N ha^{-1} in 8-12 year-old stands to about 8 kg N ha^{-1} in the 18-year-old stand (Figure 4.5b in Paper I). The original hypothesis (Figure 1 in Paper II) was therefore supported mainly by forest stands growing on the highly nutrient-rich and clay-rich soil at Vestskoven. The results suggest that new forests established on nutrient-rich and clay-rich former arable soils continue to be N-saturated and leach nitrate after afforestation, apart from a transient period early in the rotation (5-20 years after planting) when trees have a high N demand for growth.

The problem of N saturation is particularly pronounced in areas with chronic N deposition. The calculated total atmospheric input of N to the forests, as well as the measured throughfall N flux, tended to decrease along a south-west (the Netherlands) – north-east (Sweden) gradient (Table 4.3 in Paper I). The measured throughfall fluxes of inorganic N (NO_3-N + NH_4-N) varied between 10 and 20 kg ha^{-1} yr^{-1} in the oak stands and between 8 and 25 kg ha^{-1} yr^{-1} in the spruce stands. In all stands, the calculated total deposition of inorganic N ranged from 11 to 34 kg ha^{-1} yr^{-1}. The highest deposition levels were obtained at sites where surrounding agriculture was most intensive. European data have shown that nitrate leaching from forests mainly occurs above a threshold value of about 10 kg N ha^{-1} yr^{-1} in throughfall input (Dise *et al*., 1998; Gundersen *et al*., 1998; Kristensen *et al*., 2004). A similar observation was made in our study, but at high deposition rates the leaching seemed to be largely dependent on ecosystem properties such as soil type and N status (Figure 4.7 in Paper I, Figure 5 in Paper II). This is in line with previous analyses of European datasets, where about half of the variability in nitrate leaching from forests was explained by N deposition, and part of the remaining variability could be explained as an effect of ecosystem N status (Gundersen, Schmidt & Raulund-Rasmussen, 2006).

The measured throughfall deposition of N showed an increasing trend with tree height (and stand age) in all chronosequences (Figure 4.1 & Table 4.3 in Paper I). Bulk deposition exceeded throughfall deposition, especially in the youngest spruce stands, suggesting foliar uptake. Canopy exchange modelling indicated that 6-41% (1-11 kg N ha^{-1} yr^{-1}) and 5-38% (1-11 kg N ha^{-1} yr^{-1}) of the total N deposition to the spruce and oak stands, respectively, was taken up by the forest canopy (Table 4.3 in Paper I).

Effect of tree species

Our data demonstrated that tree species selection had a great impact on the magnitude and temporal pattern of nitrate leaching from the root zone after afforestation of arable land. Over the first 30 years since land use conversion, oak stands generally leached more nitrate than spruce stands of comparable age (Figure 4.5b in Paper I). On average, 7-8 kg N ha^{-1} yr^{-1} were leached from the old oak stands (>20 years) at Vestskoven, and a similar leaching rate (approx. 8 kg N ha^{-1} yr^{-1}) was obtained in the nearby 200-year-old mixed deciduous stand situated on a similar soil (Figure 4 in Paper II). In contrast, leaching of nitrate was very low or negligible under Norway spruce stands at Vestskoven, except for the oldest stand (33 years old, planted 1969) where the highest average leaching rate was observed (13 kg NO_3-N ha^{-1} yr^{-1}). In the Netherlands, nitrate leaching ranged between 8 and 26 kg N ha^{-1} yr^{-1} in the oak chronosequence at Sellingen, whereas leaching was always negligible in spruce stands of similar age at Drenthe.

Our results are partly in conflict with those of previous studies of paired mature stands of coniferous and deciduous species at the same sites showing higher seepage water nitrate concentration and higher nitrate leaching under conifers (De Vries & Jansen, 1994; De Schrijver *et al*., 2000; Rothe *et al*., 2002; Gundersen, Schmidt & Raulund-Rasmussen, 2006). Higher nitrate leaching under conifers was mainly attributed to higher N deposition on coniferous trees than on deciduous. With the exception of the 33-year-old Norway spruce stand, there was no general difference in N deposition levels between similarly-aged oak and spruce stands at Vestskoven that could explain the difference in nitrate leaching between tree species (Table 4.3 in Paper I). With similar input of N, Kristensen *et al*. (2004) found higher nitrate concentrations in seepage water beneath deciduous forests compared with coniferous. However, this difference was mainly attributed to a general soil type difference between stands of the two forest types as represented in the European intensive monitoring network (level II). In general, deciduous forests grow on more nutrient-rich soils, which are more prone to leach nitrate. At Vestskoven, where oak and Norway spruce stands grow on the same nutrient-rich soil type in the same area, possible causes could be differences in uptake or internal cycling of N between tree species.

The difference in nitrate leaching between oak and spruce may result from the difference in growth rate (and hence N uptake) between the two species. Vesterdal *et al*. (2007) found a 70% higher accumulation of biomass in spruce than in oak (section 3.1 in Paper IV), which may also indicate a higher accumulation of N in biomass, at least partly explaining the difference in nitrate leaching between oak and spruce at the same annual N deposition rate. Furthermore, larger N storage in the accumulated forest floor under spruce stands (11.9 kg N ha^{-1} yr^{-1}) than under oak (2.4 kg N ha^{-1} yr^{-1}) over 30 years might prevent N leaching from the spruce stands (Figure 2.4 in Paper IV).

The absence of a general difference in N deposition between oak and spruce at Vestskoven might partly be attributable to the short part of the rotation period in which we could compare species (0-35 years). At this young stage, commonly reported species-related differences may not yet have become fully pronounced. In a long time perspective, the higher N deposition to the evergreen spruce stands may result in larger nitrate leaching losses from spruce stands than from oak, as often reported in the literature (Gundersen, Schmidt & Raulund-Rasmussen, 2006). In addition, the different growth patterns of oak and spruce may contribute significantly to this development. Compared with oak, Norway spruce growing on rich, loamy soils has a higher initial growth rate but a shorter life cycle. In Denmark, a decrease in the growth rate of Norway spruce on clay soils have been observed at approx. 40 years of age (Hansen, 1981) and a similar observation has been made in Sweden (Johansson, 1995b). Declining increment rates (decreased N demand) in mature spruce stands may cause nitrate export from the root zone to increase relative to that from the more continuously growing and longer lasting oak stands.

Effect of afforestation on water recharge (Paper III)

The long-term effects of afforestation on the stand water balance were assessed along the four chronosequences in Denmark (oak and Norway spruce, Vestskoven; Norway spruce, Gejlvang) and Sweden (Norway spruce, Tönnersjöheden) (Paper III). The influence of land use conversion at the oak and Norway spruce sites in the Netherlands has previously been described by van der Salm *et al*. (2006). Changes in interception evaporation were investigated by measuring rainfall and throughfall volumes in the various stands. Changes in water recharge were calculated using the soil hydrological model SWAP. In general, hydrological fluxes were reasonably well reproduced by the SWAP model.

Afforestation had a clear influence on water recharge at the sites. Our study indicated a 7-32% (12-173 mm) reduction in annual water recharge across all chronosequences resulting from an increase in forest age over a period of 5 to 92 years (Figure 3 in Paper III). Previous analyses generally reveal larger reductions (approx. 25-45%) in water recharge following conversion from arable land to mature forest (Meuser, 1990; Bastrup-Birk & Gundersen, 2004; Farley, Jobbágy & Jackson, 2005; van der Salm *et al*., 2006). Results from several studies indicate that water recharge reductions are attained rapidly after afforestation, with the fastest declines occurring within the first 5 to 20 years after planting of the trees (Le Maitre & Versfeld, 1997; Farley, Jobbágy & Jackson, 2005; van der Salm *et al.*, 2006). Our field studies were mainly restricted to areas afforested 5 to 30 years ago and arable land was not represented. Enlargement of the model analyses to also include younger stands and arable land would most likely have revealed larger water recharge reductions after afforestation.

The reduction in water recharge after afforestation was highly dependent on plantation age. The general pattern revealed a sharp initial decline in water recharge in the first 30-40 years, followed by a distinct recovery in the older Swedish spruce stands (Figure 3 in Paper III). Whereas the decline in water recharge was mainly due to increased interception evaporation with age, the recovery in older stands appeared to be largely caused by decreased transpiration in the old forests (Figure 4 in Paper III). The observed age dependence of the water recharge after afforestation is supported by the review of Farley, Jobbágy & Jackson (2005).

The choice of tree species had a clear influence on the simulated hydrological fluxes. At Vestskoven, spruce stands older than about 20 years used more water and hence showed lower water recharge than adjacent oak stands of comparable age (Figures 3 & 4 in Paper III). The estimated difference in water recharge was 60-90 mm yr^{-1}. A compilation of hydrological fluxes in Danish forests (Bastrup-Birk, Gundersen & Hansen, 2003) revealed a similar difference between deciduous and coniferous tree species (80-110 mm yr^{-1} higher in oak). However, other model studies have reported slightly larger differences between related stands of oak and spruce in Denmark (Hansen, 2003; Bastrup-Birk & Gundersen, 2004).

Potential of afforestation to mitigate increasing atmospheric CO_2 (Paper IV)

Contribution of biomass and soils to total ecosystem C sequestration

For the investigated ecosystem as a whole (*i.e.* living aboveground and belowground biomass, O horizon and the mineral soil down to 25 cm), the relationship with age indicated sequestration of approximately 250 Mg C ha^{-1} (mean rate 2.8 Mg ha^{-1} yr^{-1}) over the chronosequence of 90 years for all species and sites. This is in agreement with other chronosequence studies on afforestation in temperate climates, which have reported ecosystem C sequestration of between approximately 240 and 275 Mg ha^{-1} (mean rates 2.1-2.8 Mg ha^{-1} yr^{-1}) for Norway spruce and pine plantations 93-115 years after land use conversion (Hooker & Compton, 2003; Thuille & Schulze, 2006).

The living tree biomass was the major sink of C following afforestation (Figure 2.7 in Paper IV). In stands younger than 45 years, mean rates of biomass C sequestration were more than twice as high as rates for stands between 60-90 years of age (Table 2.3 in Paper IV). However, the effect on biomass C sequestration over a full rotation remains more uncertain due to limited data (only four chronosequence stands above 40 years of age). The apparently lower rate of biomass C accretion in the older spruce stands in Sweden is thought to be partly due to a general decline in net primary production with increasing forest age (Murty & McMurtrie, 2000) and partly due to an increased impact of thinning on the biomass C storage in old stands. Moreover, forest stands older than about 50 years were established on soils subjected to less intensive agricultural practices (*e.g.* fertilisation, liming) compared with soils available for afforestation today. Furthermore, the older plantations experienced a lower level of N deposition in their early growth stages than more recently planted forests (Lövblad, 2000). Thus, forest plantations on arable soils abandoned today may constitute a greater sink for C compared with plantations on arable soils abandoned more than 50 years ago.

The aboveground and belowground biomass accounted for about two thirds of the increase in ecosystem C, while the remaining approx. one third was sequestered in the O horizon and the Ap horizon. This relationship was largely driven by data from the long 90-year-old Swedish spruce chronosequence. The contribution of soil varied among chronosequences from 0 to 31%. Other studies have reported relative contributions of soils to total ecosystem C in a range between 12 and 24% (Table 2.5 in Paper IV). The chronosequences in this study showed rates of total soil C sequestration between 0 and 1.3 Mg C ha^{-1} yr^{-1} over 30 to 90 years since afforestation. The average rate of total soil C sequestration was 0.8 Mg C ha^{-1} yr^{-1}

across all sites, which is considerably higher than the average accumulation rate of soil C (0.34 Mg ha^{-1} yr^{-1}) reported in a review by Post & Kwon (2000).

The Swedish chronosequence was outstanding in terms of its large soil C sequestration, almost solely caused by forest floor build-up. Sixty to 90 years of forest development have resulted in the accumulation of a thick (6-8 cm) O horizon with a C content comparable to that of soils with a more continuous forest history. Ninety years after forest establishment, the O horizon had accumulated 53 Mg ha^{-1} of new C, which is even higher than the 39 Mg C ha^{-1} reported as a mean value for the O horizon below coniferous forests on similar coarse-textured soil types in this part of Scandinavia (Callesen *et al.*, 2003). The maritime climate in south-western Sweden may support rather high biomass and litter production owing to the relatively long growing season (approx. 200 days) and good supply of water (precipitation approx. 1000 mm yr^{-1}). In addition, forest floor build-up may be promoted by low substrate quality (*i.e.* high lignin concentration) of the coniferous litter (Johansson, 1995a) and a sandy soil texture (Vesterdal & Rauland-Rasmussen, 1998) resulting in slow rates of litter decay.

While C sequestration was always evident in the forest floor, C change in the mineral soil was more variable among chronosequences (Table 2.4 in Paper IV). The relative contribution of mineral soil to total soil C sequestration ranged from 0 to about 80%. Chronosequences with a significant positive change in soil C were all situated on relatively poor sandy soils (Gejlvang and the Dutch chronosequences). In contrast, a decrease in mineral soil C was evident in rich loamy soil in the first 30 years after afforestation. Results from the Swedish spruce chronosequence of unchanged mineral soil C stocks over 90 years suggest that there is little reason to expect mineral soil C sequestration at all sites following afforestation of arable land, even over a full rotation. This result may be considered surprising since our flux calculations for 65-90-year-old stands (Table 4 in Paper V) suggested that more than 90% (230-280 kg ha^{-1} yr^{-1}) of DOC leaving the O horizon was retained within 0-60 cm of the mineral soil and could be sequestered. After sorption in the mineral soil, sorbed organic matter (OM) originating from the O horizon is considered to mineralise rather slowly (Kalbitz *et al.*, 2003, 2005). Kalbitz *et al.*, (2005) found significantly longer residence times for sorbed OM from the Oa horizon (91 years) compared with sorbed OM from maize straw (40 years). At the Swedish spruce sites, Karltun *et al.* (2005) showed that a substantial part of DOC in the A horizon must be derived from SOM formed before afforestation for stands younger than about 40-50 years, indicating mobilisation of old (pre-afforestation) SOM in the A horizon after afforestation. In the first decades after afforestation, rapid decomposition of SOM inherited from agriculture may limit C sequestration in the A horizon.

In addition, erratic variation in the data may partly explain the lack of a consistent trend in mineral soil C storage along the Swedish chronosequence. Considerable variability in mineral soil C at the afforested sites might be expected considering the great range in soil C content (54-93 Mg C ha^{-1}) in the A horizon of arable reference sites (Figure 2.5 in Paper IV). The variability in soil C can partly be attributed to differences in soil properties (*e.g.* Al and Fe oxohydroxides, soil texture and clay content) and management history among sites. The use of a paired plot analysis (Figure 2.6 in Paper IV) may to some extent have reduced the spatial variability attributable to differences in site properties, but did not change the interpretation of results.

Influence of soil type and tree species on C sequestration after afforestation

The total ecosystem C sequestration was higher in Norway spruce than in oak in the short term (30-40 years), whereas soil type did not clearly influence the rate of C sequestration. It is well known that biomass production for certain tree species varies considerably between sites due to soil nutrient status and climate. For example, Johansson (1995b) found significant differences in biomass growth for Norway spruce plantations on farmland with different soil types in southern Sweden. Trees grew faster on fine sandy-silty till soils and more slowly on sandy till soils, peat soils and fine-grained sediment soils. Surprisingly, the rate of biomass C sequestration differed relatively little between Norway spruce stands on highly contrasting soil types within Denmark. Spruce stands on the nutrient-rich and clay-rich soil at Vestskoven sequestered C at only a slightly higher rate than spruce stands on the nutrient-poor sandy soil at Gejlvang. This suggests that the influence of parent material on growth in the first 45 years following afforestation appears to be masked by the soil enrichment, which is a legacy of former agriculture.

At Vestskoven, the more fast-growing spruce sequestered significantly more C (4.6 Mg C ha^{-1}) in biomass over 30 years compared with oak (2.7 Mg C ha^{-1}) (Table 2.3 & Figure 2.2 in Paper IV). In the Netherlands, comparison between species was difficult because of too few spruce stands. In the short term (30 years), tree species selection had little influence on total soil C sequestration, although Norway spruce sequestered more C in the forest floor than oak (Table 2.4 in Paper IV). The difference in forest floor C content between oak (2.4 Mg ha^{-1}) and spruce (11.9 Mg ha^{-1}) at Vestskoven is in line with general observations in Denmark (Vesterdal & Raulund-Rasmussen, 1998).

For the total soil compartment studied, afforestation of nutrient-poor sandy soils resulted in larger C sequestration than afforestation of nutrient-rich clayey soils. This was mainly due to C changes in the mineral soil compartment after afforestation. Storage of C in the mineral soil was most efficient in sandy nutrient-poor soils (*cf.* Norway spruce, Gejlvang and oak and spruce, the Netherlands), whereas mineral soil C contents tended to decline in the nutrient rich soil at Vestskoven (Table 2.4 in Paper IV). We had expected the parent material and the soil type to cause larger difference in forest floor C accumulation over 40 years on sandy (0.43 Mg C ha^{-1} yr^{-1}) as opposed to clayey (0.35 Mg C ha^{-1} yr^{-1}) soils in Denmark. Forest soils with sandy texture, low nutrient availability and low pH typically store more C in the forest floor than more fine-textured soils of higher nutrient status (Vesterdal & Raulund-Rasmussen, 1998; Callesen *et al.*, 2003). The similar rates of forest floor C sequestration under Norway spruce on contrasting soil types within Denmark can be attributed to the high nutrient availability and pH in recently abandoned cropland soils.

Fluxes and retention of DOC and DON after afforestation (Paper V)

We investigated the long-term effects of afforestation on DOC and DON dynamics in the ecosystem along the Swedish 90-year-old spruce chronosequence. Additional DOC data were derived from previous sampling at the same study sites including adjacent arable plots and a few additional afforested plots.

Results showed that DOC concentrations and fluxes with throughfall were positively correlated with tree height (Figure 1 in Paper V). Possible explanations are (i) a gradually increasing biomass of old senescent leaves being more susceptible to ion leakage (Schaefer & Reiners, 1990), (ii) a larger biomass of needles and branches in older stands increasing the contact area between rainwater and needle and branch surfaces, and (iii) increased dry deposition of DOC with age due to increased roughness in taller stands. Enrichment of DOC in throughfall under forests has been reported in previous studies (Alenäs & Skärby, 1988; McDowell & Likens, 1988), but according to our knowledge no other study has described the effect of increasing stand age and tree height on throughfall fluxes of DOC and DON following afforestation. DON throughfall fluxes showed no clear trends with stand age and tree height (Table 4 in Paper V), suggesting different origins of DOC and DON in throughfall.

The highest concentrations and fluxes of DOC and DON were (as expected) found in soil leachate from the O horizon (Table 3 & 4 in Paper V). Here, DOC flux amounted to 250-310 kg ha^{-1} yr^{-1} and DON flux to 8-9 kg ha^{-1} yr^{-1} in stands afforested between 65 and 92 years ago. Interestingly, these levels are of the same order of magnitude as those typically observed in O horizon leachate at sites with a long history of forest cover (Michalzik *et al*., 2001; Fröberg *et al*., 2006).

Concentrations and fluxes of DOC and DON in the mineral subsoil were consistently low (Table 3 & 4 in Paper V). Flux calculations suggested that more than 90% (230-280 kg ha^{-1} yr^{-1}) of DOC annually leached from the O horizon was retained within 0-60 cm of the mineral soil (Table 4 in Paper V). Even though there was a large influx of DOC to the mineral soil layers in mature stands, there was no clear trend of increasing DOC concentration with stand age in the mineral subsoil. The effect of time since afforestation was partly masked by spatial variability in soil properties that influence the DOC and DON concentration in the mineral soil. Our data indicate that DOC concentrations in the A horizon were primarily related to the oxalate-extractable amounts of Al and Fe in the same horizon (Figure 6 in Paper V). Several studies have shown that the capacity to adsorb DOM relates to the presence of Al and Fe oxides and hydroxides, as these are the most important sources of variable charge in the mineral soil (Moore, Desouza & Koprivnijak, 1992; Kaiser & Zech, 1998). Furthermore, afforestation of arable land appears to induce a progressive qualitative change in SOM and DOM, as revealed by significantly increasing C/N ratios in soil and soil solution over time (Table 1 & Figure 7 in Paper V). This is thought to be partly due to an increased contribution of organic matter with high C/N ratio derived from the new forest, and partly to depletion of N-rich organic matter present in the soil prior to afforestation. We suggest two possible explanations for the observed high

DOC/DON ratio (approx. 40) in the Ap horizon (0-25 cm) under the oldest spruce stand (Figure 8 in Paper V): (i) an increased contribution of root-generated DOC in the forest stand and (ii) an increased uptake of organic N forms with time as the stand develops, resulting in DOM having a higher C/N ratio. The latter explanation is supported by several previous studies on organic N in forest ecosystems (Näsholm *et al*., 1998; Michalzik & Matzner, 1999; Andersson & Berggren, 2005).

Concluding remarks and future perspectives

The results presented in this thesis show that afforestation of former arable land in north-western Europe largely improves environmental conditions. Carbon is sequestered in biomass and soil (Paper IV) and the quality of recharging soil water in terms of nitrate is improved (Papers I and II). On the negative side, afforestation leads to a reduction in water recharge to the groundwater (Paper III), to soil acidification and may also cause nutrient deficiency (Tables 1 and 2 in Paper V). Further, the results show that soil type (loamy vs. sandy soils) and the choice of tree species (Norway spruce vs. common oak) have substantial impact on effects of afforestation. This is valuable knowledge when planning afforestation of former arable land in the future.

It is difficult to directly measure the long-term impact of afforestation, since one would need 50-100 years in order to study a whole rotation period. Instead, adjacent stands of different ages were selected to represent the time-trend following afforestation (*i.e.* chronosequence). However, distinguishing between the effects of forest succession and factors like *e.g.* spatial variability can be difficult. Even though the observed time-trends were generally very distinct they only provide a valuable estimate of changes in time following afforestation of arable land. Repeated sampling in the chronosequences about ten to twenty years after the first sampling could validate these trends and provide real evidence of the directional change for each stand.

Moreover, an increase in the number of afforested sites is needed in order to be more conclusive on whether the observed time-trends and tree species-related differences are general features for the ecosystems studied or whether large variability exists between sites showing comparable characteristics. Our hypothesis that nitrate concentrations (and leaching) in seepage water would be low in young stands and increase again after canopy closure was only supported by stands on nutrient-rich, fine-textured soil. Moreover, nitrate leaching was generally higher beneath oak than beneath Norway spruce, which is partly in contrast to results obtained in old forest ecosystems. These observed patterns need to be confirmed by measurements at other sites. In addition, extension of the chronosequences to include older stands is necessary in order to predict the long-term (forest rotation) effects of afforestation on the environmental state variables studied.

In particular, the interpretation of nitrate leaching is complicated by the transient nature of the factors that regulate leaching losses of N from the system. The afforested ecosystem is not (yet) in steady state and changes in the processes are ongoing. In order to better explain the change in nitrate leaching over time following afforestation, as well as the observed differences in N retention between contrasting tree species, we need to perform more detailed modelling of N cycling characteristics (mineralisation and nitrification rates) among sites or alternatively measure these variables in the field.

Afforestation today in north-western Europe often serves multiple purposes *e.g.* biomass production, water quality protection, restoration of biodiversity and recreational demands. However, afforestation objectives may vary considerably between countries (*e.g.* between Sweden and the Netherlands) and even within countries. Often there is a trade-off between different types of environmental objectives. For example, forestry practices leading to high C sequestration (*e.g.* fast-growing tree species and low thinning intensity) lead simultaneously to low groundwater recharge. It is evident from our results that planning and establishing new forests on arable land in an environmentally sound way requires a holistic and long-term perspective on subsequent environmental changes.

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