

# **Nitrogen deposition and nitrate leaching following afforestation**

## **Experiences from oak and norway spruce chronosequences in Denmark, Sweden and The Netherlands**

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## CHAPTER 4

### NITROGEN DEPOSITION AND NITRATE LEACHING FOLLOWING AFFORESTATION: EXPERIENCES FROM OAK AND NORWAY SPRUCE CHRONOSEQUENCES IN DENMARK, SWEDEN AND THE NETHERLANDS

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**Abstract.** Knowledge on the impact of afforestation of arable land on N deposition and leaching of nitrate to groundwater and surface waters is limited. In the AFFOREST project we evaluated nitrogen (N) deposition and nitrate leaching following afforestation of cropland. Two oak (*Quercus robur*) and four Norway spruce (*Picea abies*) afforestation chronosequences (age range 1 to 90 years) were studied with respect to deposition and nitrate leaching in Denmark, Sweden and the Netherlands. This paper presents a synthesis of these six chronosequence experiments. Three to six forest stands of Norway spruce and/or common oak were monitored in each chronosequence for a period of two years. In each stand, throughfall and soil solutions beneath the root zone were sampled and nitrate leaching was calculated. For all sites and tree species, the throughfall deposition of N increased with stand height (and age). The forests varied substantially in their ability to retain N in the ecosystem. No consistent pattern was apparent in the three countries. However, in some chronosequences nitrate leaching was low or negligible in the early phase of afforestation and increased after canopy closure (> 15-20 years). In general, nutrient-rich clayey soils leached more nitrate than nutrient-poor sandy soils. In the first approximate 35 years after afforestation, nitrate leaching below the root zone was generally higher

below oak than below Norway spruce. The presented results are compared to available studies and discussed.

## 1. INTRODUCTION

During the latest years incentives for afforestation include environmental concerns, e.g. protection of groundwater reserves. Compared to the former agricultural land use, planted trees influence the amount and quality of water reaching the soil by affecting the evaporation (Chapter 3) and nutrient cycling. Afforestation on former arable land also strongly influences the amount of atmospheric deposition since the surface roughness and collecting surface area is increased. Therefore, the amount of dry deposition of air pollutants can be expected to increase (Bleeker & Draaijers 2002).

Decreased nitrate leaching to water bodies is a specific expected environmental effect of afforestating arable land (Rijtema & De Vries 1994; Rowe & Pearce 1994; Jussy et al. 2000). Leaching losses of nitrogen (N) from ecosystems mainly occur as dissolved N in seepage water. Dissolved N may be in the form of nitrate ( $\text{NO}_3^-$ ), ammonium ( $\text{NH}_4^+$ ), or dissolved organic nitrogen (DON). Nitrate is highly mobile in the soil profile and easily lost from the system by leaching. Nitrate is therefore the most relevant N compound for water quality and addressed as such in specific EC directives (91/676/EEC and 98/83/EEC) concerned with the protection of waters against pollution from agricultural sources. In this line, the World Health Organisation has established a requirement of 11.3 mg N dm<sup>-3</sup> (50 mg dm<sup>-3</sup>  $\text{NO}_3^-$ ) for the quality of drinking water (WHO 1998).

Nitrogen leaching is at risk when ecosystems become saturated with N, i.e. when the availability of inorganic N exceeds the demand from plants and microorganisms (Aber et al. 1989; Gundersen 1991). However, even in N-limited systems substantial leaching of nitrate may occur after events of high precipitation in the winter period when evapotranspiration rates and biological uptake of N are low (Gundersen & Rasmussen 1995). Once a forest becomes N saturated, the excess nitrate is leached by percolating soil water down to the saturated zone. Leaching of nitrate may be induced by (i) increased input of N (e.g. atmospheric N deposition, fertilization, planting of  $\text{N}_2$  fixing species), (ii) decreased biological uptake of N (e.g. following clear-cutting, thinning, weed control, site preparation, degradation of vegetation), (iii) increased net mineralization and nitrification rates (e.g. resulting from liming, site preparation, change in litter substrate quality, lowering the groundwater table), and (iv) increased water recharge (e.g. decreased canopy cover after thinning, large precipitation events).

In regions dominated by agricultural activities, N is recognised as a major pollutant of water environments. During the last 40-50 years management of farmland has intensified and caused arable soils to carry large pools of N bound in organic matter, and soils have a high nitrifying capacity (Jussy et al. 2000). Fertilizer application is the dominant source of groundwater nitrate contamination (van der Voet et al. 1996). In contrast to agricultural soils, old existing forests are characterised by a more tight N cycle. Water from old forest land is, therefore, generally of good quality with a relatively low concentration of dissolved N

compared to other land uses (Thornton et al. 2000). A change in land use from agriculture to forestry may induce major changes of the N cycle, including inputs, internal cycling and losses. The external input of N to an ecosystem is dependent on the N capture efficiency of the vegetation, which again is depending on the effective surface roughness. Trees are highly efficient scavengers of pollutants (Allen & Chapman 2001) much better than the lower agricultural crops. During the years after afforestation the forest canopy structure changes continuously with increased surface roughness, which results in an increased capture of dry N deposition for each year (Hansen et al. 2006a). Net throughfall fluxes of  $\text{NO}_3^-$  and  $\text{NH}_4^+$  have been observed to correlate well with the roughness length of the canopy (Thom 1971; Draaijers 1993). The enhanced supply of N in combination with high mineralization may result in potential N saturation and increased nitrate leaching (Allen & Chapman 2001). At the same time, uptake in biomass is high when the trees are growing vividly building up the canopy. The demand for N decreases when the canopy has closed. It is an ongoing process where the relative contribution of the mechanisms that regulate N export is likely to change over time following the forest establishment. Gradually, N stores will move towards a new steady state.

Thus, an effective measure to reduce leaching of nitrate to the groundwater could be the conversion of agricultural land to forest. However, because of the high N status of former arable soils, retention of N in afforested ecosystems may be less efficient than in old forest land, resulting in an enhanced risk of nitrate leaching. Afforestation could also constitute a potential threat for groundwater quality since scavenging of pollutants leads to enhanced supply of N, which in combination with high mineralization may potentially result in N saturation and increased nitrate leaching (Allen & Chapman 2001). However, nitrate leaching may still be less than from the former agricultural land use.

The change in deposition following afforestation has not directly been studied, however, many studies have observed increased deposition when the height and canopy roughness increased as the trees grew (e.g. Draaijers 1993). Knowledge on nitrate leaching from forests established on former arable land is scarce. Rijtema & de Vries (1994) used a simple modelling approach to evaluate the effect of a change in land use from agriculture to forestry in the Netherlands. Afforestation led to a marked decrease in nitrate leaching regardless of the chosen tree species. In Denmark, Bastrup-Birk & Gundersen (2004) simulated N leaching in the Horndrup catchment with the INCA model and found that afforestation substantially reduced N leaching. Concentrations of nitrate ten years after afforestation at three afforested sites in Denmark were much lower than concentrations in agricultural soils before afforestation (Hansen & Vesterdal 1999). However, higher concentrations of nitrate in soils below the root zone were observed in recently (< 10 years) afforested land as compared to old forest ecosystems (Callesen et al. 1999).

This chapter evaluates the effect of afforestation of former cropland on N deposition to forests of changing height/age and on nitrate leaching. The basis for this evaluation is a synthesis of chronosequence experiments in three north-west European countries. The presented results from the AFFOREST project are

compared to available studies on the topic and discussed. The specific objectives are i) to estimate the change in N deposition as the forest grows in chronosequences from recently planted forest to older forest, ii) to estimate nitrate leaching in the same chronosequences, iii) to study the possible differences between afforestation with deciduous (oak) and coniferous (Norway spruce) tree species, and iv) to assess the difference in nitrate leaching after planting on contrasting soil types, i.e. sandy or clayey soils.

## 2. MATERIALS AND METHODS

To study the effect of afforestation with oak and spruce on deposition and nitrate leaching, chronosequences of afforestation stands were selected in Denmark, southern Sweden and the Netherlands (Chapter 1). The study included six chronosequences of which two were differently aged oak stands and four were differently aged Norway spruce stands. In Denmark, one oak chronosequence and one Norway spruce chronosequence were chosen at the same clay-rich and nutrient-rich soil at Vestskoven close to Copenhagen. Another spruce chronosequence in a contrasting environment was established on sandy, nutrient-poor soil at Gejlvang in southern Jutland. In the Netherlands, one chronosequence of oak and one chronosequence of spruce were studied on similar sandy soil close to Sellingen. In Sweden, one chronosequence of spruce was established in the province of Halland east of Halmstad. A map of the locations is found in Chapter 1 and more site information is given in Chapter 1 and on the AFFOREST website ([www.sl.kvl.dk/afforest](http://www.sl.kvl.dk/afforest)).

### 2.1. Precipitation and throughfall sampling

Bulk precipitation and throughfall were sampled for two years in the Danish and the Swedish chronosequences and somewhat shorter in the Dutch chronosequences (Table 4.1). Sampling of both bulk precipitation and throughfall was performed using polyethylene funnels. Detailed information on the length of the monitoring periods and the number of funnels at each site is presented on the AFFOREST web site ([www.sl.kvl.dk/afforest](http://www.sl.kvl.dk/afforest)). In the Netherlands, the funnels were placed in a cross in each stand with definite distance between them. In Sweden, the funnels were placed randomly in each forest stand. In Denmark, an experimental design with circular subplots (10 m radius) was used. In each circular subplot, five funnels were installed, four at the cardinal points and one in the centre. Three circular subplots were placed as far from stand edges as possible. The Dutch throughfall samples from the 10 individual funnels were pooled to 2 samples, each representing an axis of the sampling cross. In Denmark, the samples were pooled in 3 sub-samples, each representing a circular plot. The Dutch collecting bottles were placed in dark PVC tubes hanging above ground, to prevent the influence of direct sunlight. In Sweden and Denmark, the collection bottles were placed in soil pits to prevent them from heat and light. The bulk precipitation and throughfall

samples were collected and weighed in the field at each location. All sampling was carried out in monthly intervals.

**Table 4.1.** *Sampling periods for bulk precipitation, throughfall, and soil water in the chronosequences in Denmark, the Netherlands and Sweden.*

Country	Location	Species	Year	Start	End
DK	Vestskoven	Oak & Norway spruce	1	Jan-01	Dec-01
			2	Jan-02	Dec-02
	Gejlvang	Norway spruce	1	Apr-01	Mar-02
			2	Apr-02	Mar-03
NL	Sellingeng	Oak	1	Apr-01	Mar-02
			2	Jan-02	Dec-02
	Drenthe <sup>1</sup>	Norway spruce	1	Apr-02	Mar-03
SE	Tönnersjöheden	Norway spruce	1	Apr-01	Mar-02
			2	Apr-02	Mar-03

<sup>1</sup> Only soil water is dealt with at Drenthe.

## 2.2. Soil and litterfall sampling

The forest floor is the layer of dead organic matter, i.e. leaves, needles, twigs, branches and fruits, that blankets the mineral soil of a forest. Soil N contents and C/N ratios were assessed for the forest floor and the upper 0-25 cm of the mineral soil as well as these two soil compartments together in all chronosequences. Nitrogen was analyzed using the same soil samples collected for assessment of C, and the sampling procedure was therefore the same. The sampling design and methods are summarized in Chapter 2. More detailed descriptions of the sampling of soils in Denmark and Sweden may be found in Vesterdal et al. (2002), Ritter et al. (2003) and Chapter 2.

Total litterfall was measured for two years in Denmark and for one year only in Sweden and the Netherlands. For information on litterfall sampling see Chapter 2.

## 2.3. Soil solution sampling

Soil solution in mineral soil was sampled monthly in all chronosequences using suction cup lysimeters. Detailed information on the length of monitoring periods, number of lysimeters at each site, installation depths of lysimeters and number of replicates at each depth is presented on the AFFOREST website ([www.sl.kvl.dk/afforest](http://www.sl.kvl.dk/afforest)). In Denmark and the Netherlands, concentrations measured at 0.9 m depth represent nitrate concentrations in soil water leaching from the root zone. In Sweden, the corresponding depth was 0.6 m. In the Dutch and Swedish chronosequences, lysimeters were placed at randomly selected points within each stand. In Denmark, five lysimeters were installed in each circular subplot, as described above. In all three countries, the lysimeters were installed

about two to six months prior to the first sampling occasion. Soil water was extracted and discarded once or on several occasions before sampling in order to allow the lysimeters to equilibrate with the soil solution. In the Danish and Swedish chronosequences, concentrations of elements represent averages over monthly sampling periods. In the Netherlands, soil water was collected over 24 hours at each sampling occasion. Lysimeter samples at each site and depth were pooled before analysis. In Denmark, composite samples from each circular subplot and depth were analyzed separately.

#### 2.4. Chemical analysis

In the laboratory, sample preparation was performed within two days from sampling. In Sweden, samples were stored in a freezer (-18°C) until chemical analyses. In Denmark, samples were stored at +4°C and analyzed within one month from sampling. In the Netherlands, samples were likewise stored in a refrigerator but chemical analysis was performed within two days from sampling. In Sweden and the Netherlands, nitrate in precipitation, throughfall and soil solution was analyzed by flow injection analysis (FIA), whereas nitrate was analyzed by ion chromatography in Denmark. In all countries, ammonium was analyzed by FIA. In Sweden, dissolved organic carbon (DOC) was analysed by a total organic C analyser (TOC-5000, Shimadzu).

In Denmark and Sweden, N concentrations in litterfall, forest floor and soil were determined by dry combustion (Dumas method) in a Leco CSN Analyzer (Matejovic 1993). The Dutch litterfall, forest floor and soil samples were analyzed for N by wet oxidation according to the Kjeldahl method (Hesse 1971). More detailed descriptions of the analysis of soils in Denmark and Sweden may be found in Vesterdal et al. (2002), Ritter et al. (2003) and Chapter 3, respectively.

#### 2.5. Calculations and statistics

##### 2.5.1. Total deposition and canopy exchange modelling

Throughfall is considered an underestimate of the total deposition of N since N is taken up in the canopy. The total N deposition to the forest ecosystem has been 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 discrimination between canopy exchange and atmospheric deposition using long-term throughfall and precipitation fluxes. Dry deposition and canopy leaching of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{K}^+$  is computed by means of the so-called filtering approach, assuming a fixed relationship between wet and dry deposition of particles taking  $\text{Na}^+$  as a tracer (Ulrich 1983). The total canopy uptake of  $\text{H}^+$  and  $\text{NH}_4^+$  is assumed to equal the total canopy leaching of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{K}^+$  minus canopy leaching of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{K}^+$  associated with foliar excretion of weak acids. Based on experiments in the laboratory, Van der Maas et al. (1991) assumed that  $\text{H}^+$  has an exchange capacity six times larger than  $\text{NH}_4^+$ . Draaijers & Erisman (1995) and

Draaijers et al. (1998) assumed canopy uptake of  $\text{NO}_3^-$  to be negligible. However, this assumption may not be true, particularly in low deposition areas (de Vries et al. 2001). In the present study, canopy exchange of  $\text{NO}_3^-$  was accounted for and calculated according to the slightly adapted canopy budget model presented by de Vries et al. (2001).

The total deposition of N ( $N_{TD}$ ), i.e. the sum of wet and dry N deposition, to the forest ecosystem can thus be calculated as:

$$N_{TD} = N_{TF} + N_{SF} + N_{CE} \quad (1)$$

where  $N_{TF}$  and  $N_{SF}$  is N deposition by throughfall and stemflow, respectively, and  $N_{CE}$  is the exchange of N ( $\text{NH}_4^+ + \text{NO}_3^-$ ) by the forest canopy.

The contribution of stemflow to the total flux to the forest floor varies with tree species and is usually less than 10% of the total flux to the surface (Ivens 1990; Draaijers et al. 1996). Stemflow was not measured at the chronosequences, and the deposition estimates from throughfall measurements are therefore underestimated. At the Dutch and Danish oak sites, the contribution of stemflow to the total N deposition has been approximated using the tree species-specific relationships between throughfall and stemflow as described by Ivens (1990) and de Vries et al. (2001). In the Danish and Swedish spruce chronosequences, stemflow was considered negligible and not accounted for. Rough-barked species, like nearly all conifers, typically have low stemflow values (Augusto et al. 2002; Pypker et al. 2005). For example, stemflow amounts of various nutrients were observed to vary from between 2 to 5% of the amounts by throughfall in a 30-year-old Norway spruce stand in south-western Sweden (Bergholm et al. 2001).

The canopy budget model requires data that meet several quality criteria, including an accurate charge balance between major cations and anions (Draaijers et al. 1996; de Vries et al. 2001). The acceptable percentage difference is less than 10% for bulk deposition and less than 20% for throughfall (WMO 1992). Annual throughfall and bulk precipitation concentration data passed the applied ion balance check at most sites, however, slight discrepancies at some sites (e.g. open field and oak sites at Vestskoven and spruce planted 1972 at Tönnersjöheden) may result in uncertain estimates of total N deposition.

A major uncertainty in the calculations involves the estimation of weak acid excretion in the forest canopies. In the Danish and Dutch oak and spruce stands weak acid exchange fluxes were calculated from measured concentrations of major cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ ,  $\text{H}^+$  and  $\text{NH}_4^+$ ) and anions ( $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ ) in bulk deposition and throughfall. In the Swedish spruce stands, calculation of weak acid exchange was based on DOC and pH measurements for the same compartments according to a method described by de Vries et al. (2001). In the Danish bulk precipitation samples, the sum of major anions often exceeded the sum of major cations, which resulted in unreliable estimates of weak acid exchange fluxes and thereby subsequent errors in the calculation of canopy N uptake. It was, therefore, assumed that the contribution of weak acids from atmospheric deposition (assumed to equal twice the bulk deposition in the canopy budget model) was negligible (zero), and that weak acids found in throughfall were exclusively



derived by leaching from the canopy. This assumption was supported by field observations at the Swedish and Dutch sites of considerable enrichment of DOC in throughfall compared to the bulk deposition (Rosenqvist et al. 2006a). For comparative reasons this assumption was made at all chronosequence sites in Denmark, Sweden and the Netherlands. Excluding the contribution from weak acids in atmospheric deposition leads to slight overestimation of the calculated canopy leaching fluxes of weak acids, and thus to a slight underestimation of the total canopy N uptake. At the Swedish sites, this resulted in about 1-1.5 kg ha<sup>-1</sup> yr<sup>-1</sup> less canopy N uptake compared to if atmospheric deposition of weak acids was accounted for.

Element budgets were calculated by subtracting the leaching flux from the total deposition, in which total deposition fluxes were derived by the canopy exchange model.

### 2.5.2. Soil content and litterfall flux

Soil N dynamics (N contents and C/N ratios) are reported as the relationships with stand age. Forest floor N contents were calculated by multiplying N concentrations with forest floor mass. For the mineral soil N contents the fraction  $\geq 2$ mm were neglected (McNabb et al. 1986; Homann et al. 1995), and soil N contents ( $N_{\text{soil}}$ ) in [Mg ha<sup>-1</sup>] for the soil layers to 25 cm depth were calculated using

$$N_{\text{soil}} = \rho_i \cdot (1 - (\delta_{i, 2\text{mm}}/100)) \cdot d_i \cdot N_i \quad (2)$$

where  $\rho_i$  is the bulk density of the  $< 2$  mm fraction in g cm<sup>-3</sup>,  $\delta_{i, 2\text{mm}}$  is the relative volume of the fraction  $\geq 2$ mm (%),  $d_i$  denotes the thickness of layer  $i$  (cm), and  $N_i$  denotes the N concentration (mg g<sup>-1</sup>) in layer  $i$ .

The litterfall N flux was quantified by multiplying N concentrations with annual litterfall mass, and C/N ratios were based on the ratio between concentrations of C and N.

### 2.5.3. Hydrological modelling

Soil water fluxes were simulated using an extended version of the dynamic simulation model SWAP (Van Dam et al. 1997). Annual fluxes were calculated by summation of monthly fluxes. Detailed information on the hydrological models and their parameterisation is presented in Chapter 3 as well as in van der Salm et al. (2005) and Rosenqvist et al. (2006b). In Denmark and Sweden, the leaching of nitrate from the root zone (i.e. at 0.9 m depth in Denmark and at 0.6 m depth in Sweden) was calculated from measured monthly concentrations times estimated monthly soil water fluxes at corresponding depths. In the Netherlands, 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.

#### 2.5.4. Statistics

Relationships between stand age and soil N contents were explored by simple linear regression. 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-yr old stand in Denmark was not included in regressions, but was included in figures for comparison.

### 3. RESULTS

#### 3.1. N input to the afforested ecosystem

The total volume of bulk precipitation was presented in Chapter 3 and the bulk N deposition in the open field is presented in Table 4.2 to illustrate the difference between countries and the significance of annual variation. The volume of bulk precipitation and its chemical composition vary considerably from location to location but also from year to year. Moreover, the annual variability in volume and N deposition is not related. The lowest average precipitation was observed at Vestskoven located at Zealand in the eastern part of Denmark. The highest average precipitation was registered in the western part of Denmark at the site Gejlvang and in Sweden at Tönnersjöheden. The annual bulk N deposition was markedly lower at Vestskoven, Denmark, compared to the other sites where the annual bulk N deposition was quite high and alike. The highest bulk N deposition was observed at the Danish site Gejlvang during the first measurement year.

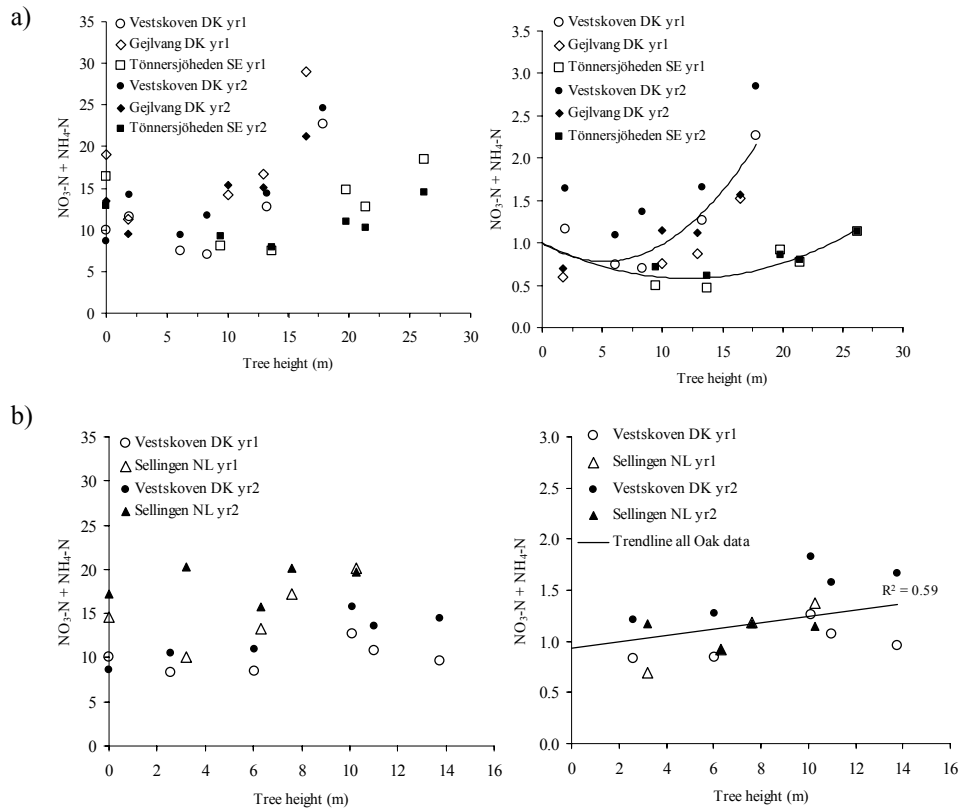
For all sites and tree species, the throughfall deposition of N increased with stand height (and age) (Figure 4.1, Table 4.3). This was most evident when the throughfall deposition of N was expressed relative to the bulk N deposition (Figure 4.1). The taller the stand, the higher the input by throughfall except in the very young stands where throughfall deposition sometimes was lower than in bulk deposition. Throughfall deposition of N in the oak stands did not exceed the bulk deposition in the open field until ~7 m height (Figure 4.1b, Table 4.3) but the N deposition was approximately twice as high for the tallest and oldest stands (25-32 years old) than for the younger stands (3-13 years old). In general, N throughfall deposition did not reach as high levels in the oak stands ( $20 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ) at Selligen in the Netherlands as in the Norway spruce stands ( $25 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ) at the Danish and Swedish chronosequences (Table 4.3).

When throughfall deposition of N in the spruce chronosequences was expressed relative to the bulk deposition two clear country-specific patterns emerge. The relative N deposition is considerably higher in the Danish spruce stands than in the Swedish stands, especially for the taller trees (> 10 m) (Figure 4.1a). For oak, a consistent pattern of increasing N deposition with height was observed, independent of the site location (Figure 4.1b).

**Table 4.2.** Annual total bulk deposition of N ( $\text{kg N ha}^{-1} \text{yr}^{-1}$ ) to the open field at the chronosequences for two years.

Country	Location	Total bulk deposition of N <sup>a)</sup> ( $\text{kg N ha}^{-1} \text{yr}^{-1}$ )	
		Year 1	Year 2
NL	Sellingen	15	17
DK	Vestskoven	10	9
	Gejlvang	19	14
SE	Tönnersjöheden	16	13

<sup>a)</sup>  $\text{NO}_3\text{-N} + \text{NH}_4\text{-N}$



**Figure 4.1.** Throughfall N ( $\text{NH}_4\text{-N} + \text{NO}_3\text{-N}$ ) deposition ( $\text{kg ha}^{-1} \text{yr}^{-1}$ ) (left) and throughfall N deposition relative to open field deposition of N (right) as a function of tree height (m) for the (a) Norway spruce and (b) oak chronosequences. The sampling periods (Year 1 & 2) are given in Table 4.1.

**Table 4.3.** The calculated total N deposition ( $N_{TD}$ ), measured total N throughfall ( $N_{TF}$ ), and calculated uptake of N ( $N_U$ ) in the canopy (in  $\text{kg ha}^{-1} \text{yr}^{-1}$ ) for Norway spruce and oak chronosequences in Sweden, Denmark and the Netherlands. Data are annual average values based on the following sampling periods: Tönnersjöheden (spruce), Nov 01-June 03; Vestskoven (spruce and oak), Jan 01-Dec 02; Gejlvang (spruce), Apr 01-Mar 03; Sellingen (oak), Apr 01-Dec 02.  $BD_N$  refers to the bulk N deposition at each location.

Location	Age	Height	$N_{TD}$	$N_{TF}$	$N_U$	$N_U$ in % of $N_{TD}$
	yr	m	$\text{kg ha}^{-1} \text{yr}^{-1}$	$\text{kg ha}^{-1} \text{yr}^{-1}$	$\text{kg ha}^{-1} \text{yr}^{-1}$	%
<i>Norway Spruce</i>						
Tönnersjöheden	$BD_N$		14.5			
SE	19	10.1	15.0 <sup>1)</sup>	10.1	4.3	29
	30	15.5	14.8 <sup>1)</sup>	8.4	4.3	29
	65	19.8	15.1 <sup>1)</sup>	12.6	1.2	8
	74	22.4	14.7 <sup>1)</sup>	11.6	1.6	11
	92	27.4	19.3	18.1	1.2	6
Vestskoven	$BD_N$		9.5			
DK	5	1.8	20.0	13.1	7.1	35
	12	6.1	11.1	8.7	2.0	18
	14	8.4	12.7	10.0	3.0	24
	29	13.9	19.8	14.1	5.9	30
	33	17.8	27.0	23.7	3.5	13
Gejlvang	$BD_N$		16.1			
DK	8	1.4	~16.1 <sup>1)</sup>	10.4	3.0	35
	21	10.4	24.1	14.2	9.9	41
	26	12.8	27.7	16.9	10.8	39
	42	16.1	32.8	25.2	7.7	23
<i>Oak</i>						
Vestskoven	$BD_N$		9.5			
DK	9	2.6	15.5	10.6	3.4	22
	14	6.0	14.8	10.4	3.0	20
	25	10.1	21.2	12.9	6.5	31
	23	11.0	22.7	14.7	6.0	26
	32	13.6	24.1	13.2	9.1	38
Sellingen	$BD_N$		15.9			
NL	4	3.2	18.2	15.1	0.9	5
	7	6.3	18.7	14.5	2.2	12
	11	7.6	27.4	18.7	6.2	23
	18	10.3	33.5	19.8	10.9	33

<sup>1)</sup> At these sites,  $N_{TD}$  was estimated by summation of wet and dry deposition of N ( $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$ ) where dry deposition of  $\text{NH}_4\text{-N}$  was set to be zero. This approximation was made since canopy exchange modelling resulted in negative dry deposition of  $\text{NH}_4\text{-N}$  and levels of  $N_{TD}$  slightly lower than the measured bulk N deposition. As a consequence, the sum of  $N_{TF}$  and  $N_U$  (stemflow of N assumed negligible in spruce) is slightly lower than the given values of  $N_{TD}$ .

The calculated total N deposition as well as the calculated uptake of N in the canopy is shown in Table 4.3 for both spruce and oak chronosequences in the three countries. The total N deposition in the Swedish spruce stands was only little

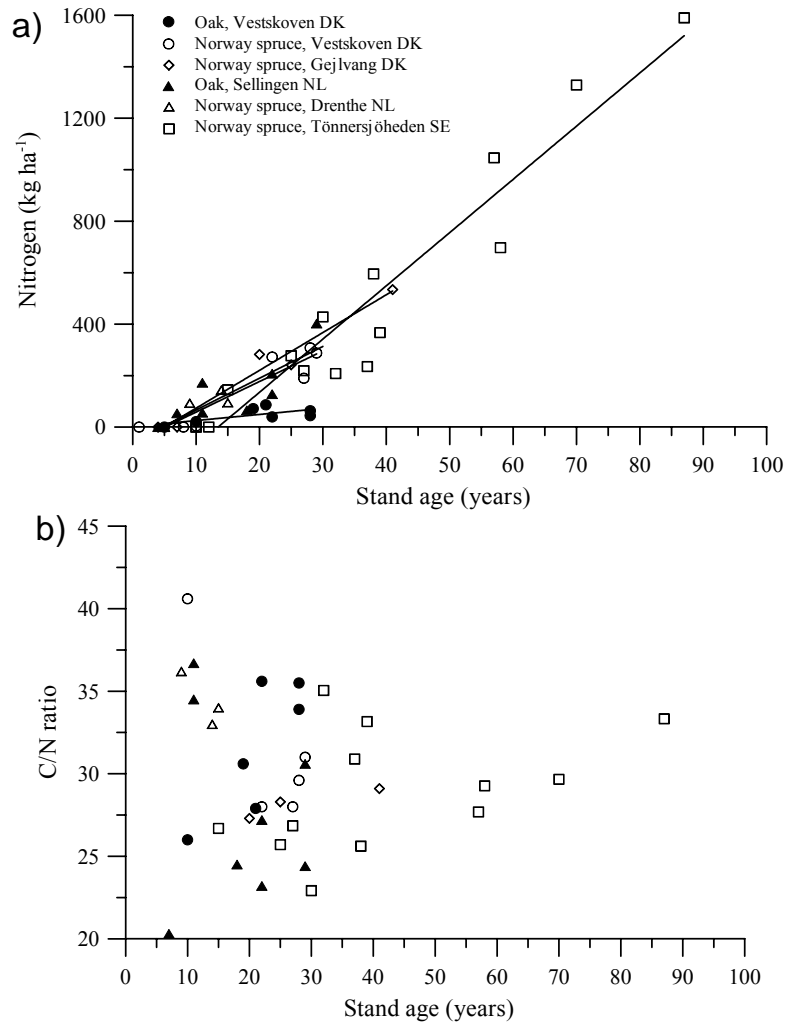
higher than the bulk deposition whereas the total N deposition was substantially higher than the bulk N deposition at the Danish and Dutch stands. Here, also a clear increase in total N deposition with stand height was observed. The total N deposition to the spruce chronosequences was highest at Gejlvang in Denmark ranging between 16-33 kg ha<sup>-1</sup> yr<sup>-1</sup> while the total N deposition to the spruce stands at Tönnersjöheden only reached a maximum value of 19 kg ha<sup>-1</sup> yr<sup>-1</sup>. At both the Dutch and Danish oak sites, the total N deposition increased with stand height. The highest total N deposition for oak chronosequences was observed in Sellingen in the Netherlands where it reached a value of 34 kg ha<sup>-1</sup> yr<sup>-1</sup>. At Vestskoven in Denmark, the highest total N deposition was 24 kg ha<sup>-1</sup> yr<sup>-1</sup>.

The measured throughfall N fluxes differed from the total N deposition fluxes as a result of canopy exchange processes (Table 4.3). Canopy uptake of N in Norway spruce varied from 1-11 kg ha<sup>-1</sup> yr<sup>-1</sup> or 6-41% of the total N deposition (Table 4.3). The canopy uptake of N decreased with age for the Swedish spruce sites but no clear pattern was observed for the Danish spruce stands. The Swedish chronosequence is more complete spanning from open field to 90 years of age. It is possible that a similar pattern would be observed if older Danish spruce stands were included in the study. For Norway spruce, the largest uptake was estimated at Gejlvang in Denmark. Canopy uptake of N in oak likewise varied from 1-11 kg ha<sup>-1</sup> yr<sup>-1</sup> or 5-38% of the total N deposition. In contrast to the spruce stands, the canopy uptake of N in the oak stands increased with age both in the Dutch and the Danish oak chronosequences.

### 3.2. Changes in soil N status and litterfall N flux

Immediately after afforestation there is no forest floor developed and N is only present in the mineral soil. Nitrogen accumulation in the forest floor started to develop rapidly after canopy closure (10-20 years, Figure 4.2a). At Vestskoven, oak accumulated very little N in forest floors over 30 years (85 kg ha<sup>-1</sup>), whereas spruce and oak at Sellingen stored more N in forest floors within 30 years (ca. 3-400 kg ha<sup>-1</sup>). Large amounts of N were accumulated in the oldest stands of the Swedish spruce chronosequence (around 1600 kg ha<sup>-1</sup>). The forest floor N accumulation rate was highest for the Swedish chronosequence and lowest for oak at Vestskoven (Table 4.4).

These patterns were mainly driven by the accumulation of forest floor mass as there was little age-related change in forest floor C/N ratios in most chronosequences (Figure 4.2b). Only in the oak chronosequence at Vestskoven and spruce chronosequences at Gejlvang and in Sweden there were trends indicating increasing forest floor C/N ratios with increasing age.



**Figure 4.2.** a) Forest floor N content (kg ha<sup>-1</sup>) and b) C/N ratio in forest floor in oak and Norway spruce chronosequences. Regression equations are given in Table 4.4.

Mineral soil N contents were unchanged or tended to decrease slightly with increasing stand age in most chronosequences (Figure 4.3a and Table 4.4). Mineral soil N significantly decreased in the Swedish chronosequence. An exception to this was the significantly increasing mineral soil N content in the Dutch chronosequence. The Danish chronosequence at Gejlvang was remarkably lower in mineral soil N content regardless of stand age (around 2 Mg ha<sup>-1</sup>) whereas the other Danish chronosequence at Vestskoven and the Swedish chronosequence had initial soil N contents of 5-6 Mg ha<sup>-1</sup>. There was no difference between the Danish spruce and oak chronosequences at Vestskoven (Figure 4.3, Table 4.4), so all data were

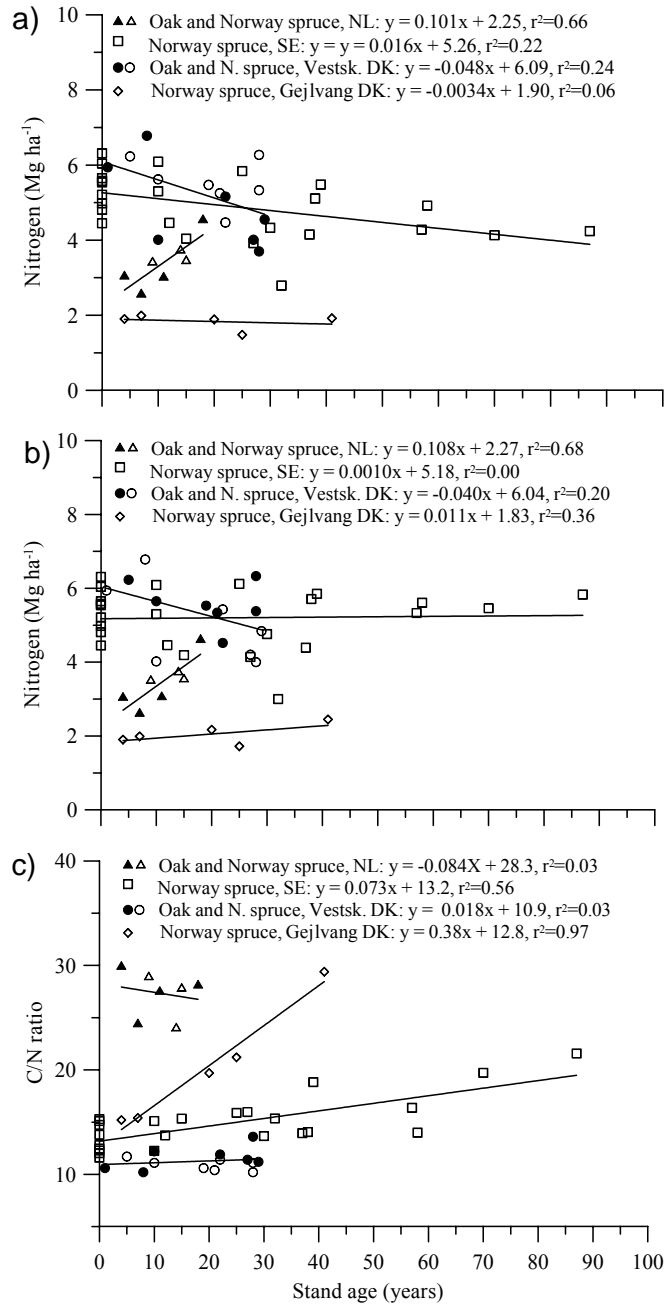
combined in the analysis of soil N (Ritter et al. 2003). For the studied soil compartments as a whole, i.e. the forest floor and the former plow layer (0-25 cm), there were patterns of constant, decreasing or increasing soil N content over 30-90 years after afforestation (Figure 4.3b). The only significant trend was the increasing soil N content in the Dutch chronosequence.

In the Danish Gejlvang chronosequence and in the Swedish chronosequence, mineral soil N content clearly decreased relative to C with increasing stand age whereas C/N ratios were unchanged in the Danish Vestskoven chronosequence. The chronosequence at Vestskoven also had the lowest C/N ratios regardless of stand age. The Danish and Swedish chronosequences had relatively similar C/N ratios of 10-15 for arable fields and stands younger than ten years. However, the development in C/N ratios differed strongly between the sites. The Dutch chronosequence differed from the other chronosequences by the much higher C/N ratios. There was no significant change along this chronosequence, which covers a time span of only 14 years.

**Table 4.4.** Rates of soil N sequestration (SE of regression slope) over 30-90 years in the AFFOREST chronosequences.

Site	Forest floor N		Mineral soil N*		Total soil N*	
	Rate kg ha <sup>-1</sup> yr <sup>-1</sup>	P value	Rate kg ha <sup>-1</sup> yr <sup>-1</sup>	P value	Rate Mg ha <sup>-1</sup> yr <sup>-1</sup>	P value
Oak, Vestskoven DK	2.4 (1.1)	0.088	-48.2 (24.3)	0.071	-40.3 (23.4)	0.111
Spruce, Vestskoven DK	11.9 (2.2)	0.003				
Spruce, Gejlvang DK	14.7 (1.7)	0.003	-3.4 (7.6)	0.683	11.2 (8.6)	0.287
Spruce SE	20.7 (1.7)	< 0.001	-15.8 (6.3)	0.020	9.7 (7.3)	0.194
Oak/spruce NL	12.1 (2.3)	< 0.001	105.3 (33.8)	0.026	108.2 (33.2)	0.023

\* Rates of soil N change for mineral soil and total soil C at Vestskoven are combined for oak and spruce.

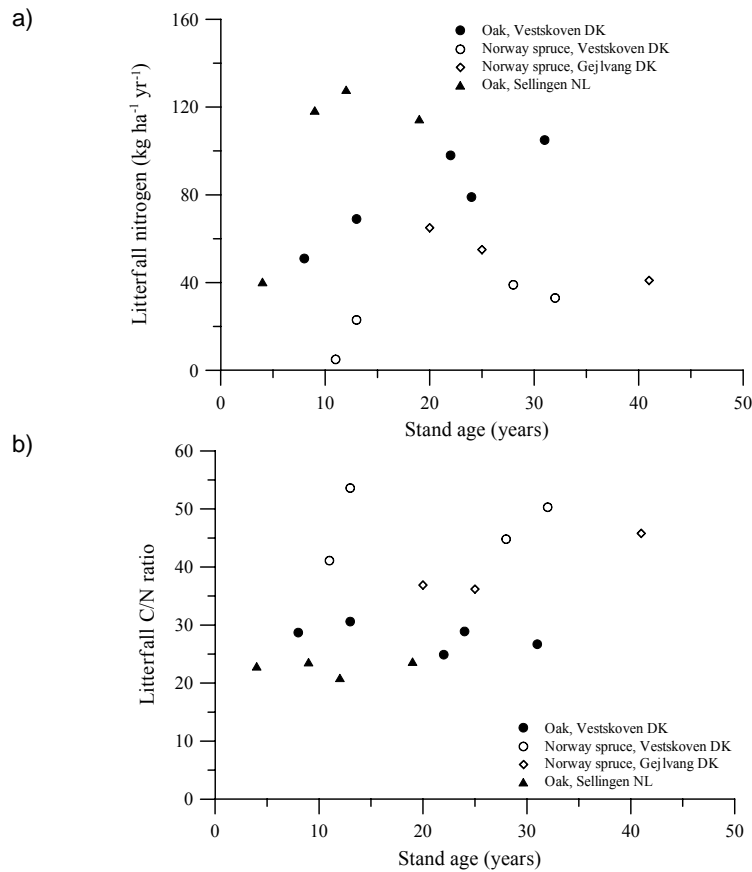


**Figure 4.3.** a) N content in mineral soil, b) N content in both mineral soil and forest floor, and c) C/N ratios in mineral soil.



There were no general trends in litterfall N flux with increasing stand age (Figure 4.3a) except that the youngest stands tended to have lower litterfall N fluxes. For oak at Vestskoven, litterfall N fluxes appeared to increase gradually with stand age. Litterfall N flux was higher in oak stands and lower in spruce stands. The few age-related dynamics were not caused by changes in litter N concentrations with stand age, as litterfall C/N ratios were quite similar along the chronosequences (Figure 4.3b).

Site- and probably also species-related differences in C/N ratio were more apparent than an effect of stand age. Oak stands had lower litterfall C/N ratios than did spruce. The oak chronosequence at Sellingen had the lowest C/N ratios (20-25) and spruce at Vestskoven had the highest C/N ratios (40-50).



**Figure 4.4.** a) Litterfall N flux ( $\text{kg ha}^{-1} \text{yr}^{-1}$ ) and b) C/N ratio of litterfall in oak and Norway spruce chronosequences.

The difference in litter C/N ratios partly contributed to the species difference in litterfall N flux, but litterfall mass and C content were also somewhat higher in oak than in spruce (Chapter 2).

### 3.3. Nitrate leaching following afforestation

At 90 cm depth, virtually all mineral N in soil solutions was found as nitrate. In the six chronosequences in Denmark, the Netherlands and Sweden, soil solution nitrate concentrations beneath the root zone were generally below the threshold value of  $50 \text{ mg dm}^{-3} \text{ NO}_3^-$  for groundwater to be utilised as drinking water as expressed in the EU Water Framework Directive. Exceptions were consistently elevated concentrations recorded under the 33-year-old (planted 1969) spruce stand at Vestskoven in Denmark, where the concentrations frequently exceeded the drinking water quality requirements.

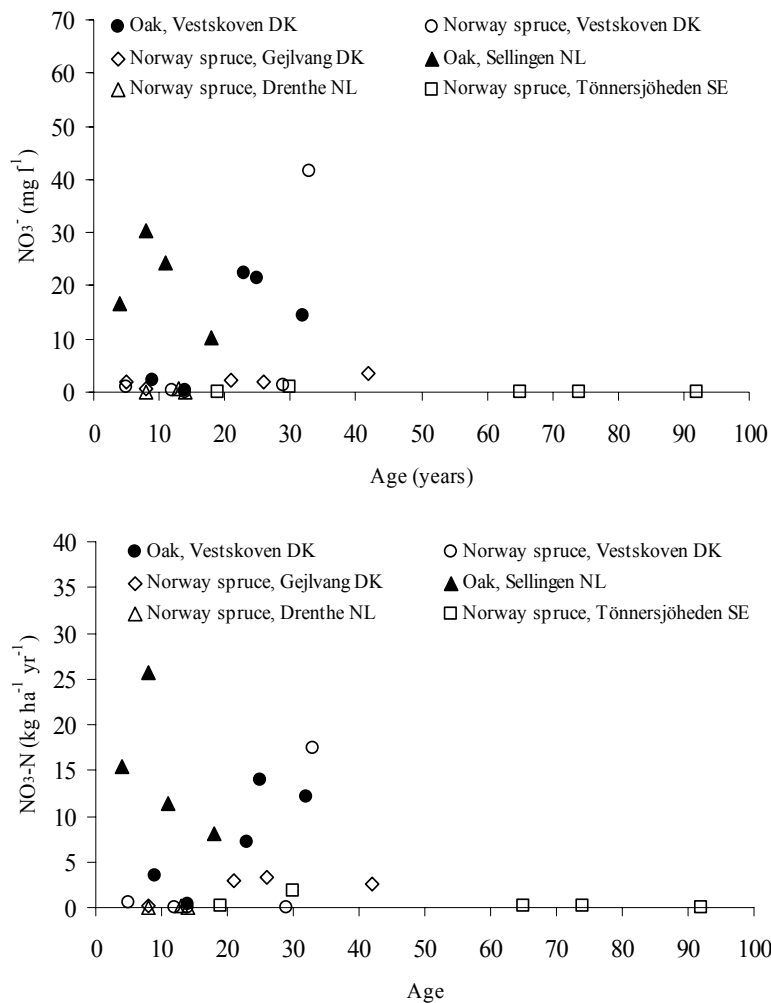
On an annual basis, high leaching losses of nitrate were related to high concentrations of nitrate in leaching water (Figure 4.5a, b). There was no clear relationship between nitrate leaching and stand age (Figure 4.5b) when data from all three countries were evaluated together. However, when looking at the countries separately, differences became apparent. In Denmark, leaching of nitrate from the root zone tended to be lower in the early phase of afforestation and increased after canopy closure. Here, nitrate losses by leaching only occurred in stands older than 20 years, whereas nitrate leaching always was low or negligible in stands younger than 15 years. In contrast to the Danish sites, there was a tendency of decreased nitrate leaching with forest age along the Dutch oak chronosequence at Sellingen. Furthermore, at Drenthe in the Netherlands and at Tönnersjöheden in Sweden, seepage water nitrate concentrations and leaching of nitrate from the root zone were generally negligible across the Norway spruce chronosequences, although slightly elevated levels were recorded in the 30-year-old stand (planted 1972) at Tönnersjöheden during summer periods.

During the measurement period, N budgets (i.e. total inorganic N deposition minus  $\text{NO}_3^-$ -N leaching) ranged from  $-7$  to  $31 \text{ kg ha}^{-1} \text{ yr}^{-1}$  (Figure 4.6). At all except for one site (the 8-year-old oak stand at Sellingen), N leaching was lower than the estimated total N input. Nitrogen budgets tended to increase with age along the chronosequences at Gejlvang and Sellingen. In the chronosequences at Tönnersjöheden and Vestskoven, N budgets appeared relatively constant and showed no distinct trends with age. Within Denmark, higher N retention was observed in closed forest stands ( $> 20$  years) growing on nutrient-poor sandy soil (Gejlvang) compared to stands of similar age on nutrient-rich clayey soil (Vestskoven).

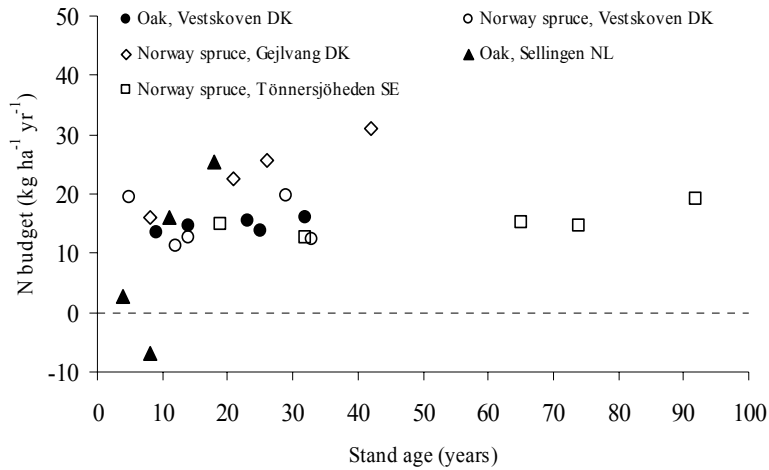
Leaching of nitrate was evident in some stands at a throughfall N deposition level of approximately  $8\text{-}10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ , and above a throughfall deposition level of approximately  $15 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  the majority of stands leached nitrate (Figure 4.7).

Over the age spans covered by the chronosequences, oak stands generally leached more nitrate than the spruce stands (Figure 4.5b). The highest leaching losses were recorded along the oak chronosequence at Sellingen in the

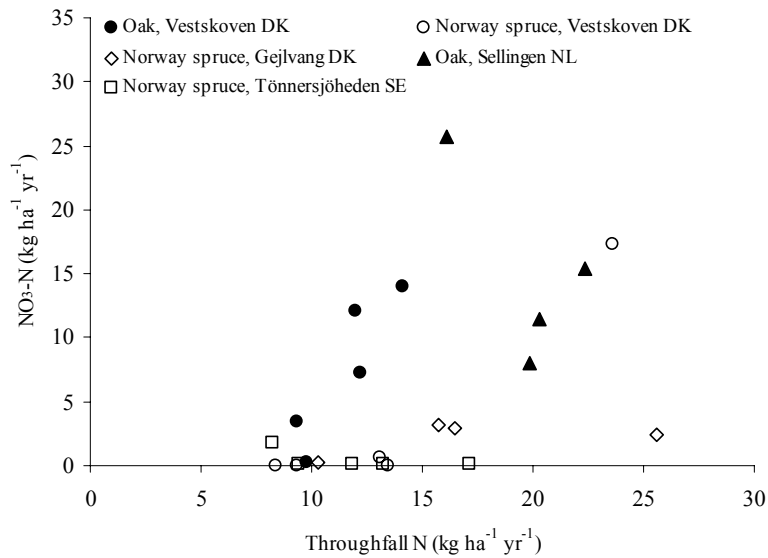
Netherlands, where nitrate leaching reached a level of  $26 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  in the 8-year-old stand. In contrast, nitrate leaching was negligible below the young Dutch spruce stands at Drenthe. At Vestskoven in Denmark, nitrate leaching appeared at an earlier age in the oak chronosequence (Figure 4.5b) and at a lower N throughfall deposition level (Figure 4.7) compared to the spruce chronosequence. Leaching was low below a throughfall deposition level of  $10\text{--}12 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  in the oak stands and below  $14 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  in the spruce stands.



**Figure 4.5.** (a) Annual average nitrate concentration ( $\text{mg dm}^{-3}$ ) in soil water at 0.9 m (Tönnersjöheden, SE: 0.6 m) and (b) average nitrate leaching ( $\text{kg NO}_3\text{-N ha}^{-1} \text{ yr}^{-1}$ ) from the root zone in 2002 as a function of time since afforestation in the six AFFOREST chronosequences.



**Figure 4.6.** Nitrogen budgets ( $\text{kg ha}^{-1} \text{yr}^{-1}$ ), i.e. total inorganic N deposition minus  $\text{NO}_3^-$ -N leaching at the bottom of the root zone, as functions of time since afforestation in five chronosequences. Positive budgets indicate that inorganic N is retained in the ecosystem, whereas negative budgets indicate a net release of inorganic N.



**Figure 4.7.** Mean annual leaching of nitrate-N ( $\text{kg ha}^{-1} \text{yr}^{-1}$ ) from the root zone in relation to throughfall N flux ( $\text{kg ha}^{-1} \text{yr}^{-1}$ ) for corresponding periods in five AFFOREST chronosequences. Data are for the year 2002.

## 4. DISCUSSION

### 4.1. Water quality before and after afforestation

The average nitrate concentrations below the root zone of sandy arable land in Denmark were close to the drinking water threshold value of  $50 \text{ mg dm}^{-3} \text{ NO}_3^-$ , while it was estimated to be  $21 \text{ mg dm}^{-3} \text{ NO}_3^-$  on clayey soils in average (Grant et al. 2004). The concentrations at the afforested stands were generally lower except for the 33-year-old spruce stand at Vestskoven. In the Netherlands, Fraters et al. (2004) showed average nitrate concentrations in groundwater beneath sandy arable land to be  $75 \text{ mg dm}^{-3} \text{ NO}_3^-$  and  $40 \text{ mg dm}^{-3} \text{ NO}_3^-$  beneath clayey arable land. Seepage water N concentrations reported for Swedish arable soils are similar to concentrations reported for arable soils in Denmark. Johnsson & Mårtensson (2002) estimated seepage water nitrate concentrations under arable land in southwest Sweden to be in average  $57 \text{ mg dm}^{-3} \text{ NO}_3^-$  for sandy soils and  $20 \text{ mg dm}^{-3} \text{ NO}_3^-$  for clay soils, which is substantially higher than concentrations recorded along the Swedish chronosequence (Figure 4.5a). As expected, the measured seepage water nitrate concentrations in the afforested stands were mostly lower than concentrations previously reported for arable soils in the three countries.

A high nitrate concentration in the soil solution seems to be a pre-condition for nitrate leaching (Figure 4.5a, b) and may indicate loss of nutrients from the ecosystem. In the Netherlands, nitrate leaching was estimated to be  $74 \text{ kg NO}_3\text{-N ha}^{-1} \text{ yr}^{-1}$  on sandy soils under agricultural crops (Rijtema & de Vries 1994). For the whole arable area of Denmark, a leaching rate of 106 and  $55 \text{ kg NO}_3\text{-N ha}^{-1} \text{ yr}^{-1}$ , respectively, were reported from sandy and clayey soils (Grant et al. 2004). Nitrate leaching below arable land in southwest Sweden (province of Halland) was estimated to be  $63 \text{ kg NO}_3\text{-N ha}^{-1} \text{ yr}^{-1}$  from sandy soils and  $18 \text{ kg NO}_3\text{-N ha}^{-1} \text{ yr}^{-1}$  from clayey soils, whereas an average value of  $22 \text{ kg NO}_3\text{-N ha}^{-1} \text{ yr}^{-1}$  was estimated for all arable land in Sweden (Johnsson & Hoffmann 1998; Johnsson & Mårtensson 2002). In general, the simulated N leaching fluxes from the root zone in the chronosequences (Figure 4.5b) were considerably lower than the simulated N leaching from arable land in the three countries. A shift in land use from agriculture to forestry therefore presumably led to decreased nitrate leaching from the root zone. However, nitrate leaching from plantations on former arable land might still be higher than that from old forests. In Denmark, higher concentrations of nitrate in soils below the root zone (75-100 cm) were observed in recently afforested land (<10 years) compared to old forest ecosystems (Callesen et al. 1999).

In the Danish chronosequences at Vestskoven and Gejlvang, stands younger than 15 years showed practically no nitrate leaching, whereas leaching tended to increase after a stand age of around 20 years (Figure 4.5b). This may reflect higher uptake of N by trees in the early phase after afforestation due to the formation of N-rich compartments. N uptake by the canopy in the youngest stands is supported by the observation that the throughfall N deposition was lower than the bulk N deposition. When the foliar canopy is complete (about 15-20 years after planting) the trees can no longer take up the available N pool, and excess N is leached to

seepage water. Increased nitrate leaching after canopy closure could also partly be attributed to increased N input with increased height (Figure 4.1, Table 4.3). Increased nitrate leaching with stand age has earlier been observed for a Sitka spruce stand in Wales (Emmet et al. 1993). The influence of stand height on N leaching has been described for Vestskoven by Hansen et al. (2006b).

At Vestskoven in Denmark, the highest concentrations of nitrate in seepage water and highest levels of nitrate leaching were encountered in the 33-year-old spruce stand (planted in 1969). Concentrations and leaching levels were negligible in the other spruce stands along the chronosequence (Figure 4.5a, b). Several factors may explain this deviant behaviour. Compared to the younger stands in the chronosequence, the annual input of N with throughfall was considerably larger in the 33-year old stand due to its larger tree height (Figure 4.1, Table 4.3) resulting in enhanced dry deposition of N. Moreover, lower uptake rates of N relative to the younger stands could have resulted in higher leaching losses of nitrate from this stand. A high rate of net N mineralization may indeed be expected in the nutrient-rich soil at Vestskoven because of high soil N contents and low C/N ratios (around 10-12) in the former plow layer (Figure 4.3a, c). Possibly, recent thinning (1998) in the stand may have resulted in increased amounts of mineral N relative to other spruce stands in the chronosequence. An increase in soil nitrate concentrations was visible in a thinning experiment in lodgepole pine where 60% of the trees were removed (Knight et al. 1991). Bäumler & Zech (1999) also observed a low increase in nitrate concentrations after a 40% thinning. The concentrations were back to pre-cutting conditions after one year.

Maintenance of high rates of N mineralization in soil in the early phase after the conversion of farmland to forest may explain the high levels of nitrate leaching beneath the oak stands at Selligen in the Netherlands (van der Salm et al. 2005) (Figure 4.5b). In addition, throughfall N deposition was quite high to the oak stands (Table 4.3, Figure 4.7). The rates of N mineralization are, as discussed earlier, likely to decline following afforestation. A decreased mineralization rate with age may partly explain the decreasing trend in nitrate leaching along the 18-year chronosequence at Selligen. A very possible contributing factor may be an increase in the rate of N uptake with age of the young aggrading oak forest. However, the stands in the Dutch chronosequence are all below the age of 20 years and it is possible that leaching of nitrate will increase when the stands mature and scavenging of atmospheric N increases. In addition, denitrification and the subsequent release of N<sub>2</sub> might be an important process that removes nitrate from the soil solution under the oak stands at Selligen due to anaerobic conditions. The groundwater level in the forest fluctuated between -20 cm in wet periods in winter to -160 cm in summer.

No age-related trends in nitrate leaching were found along the Norway spruce chronosequences at Drenthe in the Netherlands and at Tönnersjöheden in Sweden, since N leaching was generally negligible at these sites. Here, trees and field layer were able to take up all available N. Even the old Norway spruce stands (64-92 years old) at Tönnersjöheden were able to completely retain N in the ecosystem (Figure 4.5a, b) in spite of a relatively high ambient N deposition in the region (Hallgren-Larsson 2002) and an anticipated low demand for N for growth in these

mature stands. The high capacity of the old spruce stands to retain N may partly be caused by less intensive fertilisation practices during former agriculture compared to soils more recently abandoned and compared to soils abandoned in Denmark and the Netherlands. Also, the old plantations experienced a lower level of N deposition in their early growth stages than more recently planted forests (Lövblad 2000). These old stands also had abundant field layer vegetation consisting mainly of mosses, which most likely contributed to a higher uptake of N in biomass. Since the majority of Swedish forest ecosystems are N limited (Näsholm et al. 2000), low levels of nitrate leaching were anticipated. However, Andersson (2002) suggested that N saturation might be a potential problem in forests in southwest Sweden, where N deposition is relatively high ( $>15\text{ kg N ha}^{-1}\text{ yr}^{-1}$ ). In this respect, recently afforested ecosystems would be particularly vulnerable to disturbance of the N cycle and to increased N inputs.

Several authors observed nitrate leaching to occur mainly above a threshold value of about  $10\text{ kg N ha}^{-1}\text{ yr}^{-1}$  in throughfall input (Dise et al. 1998; Gundersen et al. 1998; Kristensen et al. 2004), but at high deposition rates the leaching seemed to be largely dependent on ecosystem properties (e.g. soil type, N status etc). In old-growth forest ecosystems several indices have been used in order to predict the onset of N saturation and the potential risk of nitrate leaching. Especially, the C/N ratio of the forest floor, total throughfall N flux and foliage N concentration are well established predictors of the likeliness of N leaching from old European forest ecosystems (Dise et al. 1998; Gundersen et al. 1998; Kristensen et al. 2004). It appears as if the throughfall N flux could be used as a predictor of nitrate leaching even in recently afforested stands (Figure 4.7). However, it is not possible from our data to identify whether increased nitrate leaching after canopy closure is the result of increased throughfall N input or decreased N uptake and changing mineralization or a combination of these and possibly other factors. The forest floor C/N ratio was not found to be a useful predictor in young aggrading forests planted on former arable soils, since the C/N ratio of the thin forest floors to a large extent reflects the quality of recently shed litter.

#### 4.2. Site effects on deposition and nitrate leaching

Deposition fluxes of N may show considerably local variation due to differences in e.g. climate, local N sources, stand structure, forest age, forest management and tree species (Bleeker & Draaijers 2002). In our study, the total atmospheric input of inorganic N ( $\text{NO}_3^-$  and  $\text{NH}_4^+$ ) as well as the throughfall N flux to the forests tended to decrease along a south-west (the Netherlands) – north-east (Sweden) gradient (Table 4.3). Such a gradient was, however, not observed for the measured bulk deposition fluxes of N in the open field (Table 4.2 and 3).

Throughfall N deposition in Norway spruce at Tönnersjöheden, Sweden, was considerably lower than at the Danish sites (especially for the taller trees  $> 10\text{ m}$ ) although the deposition of N in the open field was equal or higher at the Swedish location (Table 4.3, Figure 4.1a). The high bulk N deposition is confirmed by a number of Swedish studies (Kindbom et al. 2001; Hallgren-Larsson 2002). The area of Halland, where Tönnersjöheden is situated, is a part of Sweden that

receives among the highest load of atmospheric N. The lower throughfall deposition of N in the Swedish sites relative to the Danish sites was not due to high levels of canopy exchange (Table 4.3) but rather due to a lower contribution of dry deposition of N. A different contribution of dry deposition at the Danish and Swedish sites is not unexpected since the input by dry deposition is larger in Denmark than in Sweden caused by a higher concentration of livestock. Also, the wind climate differs largely between these sites. The Danish sites are more windy than the Swedish sites, which causes dry deposition to increase at the Danish sites relative to the Swedish sites. A strong relationship between the deposition velocity of  $\text{NH}_3$  and the wind velocity has earlier been observed (Bleeker & Draaijers 2002). Furthermore, lower fluxes of dry N deposition along the 90-year-old Swedish chronosequence may partly be caused by a strong thinning regime. These mature spruce stands have more open canopies than the younger more dense stands at the Danish chronosequences. Thinning of a forest stand will limit the rate of atmospheric deposition because of the reduction of the total surface area of the canopy and the accompanying reduction of the aerodynamic roughness of the canopy (Bleeker & Draaijers 2002).

Within Denmark, the deposition regime varied considerably between the stands at Gejlvang in the western part of the country and the stands at Vestskoven in the eastern part. A higher total N deposition at Gejlvang was attributed to higher rates of both wet and dry N deposition compared to Vestskoven (Table 4.3). Both higher wind velocity at Gejlvang, leading to higher dry deposition of N, and a higher local emission of  $\text{NH}_3$  due to a high concentration of livestock led to these high N depositions (Hansen 2003). Our observations of N deposition (Table 4.3) in the older Danish spruce stands (> 20 years) are in good agreement with corresponding N fluxes measured in long term monitoring plots of the same age (level II) located on similar soil types not far from the studied chronosequence stands (Hansen 2003).

Dutch agriculture is more intensive than in Denmark and Sweden in sense of both livestock and the relative area covered by agriculture. As such, the throughfall N deposition to the oak stands was higher at the Dutch stands at Selligen than at Vestskoven in Denmark. High N deposition in the Netherlands in general has been documented earlier (Draaijers 1993; Erisman et al. 2005).

Up to about 40% of the total atmospheric N deposition was taken up by the forest canopy (21 and 26 years old spruce stands at Gejlvang, Denmark) (Table 4.3). This is consistent with a study by Johnson and Lindberg (1992) conducted in several forest stands (mainly conifers) scattered over the U.S., showing that in average 40% of the total inorganic N deposition was retained by the vegetation, whereas 60% was found back in the throughfall as  $\text{NO}_3^-$  and  $\text{NH}_4^+$ . In their study, total inorganic N uptake amounted up to  $850 \text{ eq. ha}^{-1} \text{ yr}^{-1}$  ( $\sim 12 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ), which is close to the maximum calculated average rates of canopy N uptake ( $11 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ) found at Gejlvang. Even higher uptake efficiency of total inorganic N was reported by Tomaszewski et al. (2003) in a subalpine slowly aggrading 90-year-old spruce-fir-pine forest in Colorado, U.S., where approximately 85% of the total N deposition was retained by the forest canopy in the growing season. On the other hand, an investigation performed in a Douglas-fir stand at the Speulder forest in the



Netherlands showed only 9% uptake of the total  $\text{NH}_x$  deposition (Draaijers et al. 1997). However, the total N load to the forest canopies differed substantially among these studies where the Dutch study as well as our data generally showed considerably higher N deposition than the two north American studies. Often the throughfall deposition, especially in the youngest spruce stands, was lower than in bulk deposition, which also points to considerable N uptake in the canopies in the early years after afforestation.

Canopy uptake of N has earlier been estimated in the Dutch oak chronosequence at Sellingen during the same sampling period using a slightly different calculation approach (Van der Salm et al. 2005). In their study, the uptake of N was relatively constant ( $5.9\text{--}6.7 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ ) and showed no distinct trend with age. In contrast, our results suggest an increasing trend with age (and height) from 1 to  $11 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  between 4 and 18 years stand age (Table 4.3). The quantitative results therefore seem to be dependent on the assumptions taken during the calculation. However, in this study the calculations were performed alike on all data and comparisons between sites are justified.

Changes in soil C and N levels and N dynamics following afforestation of former arable land were very site specific. Among the studied sites, differences between nutrient-rich and nutrient-poor sites were reflected by C/N ratios and N contents in the mineral soil (former plow layer), with generally lower soil C/N ratios and higher soil N contents in the more nutrient-rich soils (Figure 4.3). The nitrogen status at the six AFFOREST chronosequences, as suggested by these indicators, increased in the approximate order: oak and spruce, Sellingen and Drenthe, the Netherlands  $\leq$  spruce, Gejlvang, Denmark  $<$  spruce, Tönnersjöheden, Sweden  $<$  oak and spruce, Vestskoven, Denmark.

Nitrate leaching was substantially higher from the nutrient-rich clayey soils of high N status at Vestskoven than from the nutrient-poor sandy soils of lower N status at Gejlvang (Figure 4.5b). This is consistent with previous studies reporting higher nitrate leaching (Hansen 2003) and higher seepage water nitrate concentrations (Callesen et al. 1999) below forest stands on clayey soils compared to stands on sandy soils in Denmark. However, differences in N status between sites expressed as soil C/N ratios and soil N contents could not fully explain the observed patterns of nitrate leaching between all the chronosequences. For example, seepage water nitrate concentrations and nitrate leaching were high below the young oak stands at Sellingen in the Netherlands (Figure 4.5a, b), in spite of low N status (high C/N ratio) in the sandy soils at this site. In the first years following afforestation, nitrate leaching may occur as a result of high production of N by mineralization of N-rich organic matter inherited from the former agricultural land use. Thus, in recently afforested stands the net N mineralization rate or net nitrification rate would presumably be a better predictor of the risk of nitrate leaching than the soil C/N ratio.

Within Denmark, the overall N status of the site had a marked influence on the net retention of N in the afforested ecosystem. In closed stands older than about 20 years, net ecosystem retention of N was more efficient in forests growing on nutrient-poor sandy soil at Gejlvang (western Denmark) than in forests on nutrient-rich clayey soil at Vestskoven (eastern Denmark) (Figure 4.6), regardless of tree

species and despite higher total N deposition at Gejlvang (Table 4.3). The most likely reason for this is higher soil supply rates of N (N mineralization followed by nitrification) at Vestskoven, caused by high soil N contents and pH. Soil and vegetation are unable to take up the available N (derived by mineralization and deposition), which is leached from the root zone.

At the Danish and Swedish chronosequences, mineral soil C/N ratios reflected the former land use within the first decade following afforestation, after which differences between soil types became apparent (Figure 4.3c). Chronosequences with soils of low N status (Gejlvang, Denmark as well as Sellinger and Drenthe, the Netherlands) were able to accumulate C in the mineral topsoil, which can be attributed largely to incorporation of forest derived organic matter with high C/N ratio and partly to reduced rates of decomposition in these nutrient-poor soils. The relatively unchanged (Gejlvang) and increasing (the Netherlands) mineral soil N contents (Figure 4.3) are in contrast to previous studies, which show considerable N depletion in the mineral topsoil after 40-115 years of afforestation with conifers or mixed broadleaves on sandy or loamy soil types in the U.S. (Hamburg 1984; Hooker & Compton 2003; Richter et al. 2000). An indication of depletion in mineral soil N contents after afforestation was observed in soils of higher N status (higher soil N content and lower C/N ratios) at Vestskoven and at Tönnersjöheden.

#### 4.3. Tree species effects on deposition and nitrate leaching

At Vestskoven in Denmark, the oak and spruce chronosequences were established on the same soil type (loamy till) (Chapter 1). Therefore, this site offers a unique opportunity to study the influence of different tree species on deposition and nitrate leaching following afforestation of former agricultural soils. There were no consistent differences in N throughfall and total N deposition between oak and spruce stands of similar age although there was an indication of higher levels of N deposition in the 33-year-old spruce stand than in an oak stand of similar age. Also, no distinct species difference in canopy N uptake was observed between oak and spruce stands of similar age at Vestskoven, Denmark (Table 4.3). When the two species were compared along a stand height gradient, there was also no general tree species difference in N deposition for the same stand height at Vestskoven (Table 4.3). This observation contradicts earlier studies where significant higher roughness lengths have been observed for coniferous trees compared to deciduous trees (Erisman 1992; Draaijers 1993; Bleeker & Draaijers 2002), leading to higher N throughfall deposition. However, in a review Parker (1990) found that numerous studies showed higher annual throughfall deposition under hardwoods than under adjacent coniferous stands (Cronan & Reiners 1983; Henderson et al. 1977). The absence of a tree species related difference in N deposition between oak and spruce in our study may partly be attributed to the short part of the rotation period in which we could compare species (0-35 years). Differences in N deposition to the oak and spruce stands at Vestskoven may appear when the stands grow older.

Over approximately 35 years following afforestation, oak stands generally leached more nitrate than Norway spruce stands of similar age (Figure 4.5b). These results are in contrast to findings in European studies of paired mature stands of

coniferous and deciduous species at the same sites, showing higher seepage water nitrate concentrations and higher leaching of nitrate below coniferous tree species than below deciduous tree species (De Schrijver et al. 2000; Gundersen et al. 2005; de Vries & Jansen 1994; Rothe et al. 2002). The higher N leaching losses under conifers were attributed to higher N deposition to coniferous than to deciduous tree species, since conifers are evergreen and have larger foliar surfaces over the year, which receive more deposition. With the exception of the 33-year old Norway spruce stand there was no general difference in N deposition levels between even-aged oak and spruce stands at Vestskoven in Denmark that explains the difference in nitrate leaching between the two tree species. With similar input of N, Kristensen et al. (2004) did find higher N leaching in deciduous stands than coniferous stands. However, in their case the difference was mainly attributed to a general soil type difference between stands of the two forest types as represented in the European intensive monitoring (level II). This was not the case in our chronosequences, where possible causes could be differences in uptake or internal cycling of N between oak and spruce.

The difference in nitrate leaching between oak and spruce reflects the difference in growth rate between the two tree species. The growth of oak and hereby the N uptake starts out slowly while spruce has a high initial growth rate and high uptake of N. Hence, leaching fluxes were always low or negligible in young spruce stands receiving low N deposition (Figure 4.5b, Figure 4.7). At Vestskoven, the spruce stands stored more C and N in forest floors than the oak stands of similar age, although the input of C and N from litterfall was higher in the oak stands (Figure 4.4). This suggests a higher rate of decomposition of the more easily decomposable oak litter and hence a higher rate of N mineralization in the forest floor under oak. However, there is little conclusive evidence of such a species difference for north-western Europe. While oak litter has been reported to decompose faster than spruce litter (Dziadowiec 1987), Brüggemann et al. (2005) reported higher gross N mineralization rates in the forest floor under spruce than under oak in a species trial on sandy soil in Denmark. The difference in nitrate leaching between the two tree species might also partly be due to higher groundwater recharge under oak, because of lower interception loss than in the spruce stands (Chapter 3). The difference in annual water recharge between oak and spruce was most pronounced in the Dutch stands on sandy soil types, whereas tree species related differences in deep seepage was small in the less permeable clay-rich soils at Vestskoven in Denmark (Rosenqvist et al. 2006b). Similar water yield for oak and spruce has previously been reported in areas with low precipitation and less permeable soils (Augusto et al. 2002).

## 5. CONCLUSION

Forests on former arable land vary substantially in their ability to retain N in the ecosystem. No consistent pattern was apparent in the three countries. Differences in N retention and nitrate leaching depend on a combination of local and/or regional factors such as soil N status, soil type, N deposition, forest age, tree species and hydrology. However, an effect of the former land-use is evident and

the N dynamics in the newly planted forests on former arable land differ from those in old forests. The afforested sites are not (yet) in steady state and changes in the processes are ongoing. The interpretation of results from the studied chronosequences is therefore complicated by the transient nature of the factors that regulate leaching losses of N from the system. In the course of time, the environment in the new forests will evolve towards the environment in old forests. However, the present data do not allow determination of the length of such a transition period.

Compared to nutrient-poor sandy soils, nutrient-rich clayey soils appear more vulnerable to disturbance of the N cycle and to increased N deposition, possibly leading to N saturation and enhanced nitrate leaching. Furthermore, tree species seem to influence nitrate leaching and N retention. Our results suggest that the effect of different tree species (Norway spruce vs. oak) on N leaching after afforestation must be regarded in a time perspective. In the short-term (approximately 0-35 years) ecosystem N retention is generally less efficient in oak than in spruce, leading to higher N leaching losses from the oak stands. Over larger time scales (forest harvest rotation) the higher N deposition to conifers may result in higher leaching from coniferous than from deciduous stands, as often reported in literature.

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