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ORIGINAL PAPER

Impact of long-term nitrogen addition on carbon stocks in trees and soils in northern Europe

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Abstract The aim of this study was to quantify the effects of fertiliser N on C stocks in trees (stems, stumps, branches, needles, and coarse roots) and soils (organic layer +0–10 cm mineral soil) by analysing data from 15 long-term (14–30 years) experiments in *Picea abies* and *Pinus sylvestris* stands in Sweden and Finland. Low application rates (30–50 kg N ha^{-1} year^{-1}) were always more efficient per unit of N than high application rates (50–200 kg N ha^{-1} year^{-1}). Addition of a cumulative amount of N of 600–1800 kg N ha^{-1} resulted in a mean increase in tree and soil C stock of 25 and 11 kg (C sequestered) kg^{-1} (N added) (''N-use efficiency''), respectively. The corresponding estimates for NPK addition were 38 and 11 kg (C) kg^{-1} (N). N-use efficiency for C sequestration in trees strongly depended on soil N status and increased from close to zero at C/N 25 in the humus layer up to 40 kg (C) kg^{-1} (N) at C/N 35 and decreased again to about 20 kg (C) kg^{-1} (N) at C/N 50 when N only was added. In contrast, addition of NPK resulted in high (40–50 kg (C) kg^{-1} (N)) N-use efficiency also at N-rich (C/N 25) sites. The great difference in N-use efficiency between addition of NPK and N at N-rich sites reflects a limitation of P and K for tree growth at these sites. N-use efficiency for soil organic carbon (SOC) sequestration was, on average, 3–4 times lower than for tree C sequestration. However, SOC sequestration was about twice as high at *P. abies* as at *P. sylvestris* sites and averaged 13 and 7 kg (C) kg^{-1} (N), respectively. The strong relation between N-use efficiency and humus C/N ratio was used to evaluate the impact of N deposition on C sequestration. The data imply that the 10 kg N ha^{-1} year^{-1} higher deposition in southern Sweden than in northern Sweden for a whole century should have resulted in 2.0 ± 1.0 (95% confidence interval) kg m^{-2} more tree C and 1.3 ± 0.5 kg m^{-2} more SOC at *P. abies* sites in the south than in the north for a 100-year period. These estimates are consistent with differences between south and north in tree C and SOC found by other studies, and 70–80% of the difference in SOC can be explained by different N deposition.

Keywords N-use efficiency · C sequestration · C/N ratio · C stock in trees and soil

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Introduction

Nitrogen addition to forest ecosystems will affect a number of plant and soil processes (Aber et al. 1989, 1998). Nitrogen has a potential of increasing carbon sequestration, because net primary production (NPP) in most temperate and boreal forests are chronically restricted by a lack of nitrogen (Tamm 1991; Vitousek and Howarth 1991). However, long-term nitrogen addition might also affect foliar N, N mineralisation, nitrification, N leaching, N_2O efflux and methane consumption (Aber et al. 1998). The response of needle, branch and stem growth varies depending on fertilisation procedure and site productivity. In N-limited stands, a single application of 150 kg N ha^{-1} will generally increase tree growth for 7–10 years, resulting in an additional stem growth of about 15 m^3 ha^{-1} (Pettersson 1994; Nohrstedt 2001; Pettersson and Högbom 2004). The growth response after N fertilisation is clearly lower in stands growing on soils with high nutrient status than on soils with low nutrient status (Ingerslev et al. 2001). Consequently, the effect on carbon sequestration in trees depends on both the dose of N addition and the site fertility.

N fertilisation may affect the pool of soil organic carbon (SOC) through increased litterfall because of an increase in leaf area and higher production of other tissues. The role of fine roots is more complicated, because it is unclear whether increased N availability might influence fine-root growth. Forest floor mass and root biomass has been observed to decrease with increased N status (Gundersen et al. 1998b), but there are also indications of increases in standing root biomass and root-litter production after N fertilisation (Majdi and Andersson 2005). According to a literature review by Nadelhoffer (2000), it is likely that N deposition will decrease fine-root biomass but stimulate fine-root turnover and production.

N fertilisation and N deposition may also affect SOC storage by enhanced or reduced heterotrophic respiration as a response to litter C/N-ratio. High levels of external N generally stimulate the decay of plant tissues containing low concentrations of lignin and other recalcitrant compounds, while it may reduce mass loss rate for high-lignin materials (Fog 1988; Berg and Matzner 1997; Neff et al. 2002; Knorr et al. 2005). Conifer litters are rich in lignin, and in a north/south gradient study in Europe, a positive relationship between C/N ratio and CO_2 evolution rate was observed for litter and humus layers (Persson et al. 2000).

Soil incubation experiments in the laboratory have shown lower C mineralisation rates in mor humus from N-fertilised field plots than in mor humus from unfertilised plots (Persson et al. 2000). Modelling of bomb ^{14}C data from a field experiment further supports the results indicating that repeated addition of 30 kg N ha^{-1} $year^{-1}$ for 100 years may result in a doubling (1.3 kg C m^{-2}) of the amount of C stored in the mor layer (Franklin et al. 2003). About 60% of this increase was estimated to be a result of decreased decomposition rate and the rest a result of increased litter production. N addition can increase the fraction of stable humus in soils as found by ^{13}C tracer experiments leading to more SOC in the upper mineral soil (Hagedorn et al. 2003; Jandl et al. 2007). Thus, the studies mentioned above suggest that N addition will lead to a decrease in C mineralisation rate and an accumulation of SOC in the organic layer and mineral topsoil.

In this analysis, we examine quantitative effects of added N on C pools in trees and soils by analysing data from the literature and new investigations of 15 long-term experiments in Sweden and Finland. We hypothesise that both plant C and SOC pools will increase with increased amounts of added nitrogen, and that the increase is (i) dependent on ambient N status (C/N ratio) and long-term N deposition at the site, (ii) larger in *P. abies* (higher production) than in *P. sylvestris* stands, (iii) larger in young (higher production) than in old stands, but also that (iv) the increase is further enhanced by addition of other nutrients (P and K) or water, and that (v) the increase in soil C is a constant fraction of the increase in plant C.

Materials and methods

Site descriptions

Results on changes in tree biomass and soil C as a response to repeated N fertilisation were found in the literature and in partly unpublished materials for 15 experimental field sites in Sweden and Finland (Table 1, Fig. 1). The sites are located from northern Finland (latitude 67°42′ N, longitude 26°13′ E) to southernmost Sweden (latitude 56°26′ N, longitude 14°35′ E) and with a range of mean annual air

Table 1 Site and stand characteristics arranged from north to south.

Site (code)	Lat. N	Long. E	Altitude (m a.s.l.)	MAP (mm)	MAT (°C)	Wet dep. of N (kg N ha^{-1} $year^{-1}$)	Dominant tree	Tree age (years) at exp. start	pH (H_2O) in FH layer in control plots	Parent material
Sodankylä (197)	67°42′	26°13′	240	465	−1.3	1.3	PA	61	4.00	Glacial till
Kemijärvi (194)	66°51′	27°08′	280	518	−1.1	2.4	PA	31	4.00	Glacial till
Själlarimsheden (728)	66°32′	20°10′	275	530	−0.5	3.3	PS	82	4.10	Glacial till
Häggsjöliden (728a)	65°46′	19°30′	365	520	−0.5	3.3	PS	90	4.20	Glacial till
Norråker (777)	64°27′	15°34′	280	704	1.5	2.5	PA	153	4.85	Glacial till
Norrliden (E55)	64°21′	19°46′	260	595	1.2	3.3	PS	20	4.00	Glacial till
Flakaliden (E77)	64°07′	19°27′	310–320	624	2.3	3.3	PA	24	4.40	Glacial till
Lövnäs (731)	61°21′	13°22′	450	620	0.5	4.5	PS	124	4.20	Glacial till
Bleckstugan (731a)	61°23′	13°38′	410	620	0.5	4.5	PS	96	4.20	Glacial till
Stråsan (E26a)	60°55′	16°01′	350	560	3.1	5.5	PA	10	4.12	Glacial till
Jädraås (E75)	60°49′	16°31′	185	600	3.8	5.5	PS	16	3.90	Sediment
Asa (E79)	57°08′	14°45′	225–250	700	5.5	8.5	PA	15	4.45	Glacial till
Åseda (E63)	57°06′	15°29′	225	600	6.4	8.5	PA	12	4.31	Glacial till
Skogaby (E78)	56°33′	13°13′	95–115	1145	7.5	14	PA	22	3.90	Glacial till
Farabol (131)	56°26′	14°35′	130	669	6.4	8.5	PA	55	4.20	Glacial till

MAP = mean annual precipitation, MAT = mean annual air temperature, PA = *Picea abies*, PS = *Pinus sylvestris*. Data from Ågren (pers. comm.), Andersson et al. (1998), Franklin et al. (2003), Gay et al. (1994), Hallbäcken and Popović (1985), IVL (2006), O. Langvall (pers. comm.), Lenoir et al. (2007), Popović and Andersson (1984), Persson (1980), Persson and Nilsson (2001), Persson et al. (2000), Slaney et al. (2007), SMHI (http://www.smhi.se/sgn0102/images/p34.gif), Strömgren and Linder (2002)

temperatures of 9 °C. Four of the sites belong to the temperate zone and 11 to the boreal zone. All sites were dominated by conifer trees, nine sites with Norway spruce, *Picea abies* (L.) Karst., and six sites with Scots pine, *Pinus sylvestris* L. with a great variation in stand age (12–153 years) at the start of the experiments. The field layer vegetation was sparse and dominated by forest mosses at the southern sites, whereas mosses, lichens and dwarf-shrubs (*Vaccinium myrtillus* L., *V. vitis-idaea* L. and *Calluna vulgaris* L.) were common elements at the northern sites.

With one exception (Jädraås), all soils in the study are developed in glacial till derived from granitic and gneissic bedrock. The texture is sandy loamy or sandy, and only minor fractions of clay have been found. The soils are podzolised (mostly Haplic Podsols) with an organic layer (LFH) overlying the E and B horizons (FAO 1990). The humus layers have a pH of 3.9–4.9 in the control plots indicating acidic conditions (Table 1).

The sites represent a very distinct gradient in mean wet N deposition (open field) from 1 kg N ha^{-1} $year^{-1}$ in N Finland (Sodankylä) to 14 kg N ha^{-1} $year^{-1}$ in SW Sweden (Skogaby) (Table 1).

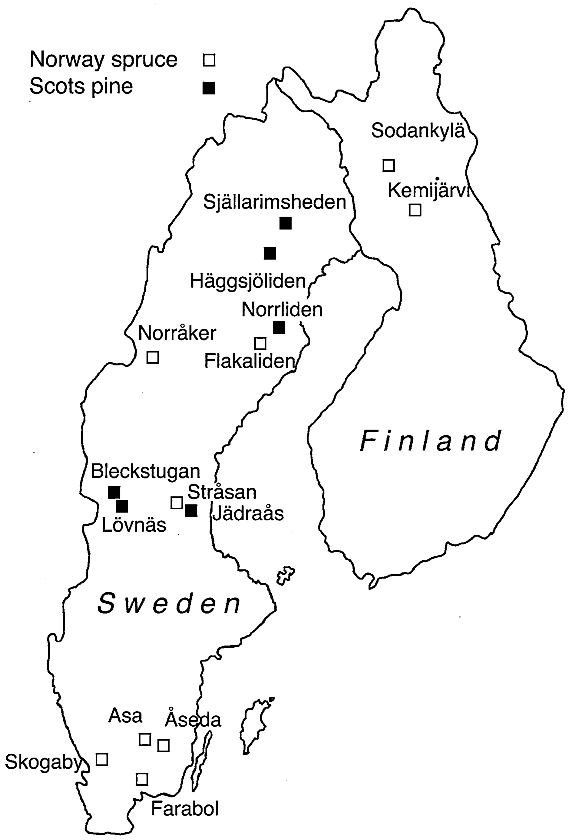


Fig. 1 Experimental sites for the 15 long-term fertilisation studies in Sweden and Finland compiled in the present study

N fertilisation

The fertilisation experiments included in this review comprise control and fertilised plots. The plots were at least 30 × 30 m, and to avoid influence from other treatments, tree growth and SOC pools were estimated for the central 20 × 20 m net plots. The fertilised ''gross'' plots received N fertilisers (ammonium nitrate, ammonium sulphate or urea) either alone or in combination with other nutrients (e.g., P, K, Mg, Ca and S) (Table 2). The number of applications before the latest tree or soil study (mostly one application per year) varied between sites, from 3 times (Farabol) to 30 times (Stråsan N1). The most complicated experiments (Stråsan and Norrliden) contained both N and NPK plots, and the treatments were given in different doses and in different N forms (Table 2). In addition, a certain treatment had often declining fertilisation rates over the treatment period. For example, N1 at Stråsan meant 60 kg N ha^{-1} year^{-1} for three years, 40 kg N for seven years and 30 kg N for the following years.

Some sites (Asa, Jädraås and Flakaliden) lacked N addition alone, and had only plots in which N was given in combination with P, K and other nutrients or N together with granulated wood ash. However, wood ash turned out to give negligible effects, implying that plots with N + wood-ash at Asa and Flakaliden could be considered as N plots. At some sites (Skogaby, Asa, Jädraås and Flakaliden), one of the treatments meant irrigation every second day (except for rainy days) with a liquid solution (IL) of N and other micro- and macronutrients during the growing season (June to mid-August). To evaluate the nutrient effect, plots with nutrient irrigation were compared with plots with just irrigation.

Most of the experiments were replicated, and only the experiments started in 1959–1965 lacked replicates. However, some of these experiments (Lövnäs/Bleckstugan, Häggsjöliden/Själlarimsheden and Sodankylä/Kemijärvi) were considered as statistical blocks within the same region. The control plot at Själlarimsheden was disturbed by a military camp (Popović and Andersson 1984) and, therefore, the Ca-amended plot was considered as control in relation to the fertilised plots.

Data analysis

Change in tree C pools

Data based on the experimental results differed in completeness between sites and time periods. At all sites, there were data on mean stem diameter at breast height and number of trees ha^{-1} on different plots and occasions (Popović and Andersson 1984; Gay et al. 1994; Andersson et al. 1998; Bergh et al. 1999; Persson and Nilsson 2001). When data on tree height was available, they were included in the calculations. This information was used to determine tree biomass above (stems, branches, needles and stumps) and below ground (coarse roots but not fine roots) according to the regression equations given by Marklund (1988) for *P. abies* and *P. sylvestris*.

The amount of C was estimated as 0.5 times the dry weight of biomass. The change in C pool was calculated for each plot, site and observation period. The effect of fertiliser N on C-pool changes was calculated as the difference between C pools in N-fertilised and control plots. In experiments that

Table 2 Experimental design and N fertilisation regimes

Site	N-fertilisation regimes	Period with N addition	No. of repl.	No. of N applic-ations	Mean N appl. dose (kg ha^{-1})	Cum. amount of N added (kg ha^{-1})	Tot. P (kg ha^{-1})	Tot. K (kg ha^{-1})	Tot. Mg (kg ha^{-1})	Tot. Ca (kg ha^{-1})	Tot. S (kg ha^{-1})
Sodankylä	N + NPK	1965–1991	1	6 times	125	752	80	180	–	?	?
Kemijärvi	N + NPK	1965–1990	1	6 times	125	752	80	180	–	?	?
Själlarimsheden	N + NPK	1959–1981	1	9 times	87	780	123	254	–	365	?
Häggsjöliden	N + NPK	1959–1981	1	9 times	87	780	123	254	–	365	?
Norråker	U + UPK	1963–1986	2	9 times	107	960	174	416	115	192	276
Norrliden	N1 + N1PK	1971–cont.	3	Annual	33	1170	280	540	–	660	370
Norrliden	N2 + N2PK	1971–cont.	3	Annual	67	2340	280	540	–	660	370
Norrliden	N3 + N3PK	1971–1989	3	Annual	109	2070	280	540	–	660	370
Norrliden	U1 + U1PK	1971–1989	3	Annual	36	690	280	540	–	660	370
Norrliden	U2 + U2PK	1971–1989	3	Annual	73	1380	280	540	–	660	370
Norrliden	U3 + U3PK	1971–1989	3	Annual	109	2070	280	540	–	660	370
Flakaliden	NA + NPK*	1987–2003	4	Annual	80	1200	212	555	119	107	119
Flakaliden	IL	1987–2003	4	Annual	80	1200	185	537	114	68	28
Lövnäs	N + NPK	1959–1981	1	10 times	78	780	123	254	–	365	?
Bleckstugan	N + NPK	1959–1981	1	10 times	78	780	123	254	–	365	?
Stråsan	N1 + N1PK	1967–cont.	2	Annual	34	1330	150–300	560	150	?	?
Stråsan	N2 + N2PK	1967–1990	2	Annual	73	1760	150–300	560	150	?	?
Stråsan	N3 + N3PK	1967–1992	2	Annual	108	2820	150–300	560	150	?	?
Jädraås	NPK	1974–1990	4	Annual	59	1000	180	360	–	?	?
Jädraås	IL	1974–1990	4	Annual	102	1740	226	1131	150	122	157
Asa	NA + NPK*	1988–2001	4	Annual	64	900	259	435	173	67	142
Asa	IL	1988–2001	4	Annual	64	900	261	522	149	15	39
Åseda	N + NPK	1974–1989	4	Annual	44	700	240	465	–	624	318
Skogaby	NS	1988–2001	4	Annual	100	1400	–	–	–	–	1596
Skogaby	IL	1988–2001	4	Annual	64	890	136	412	50	85	26
Farabol	U	1976–1985	3	3 times	200	600	–	–	–	–	–

N = ammonium nitrate, U = urea, NS = ammonium sulphate, NA = ammonium nitrate + wood ash, NPK = ammonium nitrate + PK salts including variable amounts of Ca and S, NPK* = NPK + other micro- (e.g. Fe, Cu, Zn, Mn, Mo, Na, B) and macronutrients (e.g. Mg, Ca and S) in solid form, IL = liquid fertilisation (irrigation with micro- and macronutrients), N1, N2 and N3 and U1, U2 and U3 indicate multiple doses of ammonium nitrate and urea, respectively. Total amounts of P, K, Mg, Ca and S in NPK/NPK* are also given. Data sources given in Table 1

included thinning, the differences were calculated separately for the periods before, between and after thinnings. The amount of C removed by thinning was added to the estimate of C content in the remaining stand to determine total sequestration of C. To estimate the effect of added N or NPK, the difference in C sequestration between fertilised and unfertilised (control) plots within block were expressed per unit of N added. In experiments with liquid fertilisation (IL), the fertiliser affect (N plus other nutrients added) was considered as the difference in C sequestration between IL plots and irrigated (I) plots.

Change in SOC pools

SOC pools were determined by soil sampling. Hallbäcken and Popović (1985) and Andersson et al. (1998) took 16–40 soil samples per plot with 56–72 mm diam. soil augers from the organic layer and the 0–5, 5–10 and 10–20 cm mineral soil layers.

In other studies, the topsoil layers were sampled by fewer (4–6) but wider samplers, 400-cm^2 frames for the organic layer and 100-cm^2 frames for the 0–5 and 5–10 cm mineral soil (Andersson et al. 1998; Persson and Nilsson 2001; Andersson et al. unpublished; Persson et al. unpublished). The samples were pooled for each plot and soil layer, and the pooled samples were sieved through 5-mm mesh (organic layer) or 2-mm mesh (mineral soil) to obtain the amount of fine soil. The sieved soil was dried at 105 °C for 24 h to obtain the dry matter for the appropriate surface area. C concentration was determined in a Leco WR-12 (Hallbäcken and Popović 1985) or Carlo-Erba NA 1500 analysers, and total C pool m^{-2} was obtained as the product of C concentration and dry matter m^{-2}. Because all soils were acidic (Table 2), carbonate C was negligible, and total soil C was considered as SOC.

The till soils (Table 1) had a variable degree of stoniness, and stoniness caused a huge variation in fine soil between plots within the same site. Stoniness was often more pronounced at greater soil depths. For the present comparison between fertilised and control plots, we decided to compare SOC pools in the combined organic and 0–10 cm mineral soil layers in order to reduce variability. We did not want to make a comparison between just organic layers, because different people define the borderline between organic and mineral soil differently despite identical sampling protocols.

In contrast to tree biomass, SOC pools were not estimated at the start of the experiment at many sites. Therefore, the effect of N fertilisation on SOC pool accumulation could only be evaluated as the difference between N-fertilised and non-fertilised plots for these sites.

Statistical treatment

Most of the experiments included in this review were replicated block experiments (Table 1). For each block, the difference between fertilised and control plots as regards tree C and SOC was estimated to assess the fertiliser effect, and mean and standard error of the differences were calculated for each experiment. N-use efficiency (kg C sequestered kg^{-1} N added) was calculated by dividing these estimates by the cumulative amount of N added at the appropriate time. The N-use efficiencies (mean per site) in relation to soil C/N ratios were analysed by means of regression analysis.

Results

Change in tree C pools

Tree C pools increased with time at all sites. In the control plots, mean C sequestration varied between 0.8 (Sodankylä/Kemijärvi) and 4.8 (Asa) Mg C ha^{-1} year^{-1} (Table 3), indicating low C sequestration rates in the north and high C sequestration rates in the south. The mean C sequestration at fertilised plots in excess of control plots varied between −0.1 (N treatment at Skogaby) and 4.0 (IL treatment at Flakaliden) Mg C ha^{-1} year^{-1} (Table 3). These estimates were dependent on tree species, stand age and site fertility and will be discussed in more detail for individual sites below.

Response of tree biomass C to N fertilisers

Norrliden and Stråsan

The patterns of C-pool growth in control plots (0) and after addition of ammonium nitrate (N), potassium and phosphorus (PK) and N in combination with PK (NPK) are shown in Fig. 2 for the two sites Norrliden (*P. sylvestris*) and Stråsan (*P. abies*). At both sites, annual addition of N and NPK resulted in a higher increase in tree C pool than in the control and PK treatments. The increase was more evident at the *P. abies* site Stråsan than at the *P. sylvestris* site Norrliden. Addition of PK alone had no effect on tree growth at Norrliden, whereas it had a tendency to increase tree growth in relation to control plots at Stråsan, although not significantly ($P > 0.05$).

At Norrliden, N and NPK addition significantly ($P < 0.05$) increased the sequestration of tree C in relation to unfertilised plots with the exception of N3PK ($P > 0.05$), which had great variation between blocks (Figs. 2 and 3). The cumulative increase in tree C pool as a function of cumulative N addition was higher for the low application rate (N1/U1) than for the intermediate (N2/U2) and high (N3/U3) application rates (Fig. 3). For example, N-use efficiency (kg (C sequestered) kg^{-1} (N added) was estimated to be about 35, 17 and 9 kg (C) kg^{-1} (N)

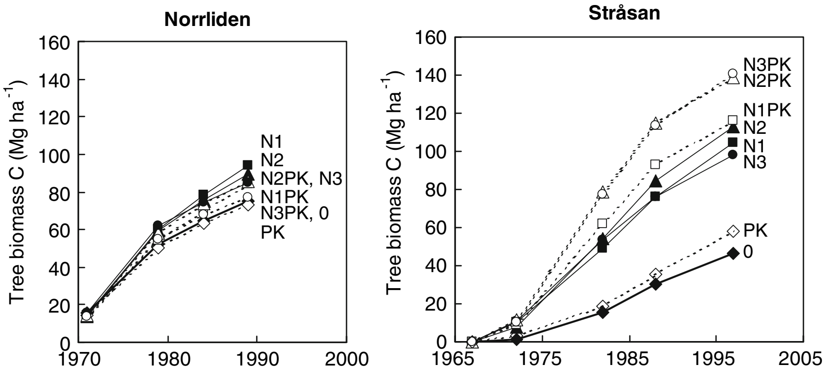
Table 3 Mean (±1 SE) N-use efficiency (kg (C sequestered) kg^{-1} (N added)) for tree biomass (including coarse roots and wood removed in thinning) and soil (LFH + 0–10 cm layers) at the sites given in Tables 1 and 2

Site	Tree species	Mean C/N in FH layer in control plots/ N plots	Time (years) from first N add. to tree/soil study	N source	Cum. amount of N (kg ha^{-1}) added before tree/soil study	Mean C seq. (± SE) in trees in control plots (Mg ha^{-1} $year^{-1}$)	Mean C seq. (± SE) in trees in excess of control plots (Mg ha^{-1} $year^{-1}$)	Mean C seq. (± SE) in soil in excess of control plots (Mg ha^{-1} $year^{-1}$)	N-use efficiency in trees (kg (C) kg^{-1} (N)	N-use efficiency in soil (kg (C) kg^{-1} (N)	Ratio between N-use eff. in trees and soil
Sodankylä + Kemijärvi	PA	45/n.e.	29	N	752	0.78 ± 0.14	0.47 ± 0.03	n.e.	18 ± 1	n.e.	n.e.
				NPK	752		1.02 ± 0.12	n.e.	39 ± 5	n.e.	n.e.
Själlarimsheden + Häggsjöliden	PS	44/38	20/23	N	780	0.95 ± 0.34	1.29 ± 0.65	0.16 ± 0.11	33 ± 17	4.6 ± 3.2	7.2
		44/36		NPK	780		1.12 ± 0.31	0.32 ± 0.12	29 ± 8	9.5 ± 3.6	3.0
Norråker	PA	34/n.e.	28	U	960	0.93	1.19	0.48	35	14	2.5
				NPK	960		0.85	0.41	25	12	2.1
Norrliden	PS	37/n.e.	18/21	N1	690/750	2.44 ± 0.19	1.33 ± 0.27	0.26[a] ± 0.03	35 ± 7	7.3[1] ± 0.7	4.8
				N1PK	690/750		0.98 ± 0.38	0.43[a] ± 0.06	25 ± 10	12[1] ± 1.8	2.1
				U1	690/750		1.21 ± 0.36	n.e.	32 ± 9	n.e.	n.e.
				U1PK	690/750		1.39 ± 0.24	n.e.	36 ± 6	n.e.	n.e.
Flakaliden	PA	37/n.e.	17	NA	1200	1.56 ± 0.09	2.76 ± 0.22	n.e.	39 ± 5	n.e.	n.e.
		37/24	17/14	NPK*	1200/1050		3.75 ± 0.28	0.66 ± 0.14	53 ± 6	8.7 ± 1.9	6.1
		37/n.e.	17	IL	1200		4.02 ± 0.19	n.e.	56 ± 4	n.e.	n.e.
Lövnäs + Bleckstugan	PS	51/43	20/23	N	780	1.00 ± 0.12	0.85 ± 0.01	0.23 ± 0.04	21 ± 0.3	6.8 ± 1.3	3.1
		51/43		NPK	780		1.17 ± 0.23	0.20 ± 0.19	29 ± 6	5.8 ± 5.7	5.0
Stråsan	PA	35/24	30/27	N1	1090/1760	1.54 ± 0.13	1.94 ± 0.14	1.21 ± 0.46	53 ± 4	19 ± 4.6	2.9
		35/27		N1PK	1090/1760		1.92 ± 0.31	0.80 ± 0.39	53 ± 9	12 ± 0.3	4.3
Jädraås	PS	36/33	25/30	NPK	1000/1000	1.80 ± 0.12	1.42 ± 0.24	0.10 ± 0.08	35 ± 6	3.1 ± 2.6	11.4
		36/29		IL	1740/1740		1.36 ± 0.09	0.28 ± 0.09	19 ± 1	5.9 ± 1.6	3.3
Asa	PA	23/n.e.	14	NA	900	4.76	0.15	n.e.	2	n.e.	n.e.
				NPK*	900		2.75	n.e.	43	n.e.	n.e.
				IL	900		3.91	n.e.	61	n.e.	n.e.
Åseda	PA	29/25	20	N	700	2.30	0.82	0.51 ± 0.13	23	15 ± 3.8	1.6
		29/27		NPK	700		1.68	0.69 ± 0.13	48	20 ± 3.7	2.4
Skogaby	PA	28/22	14/17	N	1400/1400	3.95 ± 0.31	−0.08 ± 0.29	0.70 ± 0.23	−0.8 ± 3	8.5 ± 2.7	−0.1
		28/26		IL	890/890		2.11 ± 0.28	0.72 ± 0.33	33 ± 4	14 ± 6.3	2.4
Farabol	PA	29/28	15	U	600	3.88	0.61	0.42 ± 0.01	15	10 ± 0.2	1.4

N-use efficiency was calculated for the cumulative N amount added before the measurement. PA = *Picea abies*, PS = *Pinus sylvestris*, N = ammonium nitrate, U = urea, NS = ammonium sulphate, NA = ammonium nitrate + wood ash, NPK = ammonium nitrate + P and K, NPK* = NPK + other micro- and macronutrients, IL = liquid fertilisation (irrigation with micro- and macronutrients). In the dose experiments Norrliden and Stråsan, only the lowest doses (N1 or U1) were considered (see text). Some nearby sites lacking replicates were considered as blocks ($n = 2$) (sites 197 and 194, 728, 728a, 731 and 731a). n.e. = not estimated

[a] Estimated as a mean of both N and U

Fig. 2 Development in tree biomass C (above and below ground) in control plots (0) and after annual additions of ammonium nitrate (N), potassium and phosphorus (PK) or NPK at different doses (Table 2) in the *Pinus sylvestris* and *Picea abies* stands at Norrliden and Stråsan, respectively



for N1, N2 and N3, respectively, 18 years after the start of the experiment, when total amounts of 690, 1380 and 2070 kg N ha^{-1} had been added (Fig. 3). Therefore, values of N-use efficiency at Norrliden (Table 3) were based on the estimates at low application rates (N1, N1PK, U1 and U1PK).

At Stråsan, the intermediate dose (N2) resulted in slightly higher tree biomass C than the low (N1) and high (N3) dose when given alone (Fig. 2). When N was given in combination with PK, N2 and N3 resulted in higher C sequestration than N1 after the same time (Figs. 2 and 4). Similar to Norrliden, N-use efficiency was higher for the low application rate (N1) than for the intermediate (N2) and high (N3) application rates and higher for N1PK than for N2PK and N3PK (Fig. 4). N-use efficiencies estimated after a cumulative dose of 1090 kg N ha^{-1} applied during 30 years are given in Table 3.

Asa and Flakaliden

The *P. abies* sites Asa and Flakaliden, situated in southern and northern Sweden, respectively, had similar experimental treatments comprising (i) annual additions of a solid fertiliser containing NPK and other macro- and micronutrients (NPK*), (ii) liquid fertilisation (IL, irrigation with the same mix of

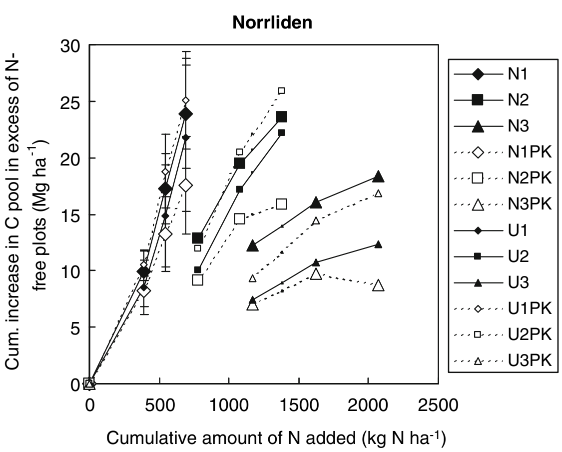


Fig. 3 Mean cumulative increase in tree biomass C in excess of control plots in the Scots pine stand at Norrliden as a function of the cumulative amount of N added. Mean application rate of N given as ammonium nitrate (N) or urea (U) was 33 for N1/U1 (diamonds), 67 for N2/U2 (quadrats) and 109 kg ha^{-1} year^{-1} for N3/U3 (triangles). For visibility, SE ($n = 2$) is only given for N1/U1. SEs are similar for the other treatments

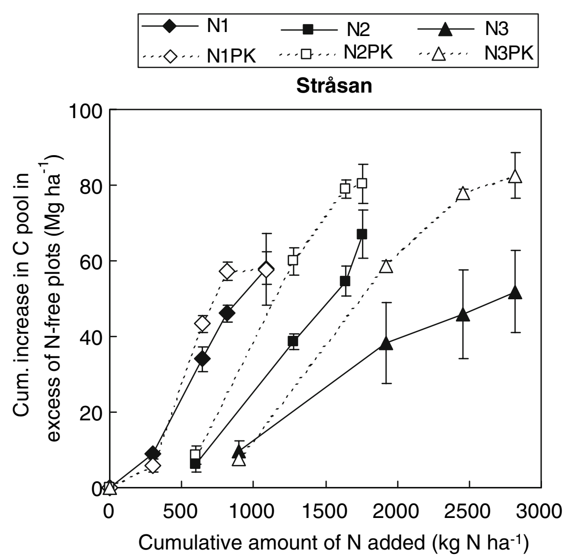


Fig. 4 Cumulative increase (mean ± SE, $n = 2$) in tree biomass C in excess of control plots (for NPK in excess of PK plots) in the Norway spruce stand at Stråsan as a function of the cumulative amount of N added. Mean application rate of N was 34, 73 and 108 kg ha^{-1} year^{-1} for N1, N2 and N3, respectively. Experimental period 30 years (see Fig. 2)

nutrients as in the solid fertiliser) and (iii) addition of wood ash (single dose at the start of the experiment) in combination with ammonium nitrate (NA) that was added annually at the same doses as in the other treatments. With the exception of N, the wood ash contained most of the nutrients found in the other treatments. However, the wood ash was granulated and did not dissolve as expected. Other experiments showed that the granulated wood ash was almost inert (B. Olsson, pers. comm.) and, therefore, the NA treatment was considered as a pure N treatment.

At Asa, the IL treatment resulted in a higher increase in tree biomass C (in relation to irrigated plots) than the NPK* treatment (in relation to control plots), whereas the NA treatment did not increase biomass C in relation to the control plots (Fig. 5). At Flakaliden, all treatments resulted in increased tree biomass C in relation to irrigated/control plots, and the increase after solid and liquid fertilisation did not differ (Fig. 5). The NA treatment resulted in a smaller increase in biomass C than the IL and NPK* treatments.

The initially low N-use efficiency in the NPK* and IL treated plots at both Asa and Flakaliden (Fig. 5) reflects a delay in biomass growth to N addition during the first 4–5 years. The up-bending curves for Flakaliden at high cumulative amounts of N added can be explained by the fact that there was a 2-year stop in fertiliser application, and during this stop the trees at the fertilised plots continued to grow better than at the control plots. A major difference between Asa and Flakaliden was the marked response to pure N fertilisation (including ash) at Flakaliden but not at Asa (39 and 2 kg C sequestered per kg N added, respectively).

Skogaby

The response of tree growth to N (given as ammonium sulphate, NS) and IL addition at the *P. abies* site Skogaby was similar to that at Asa in the same region. Addition of high doses (100 kg N ha^{-1} year^{-1}) of NS resulted in a small increase in tree biomass C in relation to control plots during the first six years in the *P. abies* experiment at Skogaby (Fig. 6). After 14 years of application with the same N-application rate, the increase had turned into an insignificant decrease in relation to the control plots. The N-use efficiency for the whole 14-year period, thus, became negative (Table 3). During the period between 6 and 14 years, the tree needles got K/N, P/N and Mg/N ratios indicating sub-optimal tree growth (Linder 1995; Persson and Nilsson 2001). This was probably caused by the acidifying effect of NH_4^+ addition that, in turn, resulted in increasing soil Al^{3+} concentrations that blocked the root uptake of K^+ and Mg^{2+} and decreased the solubility of P (Persson and Nilsson 2001). Consequently, the NS treatment strengthened the nutrient limitation in relation to the control treatment.

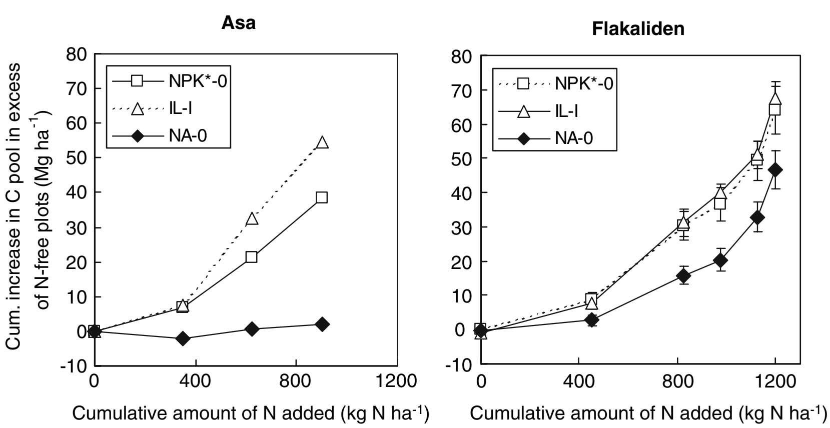


Fig. 5 Cumulative increase (n = 4) in tree biomass C after annual additions of a solid fertiliser containing NPK and other macro-and micronutrients (NPK*), liquid fertilisation (IL) and annual additions of ammonium nitrate in combination with a single addition of wood ash (NA) in the spruce stands at Asa and Flakaliden in excess of control plots (0) and irrigation plots (I) as a function of the cumulative amount of N added. N-application period 14 years at Asa and 17 years at Flakaliden (see Table 2)

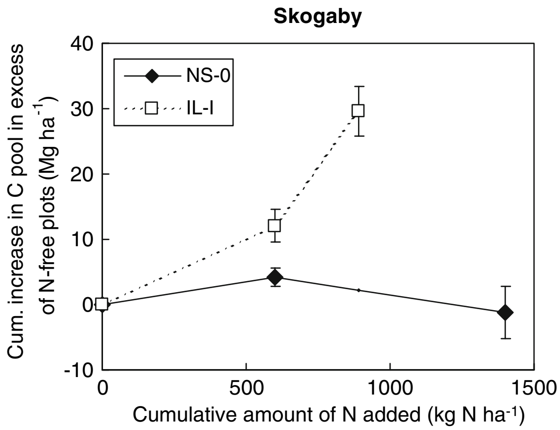


Fig. 6 Cumulative increase (mean ± SE, $n = 4$) in tree biomass C after ammonium sulphate (NS) and IL treatments in the *P. abies* stand at Skogaby in excess of control plots (0) and irrigation plots (I) as a function of the cumulative amount of N added. N-application period 14 years (see Table 2)

Liquid fertilisation with ammonium nitrate and other macro- and micronutrients resulted in a much higher increase in tree biomass C than the NS treatment. Irrigation (I) also increased biomass C, and the difference between IL and I was considered as the nutrient effect (Fig. 6).

The N application rates in IL were 100 kg N ha^{-1} year^{-1} during the first 6 years, but because of heavy nitrate leaching, the application rates were reduced to, on average, 36 kg N ha^{-1} year^{-1} during the following 8 years. The N-use efficiency for the whole period (14 years) was estimated at 33 kg (C sequestered) kg^{-1} (N added) (Table 3), but was 60 kg (C) kg^{-1} (N) during the 8-year period with low application rate and low nitrate leaching, i.e. similar to the estimate for the IL treatment at Asa (Table 3).

Other spruce stands

Andersson et al. (1998) made a review of long-term Nordic fertilisation experiments, and data from five of the *P. abies* sites in this study, Sodankylä, Kemijärvi, Norråker, Åseda and Farabol, have been used in the present compilation (Tables 2 and 3). NPK addition resulted in higher C sequestration than N addition at Sodankylä/Kemijärvi and Åseda, whereas the reverse situation was found for Norråker.

The young stand at Åseda had moderately high N-use efficiencies during the study period, 23 and 48 kg (C) kg^{-1} (N added) in the N- and NPK-treated plots, respectively, whereas the old stand at Farabol had lower (15) N-use efficiency (Table 3). The northern stands at Norråker (high age) and Kemijärvi/Sodankylä (medium age) had intermediate N-use efficiencies (Table 3).

Other pine stands

The mature *P. sylvestris* stands at Häggsjöliden och Själlarimsheden, situated in northern Sweden, and Lövnäs and Bleckstugan, situated in central Sweden had similar experimental treatments comprising ten N additions (78 kg N ha^{-1}) every second year (Popović and Andersson 1984). All stands responded with increased tree growth to addition of N and NPK during the experimental 20 years. The N-use efficiency for this period was estimated at 21–33 kg (C) kg^{-1} (N) in the N- and NPK-treated plots (Table 3). No extra effect of the PK addition could be detected.

Both NPK addition and IL resulted in increased C sequestration in the young *P. sylvestris* stand at Jädraås in relation to unfertilised and irrigated plots, respectively (Table 3). The mean N doses given were high, 76 and 146 kg N ha^{-1} year^{-1}, during the first 8 years. After this period, the doses were reduced, and after a cumulative addition of 1000 and 1740 kg N ha^{-1}, respectively, the application was stopped after 16 years. During the following nine years (1991–1999), tree growth continued to be higher in the NPK and IL plots than in the untreated plots. Thus, there was an after-effect of the fertilisation just as was observed for the trees at Flakaliden (see above). The N-use efficiency for the whole period (25 years) was higher for NPK than for IL (Table 3). These figures are most probably underestimates, especially for the IL treatment, of what could have been obtained with smaller N application rates during the initial fertilisation period.

N-use efficiency for tree growth

N-use efficiency in relation to fertiliser regime and C/N ratio

The estimates of N-use efficiency varied between −0.8 and 61 kg C sequestered per kg N added depending on geographical situation, soil status, tree species, stand age and fertiliser composition and dose

(Table 3). On average, N-use efficiency (mean ± SE) was 23 ± 6.5 (n = 8) and 30 ± 4.3 (n = 3) kg (C sequestered) kg^{-1} (N added) for *P. abies* and *P. sylvestris*, respectively, when N was added without any PK fertiliser. The corresponding estimates for solid NPK fertiliser (with or without other nutrients) were 44 ± 4.4 (n = 6) and 30 ± 2.1 (n = 4) kg (C sequestered) kg^{-1} (N added) for *P. abies* and *P. sylvestris*, respectively. Liquid fertilisation resulted in an N-use efficiency of 50 ± 8.5 (n = 3) kg (C sequestered) kg^{-1} (N added) for *P. abies*, whereas the single estimate for *P. sylvestris* (Jädraås) was very low (19 kg (C sequestered) kg^{-1} (N added)) indicating heavy leaching losses (see above).

All these estimates of N-use efficiency showed considerable variation, because other factors than fertiliser regime seemed to affect the result. One such factor was the C/N ratio of the humus layer in the control plots. In Swedish *P. abies* stands, which all were growing on soils with moderately low C/N ratios (25–37), there was a clear positive relation (R^2 = 0.76) between increasing C/N ratio and N-use efficiency after addition of N fertiliser (Fig. 7). In *P. abies* and *P. sylvestris* stands fertilised with NPK (with or without other nutrients), there was, on the other hand, a weak negative relation (R^2 = 0.16) between C/N ratio and N-use efficiency (Fig. 7). The northernmost *P. abies* stands in Finland and the *P. sylvestris* stands, all growing on soils with high C/N ratios (37–51), had similar estimates of N-use efficiency after N and NPK/NPK* fertilisation. The large difference in N-use efficiency between N and NPK fertilisers at low but not high C/N ratios indicates that N fertilisation alone has small effects on tree growth and C sequestration in N-rich soils, and that the lack of response depends on limitation of P, K and possibly other nutrients. At sites with high C/N ratios, P and K availability seems to be reasonably high also after long-term N fertilisation.

N-use efficiency in relation to tree species and stand age

The relations in Fig. 7 are confounded by the fact that *P. abies* dominated at low C/N sites, and no *P. sylvestris* sites were found in southern Sweden in this dataset. In central Sweden, Stråsan (*P. abies*) and Jädraås (*P. sylvestris*) had stands of similar age. N-use efficiency was 1.5 times higher in the *P. abies* than in the *P. sylvestris* stand (Table 3). In northern Sweden, both Flakaliden (*P. abies*) and Norrliden (*P. sylvestris*) had young stands at the start of the experiment. N-use efficiency was fairly similar after N treatment (Table 3), but higher in the *P. abies* than in the *P. sylvestris* stand (53 and 25 kg (C) kg^{-1} (N), respectively) after NPK/NPK* treatment. In the old stands of Norråker (*P. abies*) and Själlarimsheden/Häggsjöliden (*P. sylvestris*), there was practically no difference in N-use efficiency, neither after N addition nor after NPK addition (Table 3).

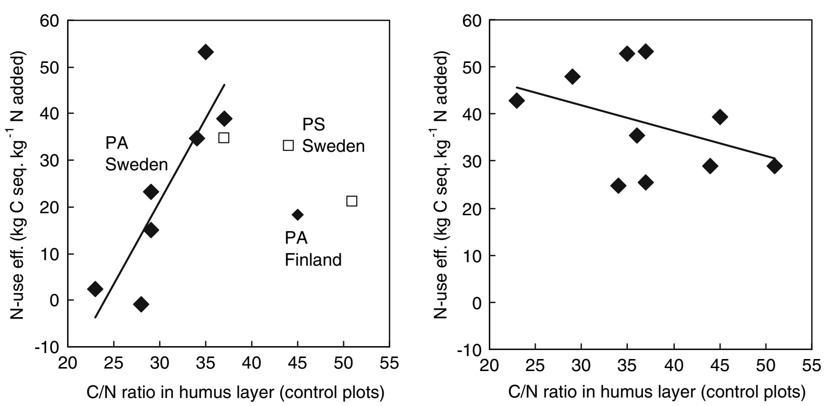


Fig. 7 N-use efficiency for C sequestration in tree biomass as a function of C/N ratios in humus layers of control plots after addition of N (left) or NPK (right) fertiliser as shown in Table 3. The regression line to the left is based on N fertilisation of *P. abies* (PA) sites in Sweden, C/N 25–37 (n = 7, R^2 = 0.76), and the regression line to the right is based on solid NPK/NPK* fertilisation of both *P. sylvestris* and *P. abies* stands (n = 10, R^2 = 0.16). Open quadrats denote *P. sylvestris* (PS) sites in Sweden (n = 3)

Also stand age seemed to have an impact on N-use efficiency. In southern Sweden, N-use efficiency at Åseda (young *P. abies*) and Farabol (old *P. abies*), both growing at sites with equal C/N ratio, was higher in the young than in the old stand (Table 3). In central Sweden, N-use efficiency was only slightly higher in young *P. sylvestris* at Jädraås than in old *P. sylvestris* at Lövnäs/Bleckstugan after NPK fertilisation. In northern Sweden, N-use efficiencies did not differ between the young *P. sylvestris* at Norrliden and the old *P. sylvestris* at Själlarimsheden/Häggsjöliden (Table 3). Similarly, N-use efficiency at Flakaliden (young *P. abies*) and Norråker (old *P. abies*) did not differ much between the young and the old stand after N fertilisation, but N-use efficiency was clearly higher in the young than in the old stand (53 and 25 kg (C) kg^{-1} (N), respectively) after NPK fertilisation.

In conclusion, young *P. abies* stands seem to have a greater capacity than old *P. abies* or young *P. sylvestris* stands to utilise fertiliser N for C sequestering, especially if P and K are not limiting growth. On the other hand, there was no indication of a difference between old *P. sylvestris* and old *P. abies* stands or between young and old *P. sylvestris* stands in N-use efficiency.

Change in soil C pools

Soil organic carbon pools often increased with time in both control and fertilised plots, but at many sites SOC was not estimated at the start of the experiment. Therefore, the effect of N fertilisation on SOC pool accumulation could only be evaluated as the difference between N-fertilised and non-fertilised plots for these sites.

SOC sequestration in fertilised plots was always higher than that in control plots. Mean SOC sequestration in excess of control plots varied between 0.1 (Jädraås, NPK treatment) and 1.2 (Stråsan, N2 treatment) Mg C ha^{-1} $year^{-1}$ (Table 3).

The estimates of N-use efficiency in SOC sequestration varied between 3 and 20 kg (C sequestered) kg^{-1} (N added) (Table 3). On average, N-use efficiency (mean ± SE) was 11 ± 1.7 (n = 8) and 11 ± 1.6 (n = 8) kg (C sequestered) kg^{-1} (N) when N and NPK/NPK* fertilisers were added, respectively. Thus, there was no significant difference in N-use efficiency for SOC sequestration between the N and NPK/NPK* fertiliser treatments (Fig. 8).

The results in Fig. 8 indicate that there is a possible relation between increasing C/N ratio in soil and reduced N-use efficiency. However, the sites with high C/N ratio and low N-use efficiency had all *P. sylvestris* stands, whereas the sites with low C/N ratio and high N-use efficiency had *P. abies* stands (Fig. 8). The *P. abies* and *P. sylvestris* sites had N-use efficiencies (mean ± SE) of 13 ± 1.7 (n = 5) and 6 ± 0.8 (n = 3) kg (C sequestered) kg^{-1} (N) added, respectively, for N-treated plots, and 13 ± 1.8 (n = 5) and 8 ± 2.0 (n = 4) kg (C sequestered) kg^{-1} (N) added for NPK-treated plots. There was a small overlap in C/N ratios between *P. abies* and *P. sylvestris* sites for NPK-treated plots indicating higher SOC sequestration at the former sites (Fig. 8, right part), but the data do not allow a firm judgement of whether differences in C/N ratio or differences in tree species are responsible for the differences in N-use efficiency.

Discussion

Imbalance in site properties

The main aim of this study was to quantify the effects of fertiliser N on C stocks in trees and soils by analysing data from 15 long-lasting experiments in Sweden and Finland, nine sites with *P. abies* and six sites with *P. sylvestris*. Some sites lacked replicates in their experimental design, and nearby experiments were, therefore, considered as statistical blocks within the same region. This meant that the dataset was reduced to eight *P. abies* sites and four *P. sylvestris* sites. Among these sites, no *P. sylvestris* site was found in southern Sweden, whereas *P. abies* was also present in northern Sweden and Finland. Young stands dominated in southern Sweden and old stands in northern Sweden and Finland. This made a proper evaluation of factors determining C sequestration more complicated.

N-use efficiency for C sequestration in trees

The experiments with different application rates of N at Norrliden and Stråsan showed that the increase in tree C pool as a function of cumulative N addition was higher

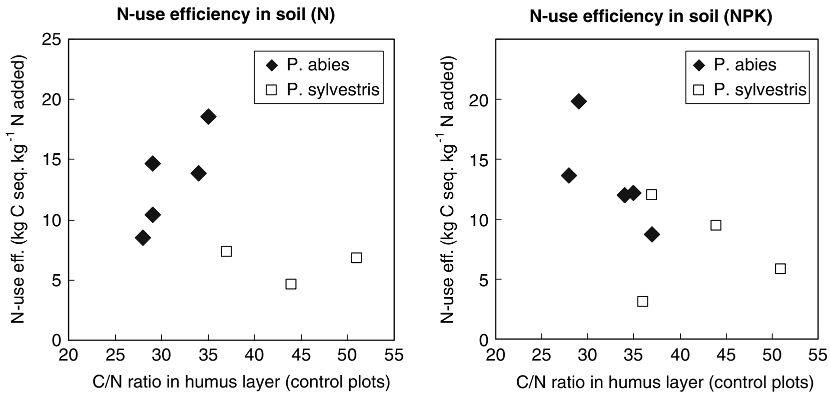


Fig. 8 N-use efficiency for C sequestration in soil (organic layer + 0–10 cm mineral soil) as a function of C/N ratios in humus layers of control plots after addition of N (left) or NPK (right) fertiliser to *P. abies* and *P. sylvestris* stands (see also Table 3)

for low application rates (30–35 kg N ha^{-1} year^{-1}) than for intermediate (60–70 kg N ha^{-1} year^{-1}) and high (90–100 kg N ha^{-1} year^{-1}) N-application rates as also demonstrated by Högberg et al. (2006). Lower N-use efficiencies for higher application rates can be explained by a combination of high N leaching (Johannisson et al. 1999) and storage of excess N in needles (Quist et al. 1999) and soil (Persson et al. 2000). Only the experiments at Norrliden, Stråsan and Åseda had mean N-application rates lower than 50 kg (N) ha^{-1} year^{-1}, and this indicates that the estimates for most other stands were probably underestimates of their potential N-use efficiency. This is further indicated by the fact that N-use efficiency increased as a response to lower N application rates during the course of some experiments, and the effect could be shown as up-bending curves in Figs. 5 and 6. During this period, both the amount of needles and the N concentration in needles increased. It is also possible that the N concentration in soil increased, but data are lacking for the initial period. The low N-use efficiency during the first years at Asa and Flakaliden (Fig. 5) cannot be explained by leaching losses of N, because N leaching was found to be very low at both sites (H. Grip, pers. comm.). Despite evidence of N-leaching associated with high N application rates, for example at Skogaby (Persson and Nilsson 2001), we had too few data to estimate the quantity of N leaching at most sites. Therefore, we could not correct the estimates of N-use efficiency for such losses, and the estimates given in Table 3 and Fig. 7 are, thus, uncorrected underestimates.

N fertilisation mostly seemed to increase C sequestration in trees. The only exception was N fertilisation of stands with N-rich soils (C/N ratios of 23–28 in the humus layer), where the effect was very small. In contrast to N fertilisation, addition of NPK (at some sites also including other nutrients) resulted in a high N-use efficiency also in N-rich soils. The great difference in N-use efficiency between addition of NPK and N at N-rich sites can be explained by limitation of P, K and possibly other nutrients for high tree growth.

This conclusion can only be drawn for *P. abies*, because *P. sylvestris* was not represented at the N-rich sites. At N-poor sites (C/N ratios 37–51), C sequestration in *P. abies* and *P. sylvestris* was almost equally stimulated by N and NPK fertilisation. However, young *P. abies* responded with higher C sequestration than young *P. sylvestris* and old *P. abies* to NPK fertilisation.

In our study, the threshold of positive tree responses to N fertilisation seemed to be at a C/N ratio of 25 in the humus layer. At this C/N ratio or lower, nitrate leaching has been found to increase in forests in Europe (Gundersen et al. 1998a; MacDonald et al. 2002) and North America (Magill et al. 2004; McNulty et al. 2005). Nitrate leaching indicates that plants and soil microorganisms cannot assimilate all inorganic N produced by N mineralisation and N deposition. Thus, it seems reasonable that application of N fertilisers to such ecosystems will not have any growth effect or will even reduce tree growth (McNulty et al. 2005). However, in our dataset addition of NPK fertilisers (also including other nutrients) meant a dramatic difference, and tree growth and C sequestration could be increased also at the C/N 23 site Asa without any nitrate losses (H. Grip, pers. comm.).

N-use efficiency for C sequestration in soils in relation to trees

We initially hypothesised that there would be a linear relation between C sequestration in trees and soil, because the amount of SOC would depend on long-term tree production and litter-fall. This hypothesis could not always be supported. As shown in Fig. 9, *P. abies* sites with low N-use efficiencies for tree growth had relatively high SOC sequestration. At one site (Skogaby), there was a marked increase in SOC accumulation despite a weak negative effect on tree growth after N addition. This indicates reduced decomposition rate after N addition. C mineralisation rate in the humus layer at N-fertilised plots at Skogaby, measured at constant temperature and moisture in the laboratory, was estimated to be 61% of that in control plots 9 years after the start of the treatment (Persson and Nilsson 2001). This is in agreement with results from Berg and Matzner (1997), Persson et al. (2000), Franklin et al. (2003) and Knorr et al. (2005).

N-use efficiency for SOC sequestration was twice as high at *P. abies* as at *P. sylvestris* sites also at sites with equal N-use efficiency for C sequestration in trees (Fig. 9). This might imply that *P. abies* sites have higher litter production and/or lower heterotrophic respiration than *P. sylvestris* sites at equal tree production. ^{13}C tracer experiments have shown that the net accumulation of tree-derived C can be greater in loamy soils with fairly low productivity than in fertile sandy soils with high productivity (Hagedorn et al. 2003; Jandl et al. 2007). Most *P. sylvestris* sites in our study had sandy soils, whereas the *P. abies* sites had soils with sandy loam. It is possible that lesser stabilisation of soil C in sandy than in loamy soils might have promoted higher heterotrophic respiration under *P. sylvestris*. In conclusion, it seems obvious that SOC sequestration in response to N fertilisation is dependent on more factors than just plant production.

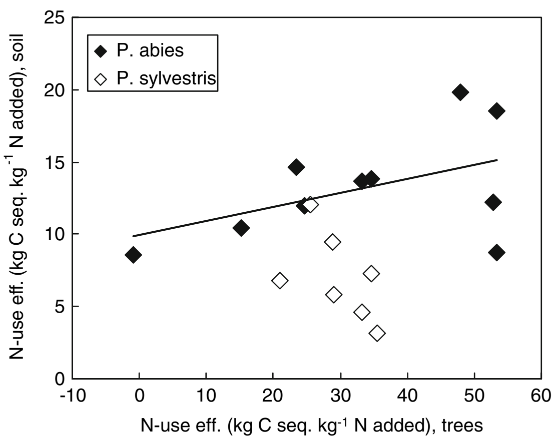


Fig. 9 Relationship between N-use efficiencies in trees and soils for *P. abies* and *P. sylvestris* sites with both tree and soil studies. The figures are based on data from both N and NPK fertilisation. The regression line is given for *P. abies* sites (y = 0.1x + 9.9, R^2 = 0.23)

There are several attempts in the literature to estimate the impact of N fertilisation and/or N deposition on C sequestration in trees and soils (e.g., Nadelhoffer et al. 1999; de Vries et al. 2006). Based on more than 100 intensively monitored forest plots and scaled up to a large part of Europe, de Vries et al. (2006) suggested that an additional N input would result in a net sequestration of 25 kg (C) kg^{-1} (N) in stem wood and 21 kg (C) kg^{-1} (N) in soil. These estimates correspond to about 50 (whole tree) + 21 (soil) kg (C) kg^{-1} (N) when using our expansion factors (1.8–2.2) for scaling up from stem to all tree fractions. Consequently, the estimates of de Vries et al. (2006), based on a combination of measurements and model calculations, were about twice as high as our mean estimates for Sweden and Finland but similar to our estimates of sites with high N-use efficiencies for trees and soils.

Impact of N deposition

N deposition (sum of ammonium and nitrate N) to *P. abies* forests was estimated at >20 kg ha^{-1} year^{-1} in southern Sweden and 3–6 kg N ha^{-1} year^{-1} in northern Sweden during early 1990s (Lövblad et al. 1995). The historical N deposition is difficult to estimate. However, Schöpp et al. (2003) estimated deposition trends of SO_2, NO_3^- and NH_3/NH_4^+ in sensitive freshwater regions in Europe during 1880–2030. By using the relative shape of the N deposition curve in Schöpp et al. (2003) for lake Gårdsjön in western Sweden and combining it with the figures for 1990 given by Lövblad et al. (1995) for southern and northern Sweden, we estimated the increase in N deposition from 6 to 20 kg ha^{-1} year^{-1} in southern Sweden and from 1 to 4 kg ha^{-1} year^{-1} in northern Sweden from 1900 to 1990, respectively, followed by a slight decline between 1990 to 2000. Based on these estimates, the cumulative N deposition during the 20th century would be 1250 and 250 kg N ha^{-1} in southern and northern Sweden, respectively. Thus, southern Sweden would have received 1000 kg (N) ha^{-1} more than northern Sweden during this 100-year period.

Data on C/N ratios in humus layers at control and N-fertilised plots (Table 3) indicate that a cumulative amount of 1250 kg (N) ha^{-1} would reduce the C/N ratio by about 4–6 units in southern Sweden. Consequently, sites with a C/N ratio of 28 today might have had a C/N ratio of 33 one hundred years ago. According to Figs. 7 and 8, sites with a humus layer of initially C/N 33 declining to C/N 28 after 100 years should have had average N-use efficiencies of 23 ± 10 (95% c.i.) and 13 ± 5 (95% c.i.) kg (C) kg^{-1} (N) in trees and soils, respectively, estimated for a mean C/N of 30.5. Addition of 1250 kg (N) ha^{-1} implies that *P. abies* sites in southern Sweden, with a humus layer of initially C/N 33, should have accumulated 2.9 kg (C) m^{-2} in tree biomass C and 1.6 kg (C) m^{-2} in soil (forest floor + 10 cm mineral soil), respectively.

Addition of a cumulative amount of 250 kg (N) ha^{-1} in northern Sweden during the last century would have reduced the C/N ratio by about two units from 41 to 39 (mean of C/N ratios at all sites north of the 64th latitude). This would imply N-use efficiencies of about 35 and 10 kg (C) kg^{-1} (N) in trees and soils, respectively. Thus, addition of 250 kg (N) ha^{-1} implies that sites in northern Sweden should have accumulated 0.9 kg (C) m^{-2} in tree biomass C and 0.25 kg (C) m^{-2} in soil. In conclusion, the difference in N deposition implies that *P. abies* sites in southern Sweden should have accumulated 2.0 kg (C) m^{-2} more in tree biomass C and 1.3 kg (C) m^{-2} more in soil, respectively, than northern Sweden during the last century.

According to data from the Swedish National Forest Soil Inventory, Olsson et al. (2007) estimated the difference between forest floor SOC at southern and northern sites at dry/mesic moisture conditions to be 1.7 kg (C) m^{-2}. Our rough estimate of 1.3 kg (C) m^{-2} 100 $years^{-1}$ more in the south than in the north implies that the difference found by Olsson et al. (2007) can to a large extent (70–80%) be explained by the effect of differences in N deposition. These estimates are also consistent with estimates from another part of the LUSTRA project of average national increases in tree C of 23 g m^{-2} $year^{-1}$ and in soil C of 7.5 g m^{-2} $year^{-1}$ over the period 1926–2000 (Ågren et al. 2007).

Conclusions

Our analysis of data from 15 long-term fertilisation experiments at *P. abies* and *P. sylvestris* sites in Sweden and Finland showed that addition of a cumulative amount of 600–1800 kg fertiliser N ha^{-1} for 14–30 years resulted in a mean (±1 SE) increase in tree and soil (organic layer + 10 cm mineral soil) C stock of 25 ± 5 and 11 ± 2 kg (C sequestered) kg^{-1} (N added), respectively. The corresponding estimates for NPK addition were 38 ± 3 and 11 ± 2 kg (C) kg^{-1} (N). These ''N-use efficiencies'' for C sequestration in trees were strongly dependent on soil N status, and increased from close to zero at C/N 25 in the humus layer up to 40 kg (C) kg^{-1} (N) at C/N 35 and decreased again to about 20 kg (C) kg^{-1} (N) at C/N 50 when N only was added. The great difference in N-use efficiency between addition of NPK and N at N-rich sites reflects a limitation of P and K for tree growth at these sites. N-use efficiency for SOC sequestration was, on average, 3–4 times lower than for tree C sequestration, and averaged 13 and 7 kg (C) kg^{-1} (N) for *P. abies* and *P. sylvestris* sites, respectively. The data implies that the C/N ratio in the humus layer can indicate where pure N fertilisation will have small or large effects on tree growth, but also that NPK fertilisation will probably increase tree growth at most sites.

N fertilisation increased SOC sequestration also at N-rich sites, where the tree-growth response was low. This indicates that reduced decomposition rate after N addition has an impact on soil C accumulation. SOC sequestration was, on average, twice as high at *P. abies* as at *P. sylvestris* sites at similar N-use efficiencies for C sequestration (and biomass production) in trees. This might imply that *P. abies* sites have higher litter production and/or lower heterotrophic respiration than *P. sylvestris* sites at equal tree production. However, the conclusion is confounded by the fact that the *P. sylvestris* sites in our study had, on average, coarser texture than *P. abies* sites, and soil texture seems to affect SOC stabilisation.

The estimates of N-use efficiency in relation to C/N ratios further implies that the difference in N deposition (about 10 kg N ha^{-1} $year^{-1}$) between southern and northern Sweden during the 20th century would have led to an extra sequestration of tree C and SOC of about 2.0 and 1.3 kg m^{-2}, respectively, in the south than in the north at least at *P. abies* sites.

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