Temporal Trends in Chemical Parameters of Upland Forest Soils in Southern Finland

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Changes in chemical soil properties during periods of 12 to 28 years were studied in 54 stands in southern Finland. Relative slopes (%/year) were calculated for the changes in soil variables in order to utilise all the sampling occasions (2–6) covered by the study period. Only the results of new analyses made on the soil samples could be used owing to unpredictable differences between the results of the original and new analyses. During the study period the acidity (pH, exchangeable acidity) of the organic layer had decreased, and the mineral soil had become more acidic only in terms of increased exchangeable aluminium concentrations. An increasing trend in the amount of soil organic matter best explained the acidity variables: it lowered acidity in the organic layer, but increased it in the mineral soil. Acid ammonium acetate extractable nutrients showed decreasing trends over time, apart from an increasing trend for sulphur in the 0–30 cm mineral soil layer. Total concentrations of most elements in the organic layer, including nitrogen and sulphur, also showed a decreasing trend. Changes in the soil variables could not be firmly connected to deposition, wood production or the amount of nutrients accumulated in woody tissues. However, the decrease in sulphur concentrations in the organic layer was clearly linked with the decrease in sulphur deposition in recent years.

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1 Introduction

Forest soils become gradually acidified over time as a result of e.g. soil forming processes (podzolisation) (Aaltonen 1935, Jauhiainen 1969), tree stand development (Tamminen 1991, Brais et al. 1995), biomass harvesting (Staaf and Olsson 1991, Olsson et al. 1996), and the deposition of acidifying compounds (Ulrich et al. 1979, Berdén et al. 1987). In Nordic conditions, i.e. in a cool, humid climate with a glaciated terrain that is primarily covered by conifers, the soils have gradually been weathered and leached, and organic matter has accumulated on and in the surface soil. These soils have been continuously subjected to acidification processes since the end of the last ice age. Slow, and therefore relatively undetectable, changes will continue up until the next glaciation. The time scales of soil acidifying processes vary considerably. In Finland, soil-forming processes have been modifying the soil over the last 8000–10 000 years. Tree stand development takes place within a relatively short period of time, ca. 70–150 years, and forest soils have been subjected to anthropogenic acidifying deposition since ca. 1950. Intensive biomass harvesting in the forests has had an acidifying effect ever since the beginning of the last century and will undoubtedly continue to do so in the future.

Changes in the soil associated with podzolisation initially occur at a relatively rapid rate, but then gradually slow down (Jauhiainen 1973, Starr 1991, Alexander and Burt 1996), i.e. approximately as a logarithmic function of soil age. In soils over 10 000 years old, changes in soil properties due to soil-forming processes are assumed to be minimal. Changes in soil properties related to tree stand ageing and growth can be explained on the basis of slower mineralization of organic matter due to cooler, moister soil (Bhatti et al. 2000), and perhaps to the occurrence of deeper, longer-lasting soil frost caused by the shading of the tree canopies (Yli-Vakkuri 1960, Päivänen 1973), on the basis of nutrient accumulation in tree biomass, the exudation of acidic material (H^+ and organic compounds) by tree roots (Gobran et al. 1998), the accumulation of acidic coniferous litter on and in the soil (Nihlgård 1971), and more acidic soil water percolating down through the forest floor (Alriksson and Olsson 1995, Brais et al. 1995, Krause 1998, Griffiths and Swanson 2001). The organic layer becomes thicker, the organic and uppermost mineral soil layers become more acidic, and the nitrogen and base cation concentrations decrease (Tamminen 1991, Brais et al. 1995). Intensive biomass harvesting reduces the amount of nutrients (e.g. Ca and Mg) in the soil, thus increasing acidity (Olsson et al. 1996, Kukkola and Mälkönen 1997). A high acidifying load also decreases base cation pools in the soil through the leaching of cations together with sulphate anions (Berdén et al. 1987, Hallbäcken 1992). A process opposite to soil acidification, i.e. neutralization, has been reported some years after clear-cutting (Nykvist and Rosén 1985), and also after the cessation of an artificial acid rain treatment (Nykvist and Rosén 1985, Stuanes et al. 1992, Rustad et al. 1996).

Short-term (10–20 years) changes are especially visible in former agricultural land, but they have also been reported in old forest soil (Alriksson and Olsson 1995, Farmer 1995, Rhoades and Binkley 1996). Medium to long-term (20–100 years) changes were reported in the 1990's in conditions similar to the boreal forests in Finland (Hallbäcken and Tamm 1986, Berdén et al. 1987, Sjöström and Qvarfort 1992). In Finland we have only weak evidence of long-term changes in soil parameters, e.g. in pH (Ahokas 1997, Kukkola and Mälkönen 1997). On the other hand, some modelling studies have predicted alarming future trends in the base cation status of forest soils (Hari et al. 1986, Johansson and Savolainen 1990).

The reasons underlying soil acidification in Sweden have been reviewed by Binkley and Högberg (1997). They concluded that much of the decline in pH_{H2O} was either associated with stand age or with changes in the ionic strength of the soil solution, as well as changes in the acid strength of the exchange complexes and decreased base saturation (Binkley and Högberg 1997). The conclusions of Binkley and Högberg could very well apply to southern Finland, where sulphur and nitrogen deposition has been relatively low. Sulphur deposition in 1980 ranged from about 4 to 13 kg ha^{-1} and in 1997 from about 2 to 3 kg ha^{-1}. Nitrogen (ammonium + nitrate) deposition in 1980 was about 4 to 12 kg ha^{-1}, and in 1997 about 2 to 6 kg ha^{-1} (Nordlund 2000, see also De Vries et al. 2001). Annual calcium, magnesium

and potassium deposition is also extremely low, 0.1 to 1 $kg\,ha^{-1}$ (Piirainen 2002). Because of the relatively low precipitation (400–750 mm) and low acidifying deposition, the leaching of nitrogen and base cations in Finland is also very low: 0.1 to 1.5 $kg\,ha^{-1}\,a^{-1}$ (Helmisaari and Mälkönen 1989, Piirainen 2002). The largest sink for soil nutrients seems to be the tree biomass. Commercial tree stands incorporate about 4–15 $kg\,ha^{-1}\,a^{-1}$ of N in the woody tissues (trunk, branches, coarse roots), 0.5–1.5 $kg\,ha^{-1}\,a^{-1}$ of P, 3 to 6 $kg\,ha^{-1}\,a^{-1}$ of K, and 4 to 11 $kg\,ha^{-1}\,a^{-1}$ of Ca (Mälkönen 1974, 1977, Hallbäcken and Bergholm 1998, Ingerslev and Hallbäcken 1999, Helmisaari et al. 2002). The weathering rates of cations (Ca, K, Mg) in southern Finland seem to be high enough to satisfy tree growth, because neither liming (even with Mg-rich limestone) nor potassium fertilization have had any effect on tree growth on mineral soils (Derome et al. 1986).

Studies carried out in the Nordic countries (Tyler et al. 1985, Hallbäcken and Tamm 1986, Falkengren-Grerup 1987, Falkengren-Grerup 1990, Falkengren-Grerup and Tyler 1991, Hallbäcken 1992), in Central Europe (Billett et al. 1988, Billett et al. 1990), in the USA (e.g. Binkley and Valentine 1991, Knoepp and Swank 1994, Drohan and Sharpe 1997) and in Canada (e.g. Brand et al. 1986, Brais et al. 1995), give good grounds to hypothesise that forest soils in southern Finland have undergone acidification, i.e. acidity has increased and base cation concentrations have decreased, during the last decades. According to Brais et al. (1995), this may also be the case for nitrogen and phosphorus. The rapid decline in the deposition of sulphur in recent years should also have decreased soil sulphur (Savolainen and Tähtinen 1990, Nordlund 2000). Hypotheses about the causes of soil acidification, e.g. acidifying deposition or the immobilisation of nutrients in the woody tissues of trees, have been verified to some extent in more polluted conditions with higher stand productivity (Tamm and Hallbäcken 1988, Hallbäcken 1992), but not in Finnish conditions.

The aim of this study is to test these hypotheses by estimating the direction and magnitude of changes in soil parameters during a period of 12–28 years, and to investigate the correlation between any such changes with soil, site, tree stand and deposition parameters.

2   Material and Methods

The study material consisted of soil samples taken from the control plots (25–30 m × 25–35 m) of fertilisation experiments established by the Finnish Forest Research Institute during 1958–1980 (Fig. 1). As most of the stands were relatively young at the time the experiments were started (Appendix 1), the 12- to 28-year periods covered by soil sampling mainly represented the first half of tree stand rotation. The experiments were located in 35 Scots pine (*Pinus sylvestris*), 15 Norway spruce (*Picea abies*), three Siberian larch (*Larix sibirica*) and one silver birch (*Betula pendula*) stand.

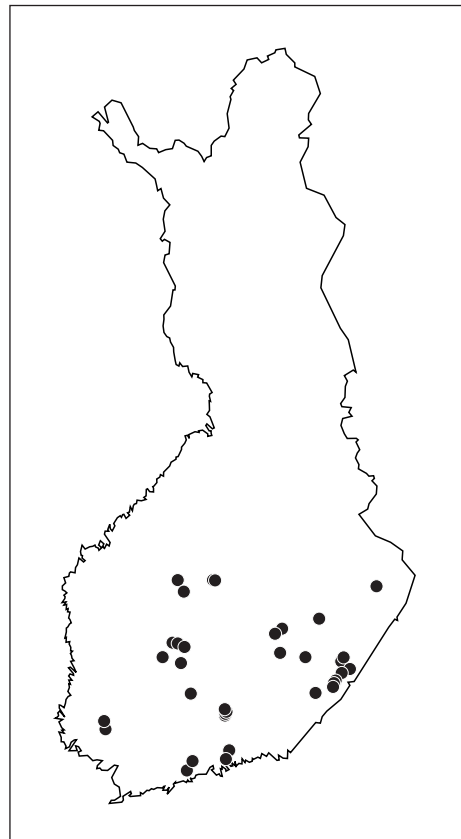During the period 1965 to 1977 the plots were sampled at relatively random intervals, and sys-

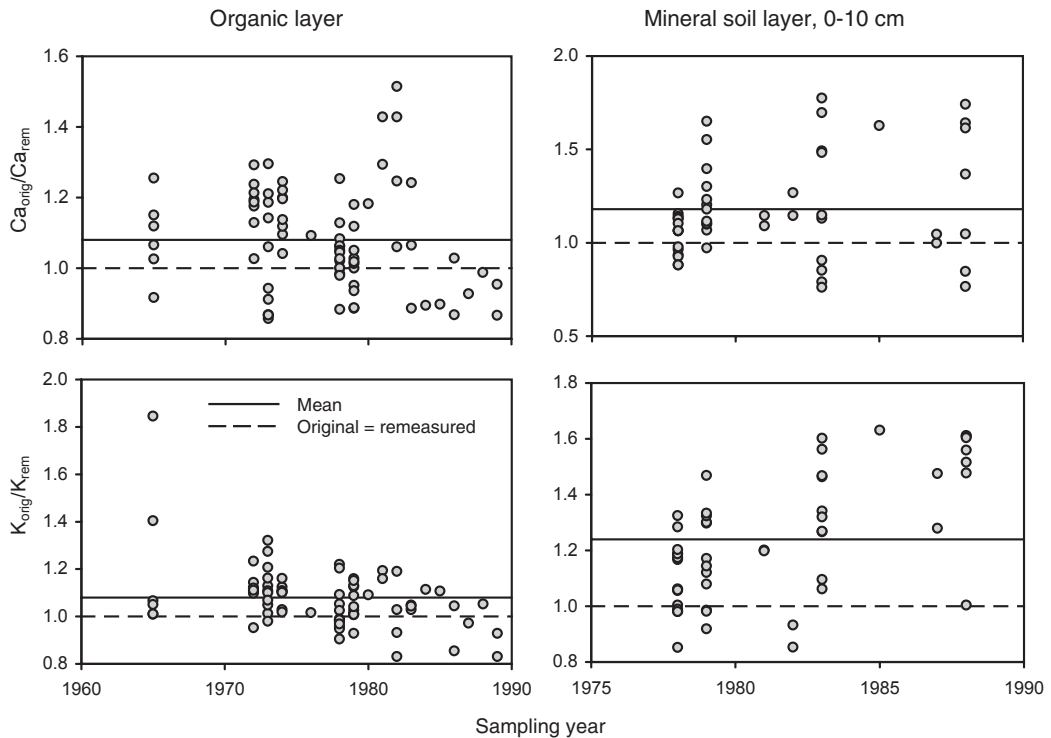**Fig. 1.** Location of the fertilisation experiments with control plots.

Fig. 2. The ratios between the original and remeasured exchangeable calcium and potassium concentrations as a function of sampling year in the organic layer and in the 0–10 cm mineral soil layer.

tematically (4- to 10-year intervals) after 1977. All the experiments were sampled in 1993 for the organic layer and either the 0–10 and 0–30 cm mineral soil layers depending on which mineral soil layer had been sampled on earlier occasions (see Appendix 1).

The average age of the tree stands in 1993 was 60 years. The soils were coarse or medium coarse (coarse sand–sandy loam) material, with an average organic layer thickness of 33 mm (13–56 mm), and an average volumetric stone content (Viro 1952, Tamminen 1991) of 20% (0–76) (Appendix 1).

Deposition of Sulphur and Nitrogen

Deposition data for the year 1986 for the network of 3000 National Forest Inventory systematic sample plots were provided by the Finnish Meteorological Institute (Tuovinen et al. 1990, Nord-

lund 2000). The deposition data corresponding to the location of the fertilisation experiments were interpolated from this network. Annual bulk deposition of SO_4-S in each experiment was estimated by adjusting the deposition values for 1986 according to the trend in annual emissions in Finland for the periods covered by soil sampling (Savolainen and Tähtinen 1990, Anttila 1991, Nordlund 2000). Annual inorganic nitrogen (NH_4-N + NO_3-N) deposition was assumed to have remained at the same level during the study period (Nordlund 2000). The average and cumulative total deposition during the study period were then calculated from these estimates (Appendix 2).

Stand Growth and Nutrient Stores in the Trees

Tree stand growth and total stem wood production

were estimated according to Vuokila and Väliaho (1980), and the amount of nutrients incorporated in woody biomass production (stem and branch wood and bark) according to Mälkönen (1975) (see Appendix 2). The experiments were thinned manually at 10- to 15-year intervals, but only merchantable stemwood was removed from the site.

Soil Sampling

Soil samples were taken, as far as possible, in a similar fashion as on earlier occasions. The organic layer was sampled with a steel cylinder ($d = 58$, earlier $d = 50$–58 mm) from 20 systematically located spots along the sides of the net plot. The mineral soil samples 0–10 cm were taken at the same spots with an auger ($d = 23$ mm) or on stony plots at 8 spots with a spade. Earlier sampling was done at systematically located spots (15–25) inside the net plot. The 0–30 cm mineral soil samples were taken at the corners of the net plot like on earlier occasions. The sampling time varied randomly during the study period from May to October. The most recent samples were collected in September and October 1993. 39% of the earlier samplings occurred in these two months.

Pretreatment and Chemical Analysis

During 1965–1981 the samples were air-dried at room temperature, and after 1982 in a ventilated chamber at a temperature of 30–40 °C. The organic samples were ground in a mill with a 2 mm bottom sieve, and the mineral soil samples were passed through a 2 mm sieve to remove stones and larger roots. The air-dry samples were stored at room temperature.

A preliminary study involving reanalysis of a sub-set of the samples indicated that there were unpredictable differences between the results of the original analyses, carried out within 1–2 years after sampling, and those of the new analyses (Fig. 2). All the samples were therefore reanalysed in 1993 and the results used in this study. However, the original pH_{H2O} values were used here because re-measurement of the old samples in 1993 gave average values that were system-

atically below those of the original pH measurement: 0.24 pH units for the organic layer, 0.22 for the 0–10 cm, and 0.20 units for the 0–30 cm mineral soil layer.

The organic matter content was determined as mass loss-on-ignition (550 °C/3 h). Carbon and total nitrogen were determined on the organic layer samples with a CHN analyser (Leco CHN-600), and nitrogen on the mineral soil samples by the Kjeldahl method. Particle size analysis was performed during 1965–79 by pipetting and sieving (Elonen 1971). pH was determined in a water suspension with a ratio of 15 ml of sieved sample and 25 ml of distilled water. Exchangeable Ca, Mg, K and Na and extractable P and S were determined by extraction with acidic ($pH = 4.65$) ammonium acetate (Halonen et al. 1983) and exchangeable acidity and aluminium with 1 M KCl using a ratio of 15 ml of sample and 150 ml of extractant. The suspensions were left to stand overnight before being shaken for 1 h and then filtered. The concentrations of Ca, Mg, K, Na, P and S in the extract were determined by ICP/AES (ARL 3580). Exchangeable acidity (EA) was determined from a 50 ml KCl extract by titration with 0.05 M NaOH to an endpoint of pH 7.0. Exchangeable Al was determined by back-titration with 0.02 N H_2SO_4 to pH 7.0 after 10 ml of 4% NaF solution had been added to the extract (Halonen et al. 1983). The pH of the KCl extract was determined at the same time. Cation exchange capacity (CEC, mmol kg^{-1} d.m.) was calculated as the sum of exchangeable cations and exchangeable acidity, $CEC = Ca^{2+} + K^+ + Mg^{2+} + Na^+ + EA^+$, and base saturation (BS) as the proportion of base cations out of CEC. Total concentrations in the organic layer samples were determined by extracting the ashed samples (550 °C for 3 h) with HCl (Halonen et al. 1983), and determining the concentrations of Ca, Mg, K, Na, P and S by ICP/AES.

Statistical Analyses

The organic layer on each control plot was represented on the average by 2.4 (2–6) samples, the 0–10 cm mineral soil layer by 2.7 (2–4) samples, and the 0–30 cm mineral soil layer by 2 samples per study period (see Appendix 1). In order to be able to utilize all the information from the

individual sampling occasions (2–6), the change in a soil variable y (OM, pH, H^+, Al^{3+}…, N) was depicted by means of the slope parameter b of the regression model $y = a + b \cdot x + \varepsilon$, where x is the sampling year. The regression coefficient b was divided by the mean value of all the observations y on the plot in question and multiplied by 100 to obtain a relative regression coefficient b_r (Eq. 1) expressing the relative annual change percentage the of soil variable during the study period.

$$b_r = \frac{100 \cdot b}{\bar{y}} \qquad (1)$$

where

b = the regression coefficient

$$\bar{y} = \frac{\sum y_i}{n}$$

y_i = the value of a variable at a sampling occasion i

The relative annual change, i.e. relative annual trend, was calculated on the basis of the mean and not on the first value of the study period. As the concentration mean values $\bar{y}$ for each variable are given in the tables, the annual change b in original concentration units for a chosen time period can be calculated from b_r values.

The statistical hypothesis that there was no change in a soil variable, i.e. $\bar{b}_r = 0$, was tested with the t test. The statistical relationships between the soil variable changes and other variables were tested by Pearson correlation coefficients.

3 Results

3.1 Organic Matter

The organic matter content showed increasing trends over time in the organic layer and in the 0–30 cm mineral soil layer, but no trend in the 0–10 cm mineral soil layer (Table 1). The trend in organic matter in the organic layer correlated negatively with organic layer thickness (r = –0.45[**]), i.e. the organic matter content had increased less on sites with a thick organic layer than on sites with a thin layer, and latitude (–0.38[**]), and positively with the estimated average annual

nitrogen deposition (0.47[***]) ([*]$p < 0.05$, [**]$p < 0.01$, [***]$p < 0.001$).

3.2 Acidity

The pH_{H2O} remeasured in 1993 was unexpectedly higher than that obtained at earlier samplings (Table 2). The trends were calculated on the basis of all the observations available from these plots using the original pH measurements. The pH trends in the organic and in the 0–10 cm layer were not correlated with any variable, but in the 0–30 cm mineral soil layer the pH change was correlated negatively with the trend in the organic matter content (–0.54[**]), i.e. pH tended to decrease along with an increasing organic matter content.

The pH of the KCl extract gave a somewhat different picture compared to the pH measured in water. pH_{KCl} showed a rising trend over time in the organic layer, no trend in the 0–10 cm mineral soil layer, and a weakly declining trend in the 0–30 cm mineral soil layer (Table 3). Exchangeable acidity and exchangeable aluminium concentrations confirmed the decreasing acidity trend in the organic layer, but in the mineral soil there were no trends for exchangeable acidity, while exchangeable aluminium showed statistically significant increasing trends (Table 3).

The trend for the pH_{KCl} of the organic layer correlated negatively with fine fractions in the mineral soil (–0.42[**]), and the trends in exchangeable acidity and aluminium correlated negatively with the trend in the organic matter content (–0.55[***] and –0.64[***]). In the 0–10 and 0–30 cm mineral soil layers only the trend in the organic matter content correlated with the trends of exchangeable acidity (0.45[*] and 0.66[***]) and exchangeable aluminium (0.50[**] and 0.73[***]).

3.3 Exchangeable and Extractable Nutrients

The trends for the organic layer were calculated using organic matter-based concentrations because the plot-wise coefficients of variation were lower when calculated on an organic matter than on a dry matter basis. All the nutrients, except for K, followed a decreasing trend in the organic layer

Table 1. Average organic matter content ($\bar{y}$, %) and its average relative annual change ($\bar{b}_r$, % a^{-1}).

Layer	Time, a[a]	n[b]	$\bar{y}$	$\bar{b}_r$	t[c]	p[d]
Organic	19 (12–28)	54	58.6	1.14	4.15	0.000
0–10 cm	15 (12–15)	30	5.5	0.42	1.54	0.135
0–30 cm	20 (14–28)	32	3.6	0.60	2.90	0.007

[a] Median and range (in parentheses) of the study period
[b] Number of plots

[c] $t = \dfrac{\bar{b}_r - \mu_0}{s / \sqrt{n}}$, where $\mu_0 = 0$

[d] p is the significance level calculated from the t distribution

Table 2. Average pH$_{H2O}$ ($\bar{y}$) and its average relative annual change ($\bar{b}_r$, % a^{-1}). Means and trends are based on the original measurements.

Layer	Time, a	n	$\bar{y}$	$\bar{b}_r$	t	p
Organic	14–28	54	3.98	0.18	3.45	0.001
0–10 cm	12–15	30	4.64	0.34	5.12	0.000
0–30 cm	14–28	32	4.83	0.10	2.08	0.046

Table 3. Average pH$_{KCl}$, exchangeable acidity (EA) and exchangeable aluminium concentrations ($\bar{y}$) and their average relative annual changes ($\bar{b}_r$, % a^{-1}).

	Layer	Time, a	n	$\bar{y}$	$\bar{b}_r$	t	p
pH$_{KCl}$	Organic	12–28	54	3.20	0.12	4.01	0.000
	0–10 cm	12–15	30	3.84	0.04	0.74	0.466
	0–30 cm	14–28	32	3.98	–0.06	–2.02	0.053
EA,	Organic	12–28	54	124	–3.62	–13.16	0.000
mmol(+) kg^{-1}	0–10 cm	12–15	30	22	–0.12	–0.38	0.705
	0–30 cm	14–28	32	14	0.17	0.48	0.634
Exch. Al,	Organic	12–28	54	34	–3.19	–6.52	0.000
mmol(+) kg^{-1}	0–10 cm	12–15	30	18	0.94	3.46	0.002
	0–30 cm	14–28	32	11	0.98	2.81	0.009

(Table 4). The Na concentrations decreased very steeply in all three soil layers (–3…–7%/year) and they were very high at the beginning of the study period. The decreasing trend in extractable S in the organic layer was steep and statistically the most significant of all the trends. In the mineral soil there were negative or zero trends for Ca, Mg, K and Na and P, but a positive trend for S in the 0–30 cm layer.

In the organic layer the trend in exchangeable Mg was negatively correlated with the proportion of fine fractions in the mineral soil (–0.52[***]) and wood production during the monitoring period (–0.39[**]). The trend in extractable P correlated positively with the trend in the organic matter content (0.44[**]), and the trend in S correlated positively with the sulphur deposition sum (0.54[***]) and the length of the study period (0.67[***]). The longer was the period covered by sampling, i.e. the earlier the first sampling occurred, the smaller was the

Table 4. Average exchangeable Ca, Mg, K and Na and extractable P and S concentrations ($\bar{y}$, mg kg^{-1}) and their average relative annual changes ($\bar{b}_r$, % a^{-1}). Concentrations in the organic layer were calculated on an organic matter basis, and in the mineral soil layers on a dry matter basis.

	Layer	Time, a	n	$\bar{y}$	$\bar{b}_r$	t	p
Ca	Organic	12–28	54	3550	–0.98	–5.45	0.000
	0–10 cm	12–15	30	118	–1.15	–2.57	0.016
	0–30 cm	14–28	32	62	–0.04	–0.10	0.923
K	Organic	12–28	54	993	–0.26	–1.46	0.150
	0–10 cm	12–15	30	30	–0.25	–0.78	0.441
	0–30 cm	14–28	32	14	–0.64	–2.45	0.020
Mg	Organic	12–28	54	441	–0.64	–4.78	0.000
	0–10 cm	12–15	30	15	–0.56	–1.55	0.133
	0–30 cm	14–28	32	8	0.01	0.05	0.963
Na	Organic	12–28	54	91	–5.48	–10.79	0.000
	0–10 cm	12–15	30	11	–7.40	–11.59	0.000
	0–30 cm	14–28	32	6	–3.35	–7.73	0.000
P	Organic	12–28	54	363	–2.87	–12.25	0.000
	0–10 cm	12–15	30	8	–1.58	–3.11	0.004
	0–30 cm	14–28	32	6	0.15	0.49	0.631
S	Organic	12–28	54	287	–4.47	–24.46	0.000
	0–10 cm	12–15	30	40	0.72	1.45	0.158
	0–30 cm	14–28	32	40	0.64	2.35	0.026

decrease in the S concentration. The relative trend (% a^{-1}) for domestic SO_2 emissions, calculated on the basis of the data of Savolainen and Tähtinen (1990), correlated relatively well with the relative trend for organic layer S concentrations:

	1965→93	1973→93	1978→93	1979→93
SO_2 emissions	–0.8	–4.0	–4.9	–5.3
Organic layer S	–2.7	–4.1	–4.9	–5.2

In the 0–10 cm mineral soil layer the trend in the organic matter content correlated positively with the trends for the K (0.81[***]), Mg (0.73[***]), P (0.62[***]) and S (0.47[**]) concentrations. The thickness of the organic layer correlated negatively with the trends for Mg (–0.57[**]) and P (–0.58[**]). In the 0–30 cm mineral soil layer the trend in the exchangeable K concentration correlated negatively with latitude (–0.52[**]), and positively with the average annual nitrogen deposition (0.57[**]).

3.4 Cation Exchange Capacity and Base Saturation

The cation exchange capacity (CEC) followed a negative trend in the organic layer, but no trend in the mineral soil layers (Table 5). Base saturation (BS) followed a positive trend in the organic layer and a negative trend in the 0–10 cm mineral soil layer.

The trend in the organic matter content almost completely explained the trend in CEC in the organic layer (r=0.93[***]) but less strongly, although significantly, in the mineral soil layers (r=0.54[**] in the 0–10 cm, and r=0.68[***] in the 0–30 cm layer). The trend in BS in the organic layer correlated negatively with total wood production (–0.47[**]) and the length (–0.37[**]) of the study period. The trend in BS in the 0–30 cm mineral soil layer correlated negatively with basal area in the spruce experiments (–0.50[**]).

Table 5. Average values of cation exchange capacity (CEC, mmol kg^{-1} d.m.) and base saturation (BS, %) and their average relative annual changes ($\bar{b}_r$, % a^{-1}).

	Layer	Time, a	n	$\bar{y}$	$\bar{b}_r$	t	p
CEC	Organic	12–28	49	209	–0.83	–3.34	0.002
	0–10 cm	12–15	29	30.2	–0.37	–1.47	0.152
	0–30 cm	14–28	32	18.2	0.03	0.10	0.922
BS	Organic	12–28	49	65.0	1.11	9.84	0.000
	0–10 cm	12–15	29	25.5	–1.12	–2.77	0.010
	0–30 cm	14–28	32	23.3	–0.48	–1.68	0.102

3.5 Total Element Concentrations in the Organic Layer

Total element concentrations were expressed on an organic matter basis because the important nutrients Ca, N, P and S were found to have the lowest coefficient of variation during the study period when calculated on this basis, and there was no difference in the coefficient of variation of concentrations calculated on a dry or organic matter basis for the other elements, except for Mg. This procedure also eliminated the direct effect of the increasing trend in the organic matter content (see Table 1).

All the elements showed a declining trend (Table 6). The trends in total concentrations correlated the strongest with the trend in the organic matter content: K –0.71[***], Mg –0.82[***], Na –0.41[**] and P –0.56[***], i.e. an increase in the organic matter content meant a decrease in the total concentrations (expressed as mg/kg o.m.). The trends in the Mg and P concentrations correlated negatively with the average annual nitrogen

Table 6. Average total element concentrations ($\bar{y}$, mg kg^{-1} o.m.) and their average relative annual changes ($\bar{b}_r$, % a^{-1}) in the organic layer (n = 54).

	$\bar{y}$	$\bar{b}_r$	t	p
Ca	5170	–0.67	–3.80	0.000
K	1250	–0.47	–2.22	0.031
Mg	1330	–1.20	–3.70	0.000
Na	190	–2.07	–5.71	0.000
P	1420	–0.71	–5.82	0.000
S	1190	–0.55	–4.97	0.000

deposition (–0.47[***] and –0.41[**], respectively), and the trend of S negatively with the total nitrogen and sulphur deposition (–0.41[**] and –0.35[**]).

The concentration trends calculated on a dry matter basis were rather different compared to the organic matter-based concentrations, as the following frequencies show:

Trend ($\bar{b}_r$)	Dry matter	Organic matter
Negative	1	6
Zero	3	-
Positive	2	-
Total	6	6

The total nitrogen concentration in the organic layer followed a decreasing, and the C/N ratio an increasing trend (Table 7). In the mineral soil there were no trends in total nitrogen, except for a negative trend in the total N concentration expressed on an organic matter basis in the 0–30 cm mineral soil layer (Table 7).

In the organic layer the trend in the total nitrogen concentration did not correlate with any variable. In the mineral soil the total nitrogen concentration (d.m.) correlated with the trend in the organic matter content in both the 0–10 cm (r = 0.78[***]) and 0–30 cm layers (r = 0.80[***]).

4　Discussion

The forest soils investigated in this long-term study had, on the average, not become more acidic in terms of pH or exchangeable acidity, but there was an increase in exchangeable aluminium in

Table 7. Average total nitrogen concentration ($\bar{y}$, g kg^{-1}) and the C/N ratio, and their average relative annual changes ($\bar{b}_r$, % a^{-1}).

Layer		Time, a	n	$\bar{y}$	$\bar{b}_r$	t	p
Organic	N, o.m.	12–28	54	17.9	–0.33	–3.53	0.001
	C/N [a]	12–28	54	35.0	0.25	2.93	0.005
0–10 cm	N, d.m.	12–15	30	1.0	0.27	1.10	0.282
	N, o.m.	12–15	30	17.6	–0.16	–0.87	0.394
0–30 cm	N, d.m.	14–28	32	0.7	–0.01	–0.03	0.975
	N, o.m.	14–28	32	18.8	–0.61	–4.71	0.000

[a] Only organic layer samples were analysed for total carbon.

the mineral soil and a reduction in base cations. In contrast, numerous studies have reported soil acidification on the basis of changes in pH and exchangeable acidity (Hallbäcken and Tamm 1986, Billett et al. 1988, Ahokas 1997).

There was an increasing trend in pH$_{H2O}$ in all the soil layers. Sampling the organic layer is a notoriously difficult and extremely subjective procedure. The border between the organic layer and the underlying mineral soil is frequently indistinct; inclusion of varying amounts of mineral soil, for instance, will have a considerable effect on the results when expressed on a dry matter basis. According to the field sampling instructions, a considerable amount of the dry litter (L horizon) on the forest floor was discarded and part of the underlying mineral soil included in samples taken prior to 1985. In the sampling carried out in 1993, on the other hand, only living (green) plant material was discarded (i.e. the L horizon was included) and the border between the organic layer and mineral soil was selected so as to exclude mineral soil. This change in the sampling procedure could account for the apparent increase in pH$_{H2O}$ because Skyllberg (1990), for instance, reported that there is a clear decrease in pH with increasing depth in the organic layer. The increasing pH trends in the mineral soil cannot be explained on the basis of differences in sampling, even though pH increases rapidly in the uppermost layer of podzolised mineral soils (Aaltonen 1935, 1939, 1941, Franzmeier and Whiteside 1963, Jauhiainen 1969, Starr and Tamminen 1992). If the positive trends in pH$_{H2O}$ were due to artefacts, then the acidification rate has, in any case, been low.

Acidification in the mineral soil was clearly apparent as an increasing trend of exchangeable Al in both the 0–10 and 0–30 cm mineral soil layers. However, there were no corresponding trends in exchangeable acidity in the mineral soil, even though exchangeable Al is the main component of exchangeable acidity in the mineral soil (Tamminen and Starr 1990). In this material, the average proportion of exchangeable Al out of exchangeable acidity was 24% in the organic layer, 78% in the 0–10 cm mineral soil layer, and 85% in the 0–30 cm layer. In most cases the trend in acidity correlated with the trend in organic matter content. An increasing trend in the organic matter content in the organic layer meant a lower level of acidity, but in the mineral soil correspondingly a higher level of acidity. The acidity variables were not correlated with the length of the monitoring period, the geographical location (cf. Hallbäcken and Tamm 1986), modelled deposition of sulphur or nitrogen (cf. Berdén et al. 1987) nor with tree growth (cf. Hallbäcken 1992).

The reducing trends for exchangeable and extractable nutrient concentrations in the organic layer corresponded rather well with corresponding values calculated from the report of Billett et al. (1990), except for Na. The concentrations of exchangeable Ca, K and Mg had declined in these Scottish sites on the average by 0.7 to 1% per year over a period of 38 years. However, base cation concentrations in the mineral soil had declined in Scotland by over 2% per year, compared to 0 to 1.2% per year in our study. The base cation concentrations in the C horizon had declined by 1.7 to 2.1% per year in southern Sweden over a period of 36 to 41 years (Falkengren-Grerup and Tyler

1992). In this Swedish material, Na had decreased by 4.1% per year, which is in good agreement with the steep Na trends in our study. The annual change in the extractable S concentration was +0.9% in the C horizon in Sweden (Falkengren-Grerup and Tyler 1992) and +0.6% in the 0–30 cm mineral soil layer in our material.

The decrease in base cations ($Ca^{+2}+K^{+}+Mg^{2+}$) seemed to be similar in the organic and 0–10 cm mineral soil layer, but lower in the deeper 0–30 cm layer, i.e. on the average –0.8, –1.0 and –0.1 % a^{-1}, respectively. Nutrients may have been taken up by the trees more efficiently from the organic and the topmost mineral soil layer, where fine roots are concentrated (Helmisaari and Hallbäcken 1997). This is also supported by the depth profile of phosphorus trends, i.e. –2.9 for organic, –0.6 for 0–10 cm and 0.2 % a^{-1} for 0–30 cm layer (Table 4).

The clear decrease in the sulphur concentration in the organic layer is most likely caused by reductions in S emissions and deposition (Savolainen and Tähtinen 1990, Ukonmaanaho et al. 1998). However, there was an increasing trend in the extractable S concentration in the 0–30 cm mineral soil. The B horizon in the mineral soil has a relatively high sulphate-adsorption capacity (Singh 1980, Autry et al. 1990, Vannier et al. 1993), and the retention of sulphur in the aboveground parts of the ecosystem and its subsequent release over the years could explain the continuous accumulation of sulphur in the mineral soil.

Base saturation showed an increasing trend in the organic layer (Table 5), despite the decline in base cation concentrations, but the increase was in line with the increased pH and decreased exchangeable acidity. In the mineral soil, on the other hand, decreased base saturation, together with the increased aluminium concentration, indicated acidification (Tables 3 and 5). The trend in the organic matter content explained a high proportion of the variation in the trend in CEC. The negative correlation between the trend of base saturation in the organic layer and wood production during the monitoring period supported the hypothesis that wood production and the retention of base cations in the woody biomass may acidify soil as a result of the uptake of base cations and corresponding release of protons into the soil (Hallbäcken 1992).

The decreasing trends of total elemental con-

centrations were partly explained by the increasing trend in the organic matter content, except for the trends for calcium and sulphur. The hypothesis that S and N deposition may decrease the amount of soil base cations in the soil was supported only by the correlation between the trend in the total magnesium concentration and annual nitrogen deposition. The trends for total P and S concentrations also seemed to be affected negatively by deposition. The trends in the total and extractable concentrations were rather similar, but the extractable concentration trends were steeper for Ca, Na, P and S, and the total concentration trends steeper for K and Mg (Tables 4 and 6). However, they both confirm that there was a decline in these nutrients during the monitoring period.

Total N concentrations expressed on an organic matter basis in the organic layer followed decreasing trends, which is completely opposite to findings reported in Germany (Buberl et al. 1994). In Germany, the C/N ratio is supposed to have decreased as a result of an increase in the N concentration in the organic matter due to nitrogen deposition. However, N deposition in Germany has been 5 to 10 times higher than that in Finland (Kauppi et al. 1990). Mälkönen et al. (1990) reported that nitrogen fertilization, at average total doses of 315 kg N/ha during a ca. 20 year period, decreased the C/N ratio in Scots pine stands in Finland, but not in the more fertile spruce stands. The declining trends in nitrogen found in our study correspond to those associated with normal stand development (Brais et al. 1995). We can thus conclude that nitrogen deposition has not had any effect on the trends in soil nitrogen. The effects of other variables, such as geographical location or tree stand properties or development, did not explain the trends in soil nitrogen.

Soil changes, i.e. mainly a decrease in nutrient concentrations and an increase in exchangeable aluminium in the mineral soil, were only weakly correlated to site and stand variables. The deposition of sulphur and nitrogen had possibly influenced some soil changes, e.g. the increase in soil organic matter. However, because of the close relationship between latitude, which correlates strongly with temperature, primary production (Koivisto 1970) and to some extent also with soil nutrients (Tamminen 2000), and the deposition of sulphur and nitrogen in Finland (correla-

tion between estimated deposition of S or N and latitude was 0.90–0.95), the use of correlations to decide which variables have a primary influence on changes in nutrient concentrations was considered to be unjustified.

The effect of tree species (here pine vs. spruce) on soil changes could not be determined. In fact, it was difficult to study these changes, because of the correlation between tree species and site fertility, i.e. pine stands on infertile and coarse-grained and spruce stands on more fertile and finer-grained soils. Stand development, which is characterized by the cumulative production of woody biomass, increasing amounts of nutrients moving from the soil to the biomass and litter accumulation, was hypothesised to affect soil changes. However, the evidence for this was weak, only the trends of exchangeable magnesium and base saturation correlated with wood production (cf. Hallbäcken 1992) but not, for instance, the trends of nitrogen, phosphorus or potassium.

The main findings reported in long-term studies of this sort have been a relatively large decline in base cation concentrations over periods of 20–50 years (Billett et al. 1990, Falkengren-Grerup and Tyler 1992, Hallbäcken 1992), as well as within short periods of time (10–15 years) (Troedsson 1985, Grimm and Rehfuess 1986). However, large relative changes in soil variables, i.e. of 1 to 5% per year, do not appear to be very plausible, especially in the mineral soil. The question inevitably arises of whether factors connected to soil sampling (Skyllberg 1990), pretreatment (Payne and Rechgigl 1989, Saarinen 1989), storage (Falkengren-Grerup 1995, Ogner et al. 2001) and analysis are the primary causes of the decreasing trends reported for base cation concentrations when old and new samples are analysed at the same time.

5 Conclusions

The organic matter content had increased in the organic and 0–30 cm mineral soil layers, and this had influenced the trends in most of the other soil variables studied.

Signs of forest soil acidification during the 12- to 28-year study period included a slight decrease in the pH_{KCl} values in the 0–30 cm mineral soil layer, an increase in exchangeable aluminium in the mineral soil, decreasing trends in base cation concentrations especially in the organic layer, and a decrease in base saturation in the mineral soil. On the other hand, the pH and exchangeable acidity values did not indicate any acidification.

Decreasing trends of extractable and total sulphur in the organic layer correlated with the decreasing trend in sulphur deposition in Finland since the 1980's. Negative or zero trends for total N suggest that there is no threat of nitrogen saturation in coniferous forests in southern Finland in the foreseeable future.

The changes in soil variables, apart from soil sulphur, could not be systematically explained on the basis of sulphur or nitrogen deposition. Annual tree growth, cumulative wood production or the amount of nutrients retained in the woody biomass did not explain the trends in soil variables, except for exchangeable magnesium and base saturation in the organic layer.

References

Aaltonen, V.T. 1935. Zur Stratigraphie des Podsolprofils besonders von Standpunkt der Bodenfruchtbarkeit I. Communicationes Instituti Forestalis Fenniae 20(6). 150 p.

— 1939. Zur Stratigraphie des Podzolprofils besonders von Standpunkt der Bodenfruchtbarkeit II. Communicationes Instituti Forestalis Fenniae 27(4). 133 p.

— 1941. Zur Stratigraphie des Podzolprofils besonders von Standpunkt der Bodenfruchtbarkeit III. Communicationes Instituti Forestalis Fenniae 29(7). 47 p.

Ahokas, H. 1997. Acidification of forest top soils in 60 years to the southwest of Helsinki. Forest Ecology and Management 94(1–3): 187–193.

Alexander, E.B. & Burt, R. 1996. Soil development on moraines of Mendellhall Glacier, southern Alaska. 1. The moraines and soil morphology. Geoderma 72: 1–17.

Alriksson, A. & Olsson, M.T. 1995. Soil changes in different age classes of Norway spruce (Picea abies (L.) Karst.) on afforested farmland. Plant and Soil 168–169: 103–110.

Anttila, P. 1991. The acidifying load in Finland. In:

Kenttämies, K. (ed.). Acidification research in Finland. Review of the results of the Finnish Acidification Research Programme (HAPRO) 1985–1990. Ministry of Environment, Environmental Protection Department, Brochure 39, Helsinki. p. 8–14.

Autry, A.R., Fitzgerald, J.W. & Caldwell, P.R. 1990. Sulfur fractions and retention mechanisms in forest soils. Canadian Journal of Forest Research 20(3): 337–342.

Berdén, M., Nilsson, S.I., Rosén, K. & Tyler, G. 1987. Soil acidification: extent, causes and consequences. An evaluation of literature information and current research. National Swedish Environmental Board Report 3292. 164 p.

Bergholm, J., Berggren, D. & Alavi, G. 2003. Soil acidification induced by ammonium sulphate addition in a Norway spruce forest in Southwest Sweden. Water, Air, and Soil Pollution 148(1–4): 87–109.

Bhatti, J.S., Fleming, R.L., Foster, N.W., Meng, F.R., Bourque, C.P.A. & Arp, P.A. 2000. Simulations of pre- and post-harvest soil temperature, soil moisture, and snowpack for jack pine: comparison with field observations. Forest Ecology and Management 138(1–3): 413–426.

Billett, M.F., Fitzpatrick, E.A. & Cresser, M.S. 1988. Long-term changes in the acidity of forest soils in North-East Scotland. Soil Use and Management 4(3): 102–107.

— , Parker-Jervis, E.A., Fitzpatrick, E.A. & Cresser, M.S. 1990. Forest soil chemical changes between 1949/50 and 1987. Journal of Soil Science 41(1): 133–145.

Binkley, D. & Valentine, D. 1991. Fifty-year biogeochemical effects of green ash, white pine, and Norway spruce in a replicated experiment. Forest Ecology Management 40(1–2): 13–25.

— & Högberg, P. 1997. Does atmospheric deposition of nitrogen threaten Swedish forests? Forest Ecology and Management 92(1–3): 119–152.

Brais, S., Camiré, C., Bergeron, Y. & Paré, D. 1995. Changes in nutrient availability and forest floor characteristics in relation to stand age and forest composition in the southern part of the boreal forest of northwestern Quebec. Forest Ecology and Management 76(1–3): 181–189.

Brand, D.G., Kekoe, P. & Connors, M. 1986. Coniferous afforestation leads to soil acidification in Central Ontario. Canadian Journal of Forest Research 16(6): 1389–1391.

Buberl, H.G., von Wilbert, K., Trefz-Malcher, G. &

Hildebrand, E.E. 1994. Der chemische Zustand von Waldböden in Baden-Württemberg. Ergebnisse der Bodenzustandserhebung im Wald 1989–92 (BZE). Mitteilungen der Forstlichen Versuchs- und Forschungsanstalt Baden-Württemberg 182: 104.

Derome, J., Kukkola, M. & Mälkönen, E. 1986. Forest liming on mineral soils. Results of Finnish experiments. National Swedish Environmental Protection Board Report 3084. 105 p.

De Vries, W., Reinds, G.J., van der Salm, C., Draaijers, G.P.J., Bleeker, A., Erisman, J.W., Auée, J., Gundersen, P., Kristensen, H.L., van Doppen, H., de Zwart, D., Derome, J., Voogd, J.C.H. & Vel, E.M. 2001. Intensive monitoring of forest ecosystems in Europe. Series Intensive monitoring of forest ecosystems in Europe. UN-ECE & EU, Brussels, Geneva. 177 p.

Drohan, J.R. & Sharpe, W.E. 1997. Long-term changes in forest soil acidity in Pennsylvania, U.S.A. Water, Air, and Soil Pollution 95(1–4): 299–311.

Elonen, P. 1971. Particle-size analysis of soil. Acta Agralia Fennica 122. 122 p.

Falkengren-Grerup, U. 1987. Long-term changes in pH of forest soils in southern Sweden. Environmental Pollution 43(2): 79–90.

— & Eriksson, H. 1990. Changes in soil, vegetation and forest yield between 1947 and 1988 in beech and oak sites of southern Sweden. Forest Ecology and Management 38(1–2): 37–53.

— & Tyler, G. 1991. Changes of cation pools of the topsoil in south Swedish beech forests between 1979 and 1989. Scandinavian Journal of Forest Research 6(2): 145–152.

— & Tyler, G. 1992. Changes since 1950 of mineral pools in the upper C-horizon of Swedish deciduous forest soils. Water, Air, and Soil Pollution 64(3–4): 495–501.

Farmer, A.M. 1995. Soil chemistry change in a lowland English deciduous woodland. Water, Air, and Soil Pollution 85(2): 677–682.

Franzmeier, D.P. & Whiteside, E.P. 1963. A chronosequence of podzols in northern Michigan. II. Physical and chemical properties. Michigan State University, Agricultural Experiment Station, Quarterly Bulletin 46(2): 21–36.

Gobran, G.R., Clegg, S., Courchesne, F. & van Breemen, N. 1998. Rhizospheric processes influencing the biogeochemistry of forest ecosystems. Biogeochemistry 42(1–2): 107–120.

Griffiths, R.B. & Swanson, A.K. 2001. Forest soil

characteristics in a chronosequence of harvested Douglas-fir forests. Canadian Journal of Forest Research 31(11): 1871–1879.

Grimm, R. & Rehfuess, K.E. 1986. Kurzfristige Veränderungen von Bodenreaktion und Kationenaustauscheigenschaften in einem Meliorationsversuch zu Kiefer (Pinus sylv. L) auf Podzol-Pseudogley in der Oberpfalz. Allgemeine Forst- und Jagdzeitung 157(11): 205–213.

Hallbäcken, L. 1992. Long term changes of base cation pools in soil and biomass in a beech and a spruce forest of southern Sweden. Zeitschrift für Pflanzenernährung und Bodenkunde 155(1): 51–60.

— & Tamm, C.-O. 1986. Changes in soil acidity from 1927 to 1982–84 in a forest area of southwest Sweden. Scandinavian Journal of Forest Research 1(2): 219–232.

— & Bergholm, J. 1998. Nutrient dynamics. In: Andersson, F., Braekke, F.H. & Hallbäcken, L. (eds.). Nutrition and growth of Norway spruce forests in a Nordic climatic and deposition gradient. TemaNord. Nordic Council of Ministers, Copenhagen. p. 127–151.

Halonen, O., Tulkki, H. & Derome, J. 1983. Nutrient analysis methods. Metsäntutkimuslaitoksen tiedonantoja 121. 28 p.

Hari, P., Raunemaa, T. & Hautojärvi, A. 1986. The effects on forest growth of air pollution from energy production. Atmospheric Environment 20(1): 129–137.

Helmisaari, H.-S. & Mälkönen, E. 1989. Acidity and nutrient content of throughfall and soil leachate in three Pinus sylvestris stands. Scandinavian Journal of Forest Research 4(1): 13–28.

— & Hallbäcken, L. 1997. Tree biomass belowground. In: Andersson, F., Braekke, F.H. & Hallbäcken, L. (eds.). Nutrition and growth of Norway spruce forests in a Nordic climatic and deposition gradient. TemaNord 1998: 566. Nordic Council of Ministers, Copenhagen. p. 80–90.

—, Makkonen, K., Kellomäki, S., Valtonen, E. & Mälkönen, E. 2002. Below- and above-ground biomass, production and nitrogen use in Scots pine stands in eastern Finland. Forest Ecology and Management 165(1–3): 317–326.

Ingerslev, M. & Hallbäcken, L. 1999. Above ground biomass and nutrient distribution in a limed and fertilized Norway spruce (Picea abies) plantation. Part II. Accumulation of biomass and nutrients. Forest Ecology and Management 119(1–3): 21–38.

Jauhiainen, E. 1969. On the soils in the boreal coniferous region Central Finland–Lapland–Northern Poland. Fennia 98(5).123 p.

— 1973. Age and degree of podzolization of sand soils on the coastal plain of northwestern Finland. Societas Scientiarum Fennica Commentationes Biologicae 68. 32 p.

Johansson, M. & Savolainen, I. 1990. Regional acidification model for forest soils. In: Kauppi, P., Kenttämies, K. & Anttila, P. (eds.). Acidification in Finland. Springer-Verlag, Berlin, Heidelberg. p. 253–269.

Kauppi, P., Anttila, P., Karjalainen-Balk, L., Kenttämies, K., Kämäri, J. & Savolainen, I. 1990. Happamoituminen Suomessa. HAPROn loppuraportti [Acidification in Finland. The final report of the HAPRO project]. Ministry of Environment, Environmental Protection Department, Series A 89. 89 p.

Knoepp, J.D. & Swank, W.T. 1994. Long-term soil chemistry changes in aggrading forest ecosystems. Soil Science Society of America Journal 58(2): 325–331.

Koivisto, P. 1970. Regionality of forest growth in Finland. Communicationes Instituti Forestalis Fenniae 71(2). 76 p.

Krause, H. 1998. Forest floor mass and nutrients in two chronosequences of plantations: Jack pine vs. black spruce. Canadian Journal of Soil Science 78(1): 77–83.

Kukkola, M. & Mälkönen, E. 1997. The role of logging residues in site productivity after first thinning of Scots pine and Norway spruce stands. Finnish Forest Research Institute, Research Papers 640: 230–237.

Mälkönen, E. 1974. Annual primary production and nutrient cycle in some Scots pine stands. Selostus: Vuotuinen primäärituotos ja ravinteiden kiertokulku männikössä. Communicationes Instituti Forestalis Fenniae 84(5). 87 p.

— 1975. Hakkuutähteiden talteenoton seurannaisvaikutuksia [Effects due to slash harvesting]. Teho 1975(10): 12–13.

— 1977. Annual primary production and nutrient cycle in birch stand. Communicationes Instituti Forestalis Fenniae 91(5). 35 p.

—, Derome, J. & Kukkola, M. 1990. Effects of nitrogen inputs on forest ecosystems. Estimation based on long-term fertilization experiments. In: Kauppi, P., Anttila, P. & Kenttämies, K. (eds.). Acidification

in Finland. Springer-Verlag, Berlin, Heidelberg. p. 325–349.

Nihlgård, B. 1971. Pedological influence of spruce planted on former beech forest soils in Scania, South Sweden. Oikos 22: 71–80.

Nordlund, G. 2000. Emissions, air quality and acidifying deposition. In: Mälkönen, E. (ed.). Forest condition in a changing environment – The Finnish case. Kluwer Academic Publishers. p. 49–59.

Nykvist, N. & Rosén, K. 1985. Effect of clear-felling and slash removal on the acidity of northern coniferous soils. Forest Ecology and Management 11(3): 157–169.

Ogner, G., Randem, G., Remedios, G. & Wikstrøm, T. 2001. Increase of soil acidity and concentrations of extractable elements by 1 M ammonium nitrate after storage of dry soil for up to 5 years at 22 °C. Communications in Soil Science and Plant Analysis 32(5–6): 675–684.

Olsson, B.A., Bengtsson, J. & Lundkvist, H. 1996. Effects of different forest harvest intensities on the pools of exchangeable cations in coniferous forest soils. Forest Ecology and Management 84(1–3): 135–147.

Päivänen, J. 1973. Harvennuksen vaikutus lumi- ja routasuhteisiin nuoressa turvemaan männikössä. Abstract: The effect of thinning on the snow cover and soil frost conditions in a young Scots pine stand on drained peat. Silva Fennica 7(2): 114–128.

Payne, G. & Rechcigl, J.E. 1989. Influence of various drying technique on the extractability of plant nutrients from selected Florida soils. Soil Science 148(4): 275–283.

Piirainen, S. 2002. Nutrient fluxes through a boreal coniferous forest and the effects of clear-cutting. The Finnish Forest Research Institute, Research Papers 859. 140 p.

Rhoades, C. & Binkley, D. 1996. Factors influencing decline in soil pH in Hawaiian Eucalyptus and Albizia plantations. Forest Ecology and Management 80(1–3): 47–56.

Roberge, M.R. 1987. Evolution of the pH of various forest soils over the past 20 years. Canadian Forestry Service Information Report – Laurentian Forestry Centre LAU-X-77. 18 p.

Rustad, L.E., Fernandez, I.J., David, M.B., Mitchell, M.J., Nadelhoffer, K.J. & Fuller, R.B. 1996. Experimental soil acidification and recovery at the Bear Brook watershed in Maine. Soil Science Society of America Journal 60(6): 1933–1943.

Saarinen, J. 1989. Effect of drying temperature on the extractable macro- and micronutrients and pH of different peat types. Suo 40(4): 149–153.

Savolainen, I. & Tähtinen, M. 1990. Sulphur dioxide and nitrogen oxide emission scenarios for Finland. In: Kauppi, P., Anttila, P. & Kenttämies, K. (eds.). Acidification in Finland. Springer-Verlag, Berlin, Heidelberg. p. 3–19.

Singh, B.R. 1980. Distribution of total and extractable S and adsorbed $^{35}SO_4^{2-}$ in some acid forest soil profiles of southern Norway. Acta Agriculturae Scandinavica 30: 357–363.

Sjöström, J. & Qvarfort, U. 1992. Long-term changes of soil chemistry in central Sweden. Soil Science 154(6): 450–457.

Skyllberg, U. 1990. Correlation between pH and depth in the mor layer of a Picea abies (L.) Karst. stand on till soils in northern Sweden. Scandinavian Journal of Forest Research 5(2): 143–153.

Staaf, H. & Olsson, B.A. 1991. Acidity in four coniferous forest soils after different harvesting regimes of logging slash. Scandinavian Journal of Forest Research 6(1): 19–29.

Starr, M.R. 1991. Soil formation and fertility along a 5000 year chronosequence. In: Proceedings of the First Symposium on Environmental Geochemistry in Northern Europe, Rovaniemi, Finland, 17–19 October, 1989. Geological Survey of Finland, Special Paper 9: 99–104.

— & Tamminen, P. 1992. Suomen metsämaiden happamoituminen. Summary: Forest soil acidification in Finland. Ympäristötieteelliset kartat ja kartoitushankkeet Suomessa. Environmental maps and environmental surveying projects in Finland. Espoo, 9.–19.10.1991. Geologian tutkimuskeskus, Tutkimusraportti. Geological Survey of Finland, Report of Investigation. 115: 7–14.

Stuanes, A.O., Van Miegroet, H., Cole, D.W. & Abrahamsen, G. 1992. Recovery from acidification. In: Johnson, D.W. & Lindberg, S.E. (eds.). Atmospheric Deposition and Forest Nutrient Cycling. Springer-Verlag, New York. p. 467–494.

Tamm, C.-O. & Hallbäcken, L. 1988. Changes in soil acidity in two forest areas with different acid deposition: 1920s to 1980s. Ambio 17(1): 56–61.

Tamminen, P. 1991. Kangasmaan ravinnetunnusten ilmaiseminen ja viljavuuden alueellinen vaihtelu. Summary: Expression of soil nutrient status and regional variation in soil fertility of forested sites in southern Finland. Folia Forestalia 777. 40 p.

— 2000. Soil factors. In: Mälkönen, E. (ed.). Forest condition in a changing environment – The Finnish case. Forestry Sciences. Kluwer Academic Publishers, Dordrecht. p. 72–86.

— & Starr, M. 1990. A survey of forest soil properties related to soil acidification in southern Finland. In: Kauppi, P., Anttila, P. & Kenttämies, K. (eds.). Acidification in Finland. Springer-Verlag, Berlin, Heidelberg. p. 235–251.

Troedsson, T. 1985. Sensitivity of Swedish forest soils to acidification related to site characteristics. National Swedish Environment Protection Board Report 3001. 51 p.

Tuovinen, J.-P., Kangas, L. & Nordlund, G. 1990. Model calculations of sulphur and nitrogen deposition in Finland. In: Kauppi, P., Anttila, P. & Kenttämies, K. (eds.). Acidification in Finland. Springer-Verlag. p. 167–197.

Tyler, G., Berggren, D., Bergkvist, B., Falkengren-Grerup, U., Folkeson, L. & Rühling, Å. 1985. Soil acidification and metal mobility in forests of southern Sweden. Effects of air pollution, especially acidific deposition, on forests, agriculture and wetlands. The NATO Advanced Research Institute workshop 12–17 May 1985, Toronto, Canada.

Ukonmaanaho, L., Starr, M. & Ruoho-Airola, T. 1998. Trends in sulfate, base cations and H$^+$ concentrations in bulk precipitation and throughfall at integrated monitoring sites in Finland 1989–1995. Water, Air, and Soil Pollution 105(1–2): 353–363.

Ulrich, B., Mayer, R. & Khanna, P.K. 1979. Deposition von Luftverunreinigungen und ihre Auswirkungen in Waldökosystemen im Solling. Schriften aus der Forstlichen Fakultät der Universität Göttingen und der Niedersächsischen Forstlichen Versuchsanstalt 58. 291 p.

van Erp, P.J., Houba, V.J.G. & van Beusichem, M.L. 2001. Effect of drying temperature on amount of nutrient elements extracted with 0.01 M CaCl$_2$ soil extraction procedure. Communications in Soil Science and Plant Analysis 32(1–2): 33–48.

Vannier, C., Didon-Lescot, J.-F., Lelong, F. & Guillet, B. 1993. Distribution of sulphur forms in soils from beech and spruce forests of Mont Lozere (France). Plant and Soil 154: 197–209.

Viro, P. 1952. Kivisyyden määrittämisestä. Summary: On the determination of stoniness. Communicationes Instituti Forestalis Fenniae 40(3). 23 p.

Vuokila, Y. & Väliaho, H. 1980. Viljeltyjen havumetsiköiden kasvatusmallit. Summary: Growth and yield models for conifer cultures in Finland. Communicationes Instituti Forestalis Fenniae 99(2). 271 p.

Yli-Vakkuri, P. 1960. Metsiköiden routa- ja lumisuhteista. Summary: Snow and frozen soil conditions in the forest. Acta Forestalia Fennica 71(5). 48 p.

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Appendix 1. Stand and soil characteristics of the experiments and soil sampling years by layer.

Plot	Spec[a]	Age[b]	Soil type	Org. layer[c] mm	Stones %	<0.06 mm %	Organic layer 1	2	3	4	5	6	0–30 cm 1	2	0–10 cm 1	2	3	4
2401	P	40	HaplPodz	15	3	4	73	93					73	93				
2507	P	56	HaplPodz	43	42	19	81	82	88	93					78	83	88	93
2602	P	32	CamPodz	45	47	26	78	81	83	87	93				78	83	87	93
2707	P	77	HaplPodz	41	53	4	65	93					65	93				
2903	S	42	CamPodz	47	61	9	65	93					65	93				
3003	S	53	CamPodz	22	42	30	76	93					76	93				
3402	S	29	CamPodz	37	17	34	65	79	81	88	93		65	93	79	88	93	
3508	S	48	CamPodz	51	17	24	79	93							79	93		
3610	S	48	CamPodz	43	14	25	79	93							79	93		
3701	P	46	HaplPodz	29	3	11	79	93							79	93		
3816	P	46	HaplPodz	28	14	11	79	93							79	93		
4001	P	29	HaplPodz	20	1	4	73	93					73	93				
4407	P	34	HaplPodz	18	6	4	73	93					73	93				
4507	P	34	HaplPodz	13	6	4	73	93					73	93				
4907	S	27	CamPodz	33	17	43	65	78	93				65	93	78	93		
5005	P	24	CamPodz	27	3	62	65	93					65	93				
5208	P	28	HaplPodz	26	0	15	78	93							78	83	88	93
5305	P	31	HaplPodz	46	29	14	78	93							78	83	88	93
5505	P	20	CamPodz	31	3	48	65	78	93				65	93	78	93		
5608	P	47	HaplPodz	28	76	19	79	93							79	83	88	93
5701	P	49	HaplPodz	27	24	38	79	93							79	83	88	93
5808	S	47	HaplPodz	34	15	38	79	93							79	83	88	93
6007	S	49	HaplPodz	40	26	29	79	93							79	83	88	93
6403	P	46	HaplAreno	25	1	20	72	78	93				72	93	78	93		
6601	S	40	CamPodz	25	12	38	78	93							78	93		
6703	P	43	HaplPodz	31	14	33	78	93							78	93		
6808	P	41	HaplPodz	37	3	12	65	78	80	83	89	93	65	93	78	93		
6901	P	23	CamPodz	25	34	16	78	93							78	93		
7103	P	21	HaplPodz	35	1	9	78	93							78	93		
7306	P	25	HaplPodz	33	17	30	78	93							78	93		
7507	P	26	HaplPodz	27	23	30	79	82	89	93					79	93		
7601	P	26	HaplPodz	17	34	27	79	82	89	93					79	93		
7702	P	26	HaplPodz	32	53	21	79	82	89	93					79	93		
8502	B	44	HaplPodz	26	67	27	78	82	83	86	93				78	93		
11115	S	61	CamPodz	37	6	14	72	78	93				72	93	78	93		
11205	S	46	CamPodz	50	20	21	72	93					72	93				
11317	S	29	CamPodz	33	20	17	78	80	83	87	93				78	83	87	93
11506	P	47	HaplPodz	32	6	22	72	93					72	93				
12612	P	82	HaplPodz	40	53	8	72	93					72	93				
13007	S	56	HaplPodz	43	20	32	72	93					72	93				
13108	P	56	HaplPodz	34	25	48	72	79	82	85	93		72	93	79	85	93	
14602	S	60	HaplPodz	41	6	11	72	93					72	93				
15711	P	28	HaplPodz	23	39	25	74	93					74	93	81	93		
18203	S	75	HaplPodz	47	34	26	73	93					73	93				
22431	P	80	HaplPodz	56	12	8	73	93					73	93				
22437	P	90	HaplPodz	32	56	7	73	93					73	93				
34506	S/B	60	CamPodz	52	39	18	74	93					74	93				
34608	L	32	HaplPodz	45	45	20	74	93					74	93				
34706	L	30	CamPodz	27	56	21	74	93					74	93				
34805	B	27	HaplPodz	34	0	64	74	93					74	93				
35506	P	59	HaplPodz	31	23	21	74	93					74	93				
35510	P	59	HaplPodz	32	20	24	74	93					74	93				
35703	P	49	HaplPodz	33	9	9	79	93					79	93				
35709	P	49	HaplPodz	41	26	17	79	93					79	93				

[a] Tree species: P=pine, S=spruce, B=birch, L=larch
[b] Age at the time of the first soil sampling
[c] Thickness of the organic layer, mm

Appendix 2. Estimated annual average (S_{an}, N_{an}, $kg\,ha^{-1}\,a^{-1}$) and total amount during the monitoring period (S_{tot}, N_{tot}, $kg\,ha^{-1}$) of bulk deposition of SO_4-S and inorganic nitrogen and estimated average volume increment (I_v, $m^3\,ha^{-1}\,a^{-1}$), and stem wood production (Prod, $m^3\,ha^{-1}$) and amount of nutrients retained in stem and branches ($kg\,ha^{-1}$) during the study period.

Plot	S_{an} $kg\,a^{-1}$	S_{tot} kg	N_{an} $kg\,a^{-1}$	N_{tot} kg	I_v $m^3\,a^{-1}$	Prod m^3	N_{ret} kg	Ca_{ret} kg	K_{ret} kg	Mg_{ret} kg	P_{ret} kg
2401	10.7	214	6.4	127	4.1	82	37	38	21	4.8	1.6
2507	7.7	92	4.8	58	8.1	97	44	45	25	5.6	1.9
2602	8.8	132	5.3	80	10.3	155	70	72	40	9.0	3.1
2707	13.1	368	5.4	150	2.7	77	35	36	20	4.5	1.5
2903	10.4	291	4.8	135	8.8	246	136	209	63	18.7	11.8
3003	9.1	155	4.8	82	9.9	168	93	142	43	12.8	8.1
3402	9.1	254	4.3	121	8.9	248	137	210	63	18.8	11.9
3508	6.5	91	3.9	55	7.3	103	57	87	26	7.8	4.9
3610	6.5	91	3.9	55	8.6	121	67	102	31	9.2	5.8
3701	10.8	151	6.3	88	4.8	67	30	31	17	3.9	1.3
3816	10.8	151	6.3	88	5.2	73	33	34	19	4.3	1.5
4001	10.7	214	6.4	127	3.2	64	29	30	17	3.7	1.3
4407	10.7	214	6.4	127	3.7	74	34	34	19	4.3	1.5
4507	10.7	214	6.4	127	3.7	74	34	34	19	4.3	1.5
4907	10.8	301	5.4	151	10.7	300	166	254	76	22.8	14.4
5005	10.8	301	5.4	151	9.4	263	120	122	68	15.3	5.3
5208	8.5	127	4.5	67	10.7	161	73	75	42	9.3	3.2
5305	9.3	140	4.1	62	10.3	155	70	72	40	9.0	3.1
5505	11.3	315	5.5	154	10.7	300	137	139	78	17.4	6.0
5608	6.1	85	3.8	53	5.2	73	33	34	19	4.3	1.5
5701	6.1	85	3.8	53	5.2	73	33	34	19	4.3	1.5
5808	6.1	85	3.8	53	8.6	121	67	102	31	9.2	5.8
6007	6.1	85	3.8	53	9.0	126	70	107	32	9.6	6.0
6403	11.2	236	5.6	118	5.0	104	47	48	27	6.0	2.1
6601	9.9	149	5.9	88	11.1	166	92	141	42	12.6	8.0
6703	9.9	149	5.9	88	4.7	70	32	33	18	4.1	1.4
6808	8.2	230	4.1	115	5.0	139	63	64	36	8.0	2.8
6901	6.5	98	4.0	59	9.4	141	64	66	37	8.2	2.8
7103	6.5	97	4.0	60	3.7	56	25	26	14	3.2	1.1
7306	6.2	93	3.6	54	10.3	155	70	72	40	9.0	3.1
7507	4.9	54	3.5	39	10.7	118	54	55	31	6.8	2.4
7601	4.9	54	3.5	39	10.3	113	52	53	29	6.6	2.3
7702	4.9	54	3.5	39	5.0	55	25	26	14	3.2	1.1
8502	5.5	83	5.3	80	5.0	75	41	64	19	5.7	3.6
11115	12.2	256	5.3	110	9.0	189	105	160	48	14.4	9.1
11205	12.2	256	5.3	110	9.8	205	113	174	52	15.6	9.8
11317	10.6	159	5.3	79	10.8	162	89	137	41	12.3	7.8
11506	12.2	256	5.3	110	7.5	158	72	73	41	9.1	3.2
12612	11.0	230	4.1	87	3.0	63	29	29	16	3.7	1.3
13007	8.5	178	3.8	80	8.4	176	98	149	45	13.4	8.5
13108	8.5	178	3.8	80	4.9	103	47	48	27	6.0	2.1
14602	15.1	318	6.1	127	4.1	86	39	40	22	5.0	1.7
15711	11.7	223	5.3	100	8.1	154	70	72	40	8.9	3.1
18203	9.4	187	4.7	93	6.4	128	71	108	33	9.7	6.1
22431	9.8	195	4.8	97	3.0	60	27	28	16	3.5	1.2
22437	9.8	195	4.8	97	2.9	58	26	27	15	3.4	1.2
34506	15.8	300	6.2	117	7.8	148	82	126	38	11.3	7.1
34608	16.9	321	7.1	135	8.1	154	85	130	39	11.7	7.4
34706	11.9	227	5.2	99	10.7	203	112	172	52	15.5	9.8
34805	9.5	180	4.5	85	8.1	154	85	130	39	11.7	7.4
35506	9.5	181	4.8	92	4.7	89	41	42	23	5.2	1.8
35510	9.5	181	4.8	92	4.7	89	41	42	23	5.2	1.8
35703	7.6	106	4.5	63	6.7	94	43	44	24	5.4	1.9
35709	7.6	106	4.5	63	6.7	94	43	44	24	5.4	1.9