

# Comparison of throughfall and soil solution chemistry between a high-density Corsican pine stand and a naturally regenerated silver birch stand

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**“Capsule”:** Nitrogen throughfall deposition in a naturally regenerated birch stand was 3.5 times lower than in a neighbouring Corsican pine plantation.

## Abstract

In Flanders, critical loads for acidification and eutrophication are exceeded in the majority of the forest stands, and many previously nitrogen limited forest ecosystems have become nitrogen saturated. The present study investigates whether a naturally regenerated stand of silver birch (*Betula pendula* Roth) contributes less to the acidification and eutrophication of the forest soil than a high-density plantation of Corsican pine (*Pinus nigra* ssp. *laricio* Maire). Throughfall deposition of inorganic nitrogen was about 3.5 times higher in the Corsican pine stand than in the birch stand. Potassium throughfall deposition was significantly higher under birch due to higher canopy leaching. Magnesium throughfall deposition was significantly higher under the pine canopy due to higher dry deposition. The lower nitrogen throughfall deposition in the birch stand was reflected in a 60% lower nitrate percolation at 1 m depth compared with pine. Nitrate soil percolation is linked to losses of aluminium and base cations.

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## 1. Introduction

With a forest cover of less than 10%, the northern part of Belgium (Flanders) has one of the lowest afforestation rates of Europe (Luysaert et al., 1999). About 30% of the Flemish forest consists of first or second generation Scots pine (*Pinus sylvestris* L.) and Corsican pine (*Pinus nigra* ssp. *laricio* Maire) stands, located on sandy spodosols formerly covered by heath. Most of these forests are homogeneous plantations on aeolian

soils with low buffering capacity, and have shown to be susceptible to infestations (like *Sphaeropsis sapinea* (Fr.) Dyko & Sutton). The condition of these forests has become even more critical due to increasing stress from high atmospheric deposition levels. In 2001, the average acid deposition in Flanders amounted to 4605 mol ha<sup>-1</sup> y<sup>-1</sup>, of which 1183 mol ha<sup>-1</sup> y<sup>-1</sup> came from SO<sub>x</sub>, 1287 mol ha<sup>-1</sup> y<sup>-1</sup> from NO<sub>y</sub>, and 2135 mol ha<sup>-1</sup> y<sup>-1</sup> from NH<sub>x</sub> (NH<sub>4</sub><sup>+</sup>-deposition was converted to deposition of H<sup>+</sup> assuming complete nitrification in the soil) (Van Avermaet et al., 2002). Although the total acid deposition has shown a decreasing trend over the past decade (5928 mol ha<sup>-1</sup> y<sup>-1</sup> in 1990), it far exceeds all medium and long-term targets as formulated in the Protocol of Göteborg and the European NEC directive.

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Due to the implementation of air pollution agreements, sulphur deposition in particular has decreased over the past decade (–22% compared to 1990) (Van Avermaet et al., 2002). The high exogenous input of nitrogen and sulphur is due to (i) high concentrations of atmospheric pollutants (VMM, 2001), (ii) high humidity, which increases the collection efficiency for airborne material (Ruijgrok et al., 1997) and (iii) a high degree of forest fragmentation leading to a reinforcement of edge deposition effects (Draaijers et al., 1988). As a result, the deposition of nitrogen and sulphur in Flanders exceeds the critical loads for acidification and eutrophication in the majority of forest stands (Craenen et al., 2000).

The high deposition of nitrogen has provoked N saturation of many previously N-limited temperate forest ecosystems (Nihlgard, 1985; Aber et al., 1989). When a forest becomes N saturated, the capacity of plants and soil to accumulate N is exceeded (Aber et al., 1989) and nitrate starts to leach and impair groundwater and surface water (Nihlgard, 1985; Agren and Bosatta, 1988). The presence of inorganic nitrogen below the rooting zone is therefore a key index to distinguish N-limited and N-saturated forest ecosystems (Aber et al., 1989; Gundersen et al., 1998). Nitrification of ammonium derived from atmospheric deposition does not only cause nitrate leaching, but may also be harmful to the ecosystem itself. The effects comprise acidification with concomitant loss of nutrient cations (calcium, magnesium and potassium) and/or mobilisation of aluminium (Ulrich, 1987; Cole et al., 1992).

Forest structure, canopy density and tree species composition were reported to have a significant impact on dry deposition. Dry deposition is increasing with increasing tree height (Lovett and Reiners, 1986) and increasing leaf area index (Ivens, 1990). Dry deposition is furthermore dependent on tree species composition, as conifers are found to be more efficient in collecting particles and cloud droplets compared to broadleaf trees (Brown and Iles, 1991, Draaijers, 1988). Also nutrient exchange processes in the canopy (Alcock and Morton, 1985), uptake capacity by roots (Cole and Rapp, 1981; Magill et al., 1997), and litter decomposition rate (Howard and Howard, 1990; Johansson, 1995) were reported to depend on forest structure and composition.

This study's hypothesis is that, given the high atmospheric nitrogen and acid deposition in Flanders, naturally regenerated forests of broadleaf trees contribute less to soil acidification and eutrophication than homogeneous plantations of coniferous trees. Forest conversion from these high-density plantations to deciduous stands is therefore expected to have an impact on the forest soil by decreasing nitrate percolation, loss of base cations and mobilisation of toxic aluminium. This hypothesis is tested by comparing biogeochemical fluxes through (i) atmospheric deposition, (ii) litterfall, and (iii) soil leaching, between a high-density plantation of Corsican pine and a naturally regenerated stand of silver birch.

## 2. Materials and methods

### 2.1. Site description

The experimental site is located in the 'Hoogmoerheide' nature reserve in Merksplas (51°15' N, 4°54' E), in the northeastern part of Belgium (Flanders). Hoogmoerheide has a surface area of 105 ha and is mainly dominated by heath, homogeneous plantations of Corsican pine (*P. nigra* ssp. *laricio* Maire), homogeneous plantations of Scots pine (*P. sylvestris* L.), and naturally developed stands of silver birch (*Betula pendula* Roth). Neighbouring stands of Corsican pine and silver birch were selected. Both stands had the same age and were growing on the same parent material. The management history of the stands was also very similar. Following the FAO classification (FAO, 1988), the coarse sandy soils were classified as Haplic podzols.

Exchangeable cations were measured using flame atomic absorption spectrophotometry after extracting the soil during 30 min in an NH<sub>4</sub>–EDTA-solution (1/5). Extraction solution was filtered before measurement through a 589 Blauband filter. Although the soil characteristics of both stands are similar, exchangeable aluminium of the upper 50 cm is much higher in the Corsican pine stand (Table 1). In deeper soil, exchangeable aluminium is highest in the silver birch stand. The history of the stands was reconstructed using historical topographic maps. In 1850, the vegetation was transformed

Table 1  
Soil characteristics of the silver birch and Corsican pine stands at Merksplas, 1999

Soil depth (m)	pH-H <sub>2</sub> O		C/N-ratio		Exchangeable base cations (meq/100 g)		Exchangeable Al (meq/100 g)	
	Birch	Pine	Birch	Pine	Birch	Pine	Birch	Pine
0–0.1	3.7	3.6	30.0	31.2	0.10	0.09	0.18	0.47
0.1–0.3	3.9	4.2	28.0	26.7	0.06	0.05	0.67	0.95
0.5–1	4.1	4.3	14.0	15.0	0.04	0.04	1.29	0.47

from heath to Scots pine. Scots pine was harvested and the stand was replanted with Corsican pine around 1955. Between 1955 and 1985, the birch stand developed naturally as the sublayer of a Scots pine stand. In 1985, the Scots pines were cut and the birches became the dominant tree species.

Both stands are about 40 years old. In 1999, average tree height, basal area and stem volume were 14.4 m,  $16 \text{ m}^2 \text{ ha}^{-1}$  and  $111 \text{ m}^3 \text{ ha}^{-1}$  for the silver birch stand, and 16.1 m,  $45 \text{ m}^2 \text{ ha}^{-1}$  and  $365 \text{ m}^3 \text{ ha}^{-1}$  for the Corsican pine stand, respectively. These stand characteristics are representative for Flanders and the Netherlands (Jansen et al., 1996). The leaf area index (LAI) of the pine stand was visually estimated to be three times higher than the LAI of the birch stand.

## 2.2. Experimental set-up

The experiment was set up as three replicated plots under Corsican pine and silver birch. To avoid a forest edge effect on the deposition values (Draaijers et al., 1988; De Schrijver et al., 1998), all plots were established at a fixed distance of 50 m from the forest edge. Each plot was equipped with four throughfall collectors, suction lysimeters with ceramic cup at three depths (0.1 m, 0.5 m and 1 m), and five circular litter traps of  $0.3 \text{ m}^2$ . The applied suction on the porous cup lysimeters was  $-50 \text{ kPa}$ . Bulk deposition was collected using four bulk collectors placed above the heath adjacent to the forest. Throughfall and bulk precipitation were collected using polyethylene funnels (15 cm diameter) supported by and draining into 2-l polyethylene bottles. The bottles were placed below ground level to avoid the growth of algae and to keep the samples cool. A nylon wire mesh was placed in the funnels to prevent contamination by large particles. Stemflow water was not collected because of its low contribution to nutrient fluxes in young pine and birch stands.

Water fractions were collected and measured fortnightly from September 1998 to April 1999 and monthly from May 1999 to February 2000. On each sampling occasion, the water volume in every collector was measured in the field, and the bottles were replaced by bottles rinsed with distilled water. The four throughfall samples of each plot were pooled to one sample for the chemical analyses. All water samples were transported and stored at a maximum temperature of  $5 \text{ }^\circ\text{C}$ . After the samples had been analysed for pH (ion-specific electrode), they were filtered through a glass microfibre filter (WHATMAN GF/A) and a nylon membrane filter (GELMAN, nyloflo) of  $0.45 \text{ }\mu\text{m}$ . Samples were analysed within a week for  $\text{Cl}^-$  (ion-specific electrode),  $\text{NO}_3^-$  (UV photometric method at  $\lambda = 210 \text{ nm}$  according to the Dutch standard method NEN 6581),  $\text{NH}_4^+$  (photometric determination of a reaction product of  $\text{NH}_4^+$  at

$\lambda = 660 \text{ nm}$  according to the Dutch standard method NEN 6576), and  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$  and  $\text{Al(III)}$  (flame atomic absorption spectrophotometry).

Litterfall was collected fortnightly between September 1998 and January 1999, and dry litterfall weight was determined after drying for 48 h at  $70 \text{ }^\circ\text{C}$ . Litterfall samples were analysed on total N (modified method of Kjeldahl) and  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{Na}^+$  (flame atomic absorption spectrophotometry) after oxidation with a  $\text{HNO}_3$  digestion.

## 2.3. Quality control

The quality of the chemical analyses was checked by including method blanks, repeated measurements of internal and certified reference samples, and by inter-laboratory tests. The methods were validated with samples from inter-laboratory profession tests and standard reference materials (CRM 100, CRM 409). For water samples the relative standard deviation ( $\text{Stdev/average} \times 100$ ) based on repeated measurements of internal quality controls was 4.6% for  $\text{NH}_4^+$ , 3.3% for  $\text{NO}_3^-$ , 5.1% for  $\text{Na}^+$ , 4.3% for  $\text{K}^+$ , 4.1% for  $\text{Ca}^{2+}$ , 4.3% for  $\text{Mg}^{2+}$ , and 4.7% for  $\text{Al(III)}$ . For plant samples the relative standard deviation based on repeated measurements of CRM 100 was 0.8% for N, 4.4% for K, 3.5% for Ca and 4.6% for Mg. The rsd of the analytical methods is a lot smaller than the rsd of the observed nutrient concentrations, so the analytical methods can only be held responsible for a minor part of the total and temporal variability.

## 2.4. Element input fluxes

Litterfall and deposition are both considered as sources of element input to the soil. From the viewpoint of the ecosystem, however, litterfall has to be considered as an internal element cycling.

Element deposition was calculated by multiplying the water volume with the element concentration in that volume. Element input by litterfall was calculated by multiplying the amount of litterfall with the element concentration in the litter.

Compared to bulk precipitation, the chemical composition of throughfall water elements is generally altered, and it is widely acknowledged that this transformation results from (i) the washing off of dry deposition of aerosols and gases, as well as (ii) canopy leaching, i.e. release of ions from plant tissues or (iii) canopy uptake (Parker, 1983). The canopy budget method was used (Ulrich, 1983; De Vries et al., 1998) to estimate the contribution of dry deposition and canopy leaching or uptake to net throughfall. Following Parker (1983), we calculated the deposition quantity of net throughfall

water (NTW, mol ha<sup>-1</sup> y<sup>-1</sup>) to obtain the total effect of the canopy on deposition in the forest:

$$\text{NTW} = \text{TF} - \text{BD} = \text{DD} + \text{CL} \quad (1)$$

where BD = bulk deposition, TF = throughfall, DD = dry deposition, and CL = canopy leaching.

In the canopy budget method, Na<sup>+</sup> is assumed to be inert with respect to the canopy, i.e. neither uptake nor leakage occurs. Furthermore, particles containing K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup> are assumed to have the same deposition velocity as Na<sup>+</sup>, as expressed by a dry deposition factor (DDF):

$$\text{DDF} = \frac{(\text{TF} - \text{BD})_{\text{Na}}}{\text{BD}_{\text{Na}}} \quad (2)$$

Dry deposition of K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup> is then calculated as bulk deposition multiplied by this dry deposition factor:

$$\text{DD}_x = \text{BD}_x \text{DDF} \quad (3)$$

where  $x = \text{K}^+, \text{Ca}^{2+}$  or  $\text{Mg}^{2+}$ .

Canopy leaching of base cations is calculated by subtracting the estimated dry deposition from net throughfall water. Assuming that the leaching of base cations from the canopy equals the uptake of protons and ammonium in a proportion equal to their fluxes in bulk deposition and throughfall (De Vries et al., 1998), the canopy uptake (CU) of H<sup>+</sup> and NH<sub>4</sub><sup>+</sup> is subsequently calculated from the sum of the exchanged cations of K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup>:

$$\text{CU}_{\text{NH}_4} = \frac{(\text{BD} + \text{TF})_{\text{NH}_4}}{(\text{BD} + \text{TF})_{\text{NH}_4} + (\text{BD} + \text{TF})_{\text{H}}} \text{CL}_{\text{K} + \text{Ca} + \text{Mg}} \quad (4)$$

Knowing the canopy uptake of NH<sub>4</sub><sup>+</sup>, the dry deposition flux of NH<sub>4</sub><sup>+</sup> (NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup> aerosol) can be computed from TF + CU - BD. Finally it is assumed that canopy leaching of NO<sub>3</sub><sup>-</sup> is negligible (Neary and Gizyn, 1994), allowing the calculation of dry deposition of NO<sub>3</sub><sup>-</sup>.

### 2.5. Element percolation flux

Element percolation flux was calculated by multiplying the calculated water percolation volume at 1 m depth with the average element concentration in the soil solution at 1 m depth. Since meteorological parameters were not measured in the stands, the percolation water flux at 1 m depth was calculated using the Chloride Mass Balance (CMB) method. This method is based on the assumption of conservation of mass between the input of atmospheric chloride and the chloride flux in the subsurface (Eriksson and Khunakasem, 1969) according to the formula

$$P = \frac{\text{TD}}{\text{Cl}_{\text{ss}}} \quad (5)$$

where  $P$  = percolation flux, TD = total atmospheric chloride deposition and Cl<sub>ss</sub> = average chloride concentration in the soil solution.

This method is based on the fact that Cl<sup>-</sup> is biologically inert, i.e. not preferentially taken up or excluded by roots. As a consequence, water uptake does not result in a change in Cl<sup>-</sup> concentration. Moreover, concentration of Cl<sup>-</sup> is not affected by mineralisation (Thomas and Büttner, 1998). A limitation of the CMB-method is that it requires long-term averages of chloride concentrations in throughfall water and soil solution. Therefore, the obtained results only count for the measuring period (September 1998 to February 2000).

This article only discusses the results of nutrient concentrations in the soil solution at 1 m depth, as concentrations in soil solution at 0.1 and 0.5 m depth did not provide additional information.

### 2.6. Statistical analysis

As water fractions were collected and measured fortnightly from September 1998 to April 1999, but monthly from May 1999 to February 2000, the fortnightly deposition data were summed and the fortnightly data of soil solution concentrations were averaged on a volume-weighted basis.

To compare the element fluxes of both tree species throughout time, a repeated measures test (also called: a within-subjects test) was performed. This is a variant on the Factorial Design (an experiment which tests the effect of one or more factors) in which at least one factor is a repeated measure. In this case, time (within factor) and species (between factor) are two factors determining throughfall depositions and soil solution concentrations. A repeated measures test is more powerful than a One-Way ANOVA test since the procedure accounts for variability in time. This information is used to provide a more precise estimate of the experimental error. The repeated measures test was conducted after testing the preconditions of normality and homoscedasticity (Neter et al., 1996), which were both fulfilled. The major problem with repeated measures designs is the potential for carry-over effects. Therefore, the precondition of absence of autocorrelation was tested (Neter et al., 1996), and found to be satisfactory.

Statistical analyses were performed on monthly data for the growing season (May to November;  $n = 10$ ) and the dormant season (December to April;  $n = 8$ ), as well as for the complete measuring period ( $n = 18$ ). To allow comparison between nutrient inputs via throughfall deposition and litterfall, the annual data of September 1998 to August 1999 are presented.

### 3. Results

#### 3.1. Hydrologic fluxes

During the 18-month measuring period, total bulk precipitation was 1538 mm. Average interception by the canopy represented 17% of the total precipitation for the deciduous stand and 37% for the coniferous stand. This resulted in a significantly different ( $p < 0.001$ ) total throughfall amount of 1250 mm in the silver birch stand and 961 mm in the Corsican pine stand.

Using  $\text{Cl}^-$  as a tracer (data not shown), the calculated quantity of percolation water under both stands was comparable. Percolation output was somewhat higher under the birch stand (541 mm) than under the pine stand (489 mm), which corresponds to the throughfall quantities.

#### 3.2. Ion deposition

The bulk and throughfall deposition of nitrogen consisted mainly of ammonium (Fig. 1, Tables 2 and 3). Throughfall nitrogen deposition was 1.5 times higher than bulk deposition in the birch stand, and five times higher than bulk deposition in the pine stand (Fig. 1, Table 3). Throughfall ammonium and nitrate deposition in the pine stand were significantly higher than in the birch stand (Table 2). Moreover, nitrogen deposition was significantly influenced by time, indicating a strong seasonality in N deposition (Fig. 1). For the total measuring period, a significant interaction was found between tree species and time concerning N deposition.

In both stands, throughfall deposition of ammonium-N mainly resulted from dry deposition (Table 3). According to the canopy budget model,  $471 \text{ mol ha}^{-1} \text{ y}^{-1}$

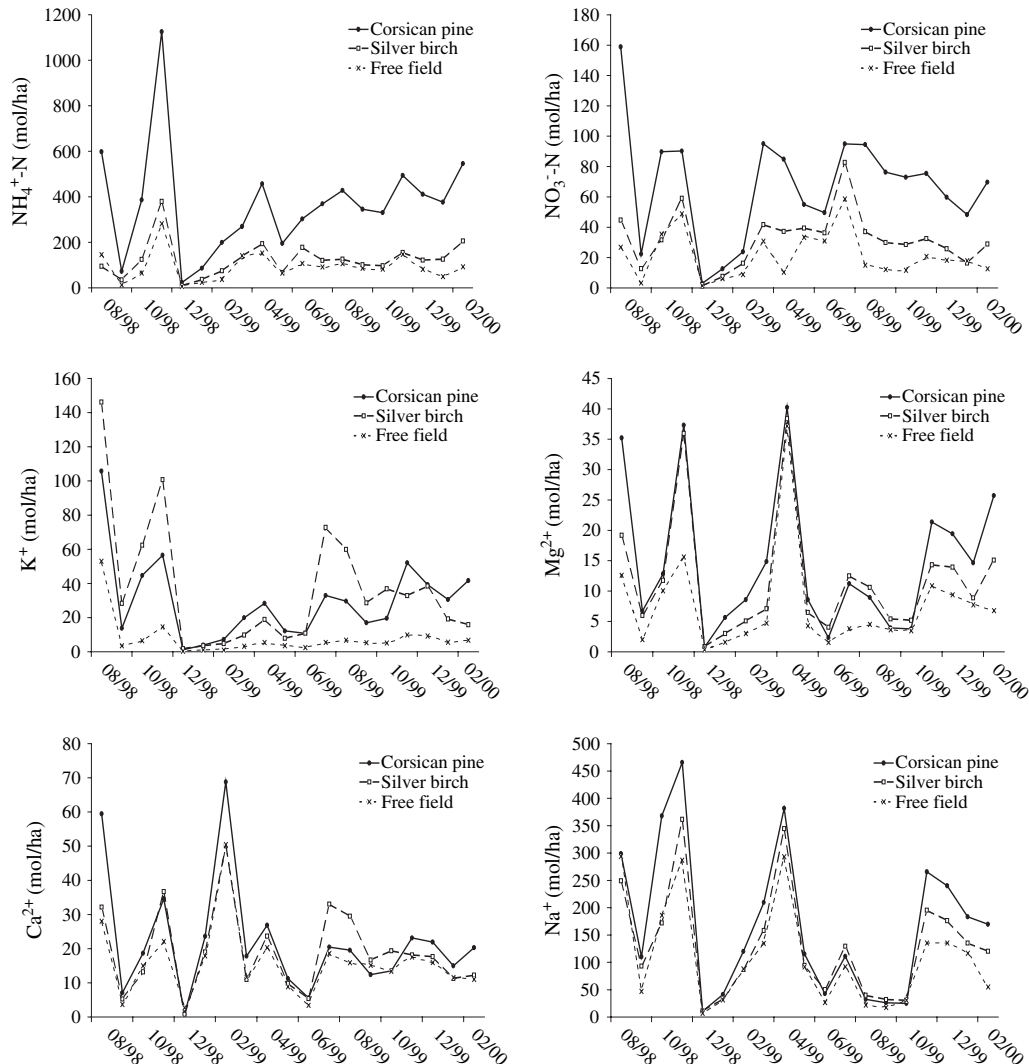


Fig. 1. Bulk and throughfall deposition ( $\text{mol ha}^{-1} \text{ month}^{-1}$ ) of  $\text{NO}_3^-$ -N,  $\text{NH}_4^+$ -N,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$  and  $\text{Na}^+$  in the Corsican pine plantation and the silver birch stand.

Table 2

Average monthly bulk and throughfall depositions ( $\pm$  standard deviation) expressed in mol ha<sup>-1</sup>, during the growing season, the dormant season and the whole year for the measuring period (September 1998–February 2000) (significance levels of species (Sp.), time (Ti.) and interaction between species and time (Int.) are indicated (ANOVA, repeated measures design))

	Growing season			Dormant season			Study period					
	Mean $\pm$ s.d.	Sign.			Mean $\pm$ s.d.	Sign.			Mean $\pm$ s.d.	Sign.		
		Sp.	Ti.	Int.		Sp.	Ti.	Int.		Sp.	Ti.	Int.
<b>Ammonium-N</b>												
Bulk	108 $\pm$ 69				73 $\pm$ 54				93 $\pm$ 64			
Silver birch	135 $\pm$ 90				113 $\pm$ 70				126 $\pm$ 81			
Corsican pine	423 $\pm$ 271	*	***	***	296 $\pm$ 184	**	***	**	369 $\pm$ 241	**	***	***
<b>Nitrate-N</b>												
Bulk	27 $\pm$ 17				13 $\pm$ 9				21 $\pm$ 15			
Silver birch	40 $\pm$ 18				22 $\pm$ 14				32 $\pm$ 18			
Corsican pine	80 $\pm$ 34	**	***	***	50 $\pm$ 34	**	***	*	67 $\pm$ 37	**	***	***
<b>Potassium</b>												
Bulk	11 $\pm$ 14				4 $\pm$ 3				8 $\pm$ 11			
Silver birch	53 $\pm$ 41				14 $\pm$ 12				37 $\pm$ 38			
Corsican pine	36 $\pm$ 28	**	***	**	22 $\pm$ 16	n.s.	***	***	30 $\pm$ 24	n.s.	***	***
<b>Calcium</b>												
Bulk	15 $\pm$ 7				18 $\pm$ 14				16 $\pm$ 11			
Silver birch	20 $\pm$ 11				18 $\pm$ 15				19 $\pm$ 12			
Corsican pine	21 $\pm$ 15	n.s.	***	**	24 $\pm$ 20	n.s.	***	***	22 $\pm$ 17	n.s.	***	***
<b>Magnesium</b>												
Bulk	7 $\pm$ 5				9 $\pm$ 12				8 $\pm$ 8			
Silver birch	12 $\pm$ 9				12 $\pm$ 12				12 $\pm$ 10			
Corsican pine	14 $\pm$ 12	n.s.	***	n.s.	16 $\pm$ 12	*	***	***	15 $\pm$ 12	*	***	**
<b>Sodium</b>												
Bulk	112 $\pm$ 103				108 $\pm$ 89				110 $\pm$ 95			
Silver birch	132 $\pm$ 105				133 $\pm$ 103				132 $\pm$ 101			
Corsican pine	160 $\pm$ 155	n.s.	**	n.s.	170 $\pm$ 117	n.s.	***	n.s.	169 $\pm$ 136	n.s.	***	n.s.
<b>Protons</b>												
Bulk	4 $\pm$ 4				7 $\pm$ 6				5 $\pm$ 5			
Silver birch	2 $\pm$ 2				5 $\pm$ 5				4 $\pm$ 4			
Corsican pine	1 $\pm$ 1	**	***	***	1 $\pm$ 1	***	***	**	0 $\pm$ 1	***	***	***

\* $p < 0.05$ ; \*\* $p < 0.01$ ; \*\*\* $p < 0.001$ .

and 150 mol ha<sup>-1</sup> y<sup>-1</sup> ammonium-N were retained in the canopies of the birch and the pine stands, respectively (Table 3), especially during the growing season (not shown). Nitrate-N mainly entered the birch stand by bulk deposition, and only about 30% originated from dry deposition. In the pine stand on the other hand, throughfall deposition of nitrate-N was mainly the result from dry deposition, and only 29% originated from bulk deposition.

Input of N via litterfall was 1.8 kmol ha<sup>-1</sup> y<sup>-1</sup> in the birch stand and 2.3 kmol ha<sup>-1</sup> y<sup>-1</sup> in the pine stand (Table 3). Adding inorganic nitrogen throughfall deposition and total nitrogen input via litterfall gave 3.8 kmol N ha<sup>-1</sup> y<sup>-1</sup> in the birch stand and 9.2 kmol N ha<sup>-1</sup> y<sup>-1</sup> in the pine stand.

Throughfall deposition of Ca<sup>2+</sup>, Mg<sup>2+</sup> and K<sup>+</sup> was higher than bulk deposition during both growing and dormant seasons (Fig. 1, Table 2). Relative to bulk deposition and taken over a period of a whole year, the Ca<sup>2+</sup>, Mg<sup>2+</sup> and K<sup>+</sup> throughfall depositions were

enriched by a factor 1.2, 1.6 and 5, respectively, under the deciduous canopy, and by 1.4, 1.9 and 3.4, respectively, under the coniferous canopy. Throughfall deposition of K<sup>+</sup> and Mg<sup>2+</sup> was significantly different between the birch and pine stand for the whole measuring period (Table 2). A significant tree species effect was also observed for K<sup>+</sup> during the growing season and for Mg<sup>2+</sup> during the dormant season. Throughfall deposition of all base cations was significantly affected by time (Fig. 1). The interaction between tree species and time was significant for Ca<sup>2+</sup>, Mg<sup>2+</sup> and K<sup>+</sup>.

Throughfall deposition of protons was generally lower than bulk deposition. The repeated measures design revealed a significant effect of tree species and time on throughfall deposition of protons (Table 2) for the whole year. Both forest canopies neutralized significant amounts of protons, especially during the growing season. Birch and pine retained 34% and 82% of the incoming protons, respectively.

Table 3

Bulk deposition, throughfall deposition split up calculated dry deposition and canopy leaching (+)/uptake (-), input of nutrients via litterfall, and total input to the forest floor (throughfall deposition + litterfall) for the period September 1998–August 1999 (all fluxes in mol ha<sup>-2</sup> y<sup>-1</sup>)

	Bulk deposition	Dry deposition	Leaching/uptake	Throughfall deposition	Litterfall	Total input
Birch stand						
Ammonium-N	900	1114	-471	1543	—	—
Nitrate-N	314	136	0	450	—	—
Total nitrogen	1214	1250	-471	1993	1779	3772 <sup>a</sup>
Potassium	118	31	445	594	128	722
Calcium	247	67	-7	307	140	447
Magnesium	107	29	37	173	91	264
Pine stand						
Ammonium-N	900	5057	-150	5807	—	—
Nitrate-N	314	771	00	1085	—	—
Total nitrogen	1214	5828	-150	6892	2279	9171 <sup>a</sup>
Potassium	118	74	212	404	95	499
Calcium	247	152	-57	342	157	499
Magnesium	107	66	29	202	49	251

<sup>a</sup> Throughfall deposition of inorganic nitrogen + total nitrogen in litterfall.

### 3.3. Element output

In contrast to throughfall water, percolation fluxes at 1 m depth were dominated by nitrate and not by ammonium (Table 5). Nitrate concentrations in the soil solution at 1 m depth were significantly influenced by both tree species and time (Fig. 2, Table 4) for the whole year as well as for the dormant and the growing season separately. Compared to the soil solution under birch, nitrate concentrations under pine were on the average three times higher in the growing season and two times higher in the non-growing season. Only time was of significant influence on the low NH<sub>4</sub><sup>+</sup> soil solution concentrations.

Concentrations of K<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup> and Al(III) in the soil solution at 1 m depth were clearly time and species dependent (Table 4). Tree species significantly influenced the soil solution concentration of base cations and aluminium, annually and on a seasonal basis, except for K<sup>+</sup>. On a yearly basis, the interaction between tree species and time was significant for the base cations, but not for Al(III). For the dormant season, K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup> concentrations in the soil solution were significantly affected by tree species, time and the interaction between these two factors. Total element percolation of all base cations and aluminium was higher under Corsican pine than under silver birch (Table 5).

Proton concentrations in the soil solution at 1 m depth were only time dependent. No significant differences between species were found.

## 4. Discussion

### 4.1. Element input

Compared to bulk deposition, a net NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> enrichment in throughfall water was observed for both

stands. In the birch stand, throughfall deposition of inorganic nitrogen was 3.5 times higher in the pine stand. Von Wilpert et al. (2000) found that the deposition load in deciduous beech stands was 45–85% lower than in spruce stands. In the Netherlands, nitrogen deposition was significantly higher in stands of Douglas fir and Scots pine than in oak stands (Van Ek and Draaijers, 1994). Differences in stand and crown characteristics, such as lower stand and crown density, lower LAI, and the summergreen character, are important factors for lower throughfall element fluxes in deciduous forest stands (Alcock and Morton, 1985; Draaijers, 1993; Houle et al., 1999).

It was calculated that ammonium was retained by the canopy of both forest types, but to a greater extent in the birch stand than in the pine stand. The performed calculations are based on the assumption that total canopy uptake of protons and ammonium is equal to total canopy leaching of Ca<sup>2+</sup>, Mg<sup>2+</sup> and K<sup>+</sup> due to cation exchange. The reliability of the calculated ammonium uptake by the canopy depends on (i) the calculated base cation leaching, and (ii) the relationship between canopy leaching of base cations and canopy uptake of ammonium and protons. Reliability of estimated base cation leaching will be discussed later. The calculated ammonium uptake might be overestimated since no corrections were made for passive cation leaching, which may be due to senescence and sprouting or other possible metabolic processes. However, the estimated canopy uptake is in line with literature data: the aboveground uptake of inorganic nitrogen by forests was suggested to be between 150 and 350 mol N ha<sup>-1</sup> y<sup>-1</sup> (Ivens, 1990), and between 200 and 300 up to 850 mol N ha<sup>-1</sup> y<sup>-1</sup> (Lovett, 1992). Both studies were conducted in areas with relatively low air concentrations of N compounds. It is not yet clear if higher deposition results in a higher uptake rate (Bleeker

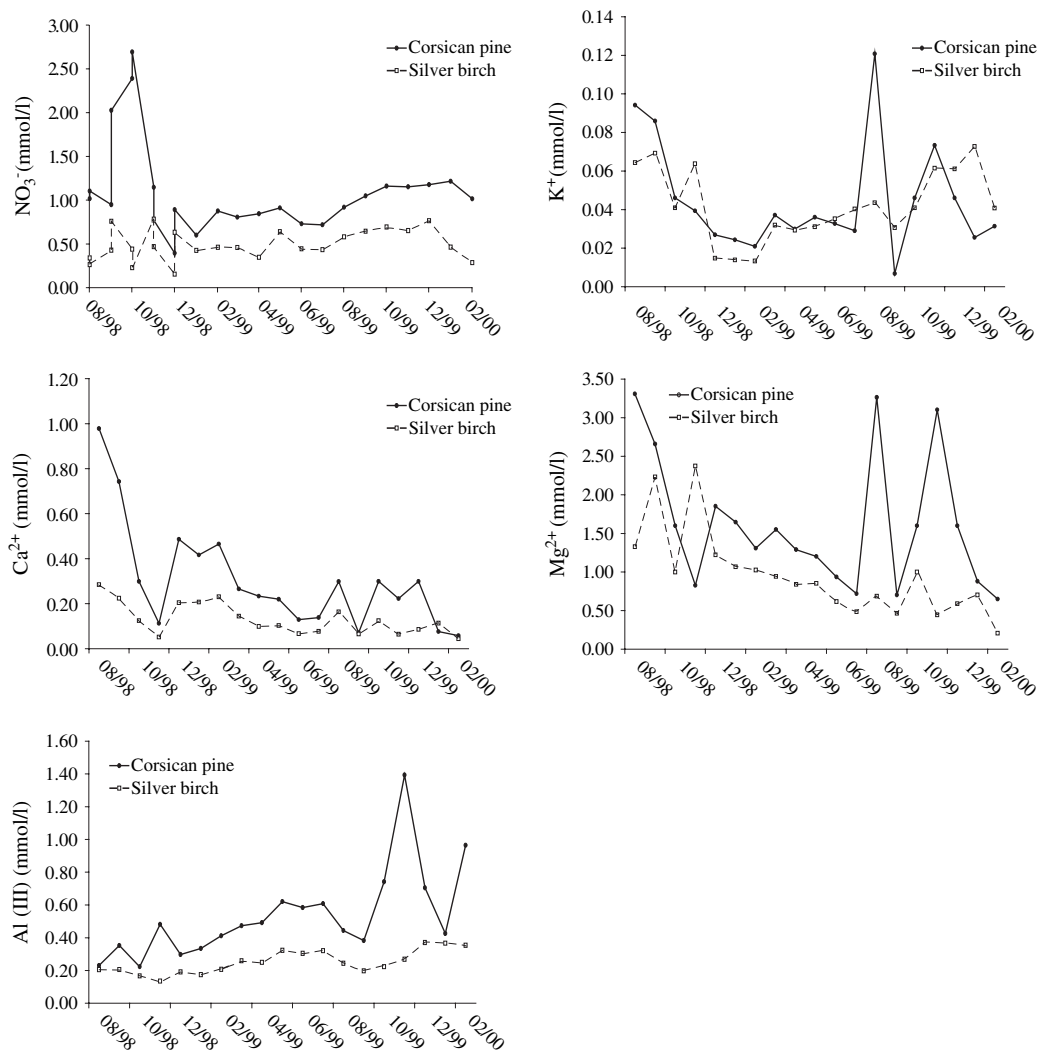


Fig. 2. Concentrations ( $\text{mmol l}^{-1}$ ) of  $\text{NO}_3^-$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{Al(III)}$  in percolation water of the Corsican pine plantation and the silver birch stand.

and Draaijers, 2002). Although nitrogen saturation in the canopy might be expected, the canopy uptake of nitrogen is governed by many other factors such as the tree's nitrogen demand and the soil N pool. However, our results contrast with the research of Neary and Gizyn (1994) and Houle et al. (1999) who found  $\text{NH}_4^+$  retention to be higher at a coniferous site than at a deciduous site, in regions with a much lower atmospheric  $\text{NH}_4^+$  deposition. Brumme et al. (1992) and Harisson et al. (1991) reported faster uptake rates for nitrogen in the canopy of beech and birch compared to spruce. According to Harisson et al. (2000), these differences may be attributable to a combination of thinner cuticles and higher wettability of foliage and bark of beech and birch.

The net throughfall flux of  $\text{H}^+$  was negative for both species throughout the measurement period, which indicates a net uptake of protons in the canopy and demonstrates the ability of both forest covers to

neutralize acidic input. Since both forest stands receive similar wet-only atmospheric loads and are subjected to similar ecological and meteorological conditions, tree species must be the main regulating factor of neutralization (Mahendrappa, 1989). Dry deposition of atmospheric ammonia leads to the consumption of protons, and is much lower for the birch canopy than the pine canopy. This partly explains why proton deposition to the forest floor is significantly higher in the birch stand. The lower acid neutralizing capacity of birch compared to pine could also be due to the canopy exchange of  $\text{NH}_4^+$  for  $\text{H}^+$  (Neary and Gizyn, 1994), as birch retains more  $\text{NH}_4^+$  in the canopy.

In the present study, nitrate leaching out of the canopy was assumed to be negligible, based on the results of Neary and Gizyn (1994). In earlier research, Garten and Hanson (1990) also found that  $^{15}\text{NO}_3^-$  deposited to deciduous tree leaves was easily removed by washing with water, while  $^{15}\text{NH}_4^+$  was retained and



Table 4

Average monthly concentrations ( $\mu\text{mol l}^{-1}$ ) and standard deviation of elements in the soil solution at 1 m depth during growing season, dormant season and the whole year for the measuring period (September 1998–February 2000) (significance levels of species (Sp.), time (Ti.), and interaction between species and time (Int.) are indicated (ANOVA, repeated measures design))

	Growing season			Dormant season			Study period					
	Mean $\pm$ s.d.	Sign.			Mean $\pm$ s.d.	Sign.			Mean $\pm$ s.d.	Sign.		
		Sp.	Ti.	Int.		Sp.	Ti.	Int.		Sp.	Ti.	Int.
<b>Ammonium-N-N</b>												
Silver birch	7 $\pm$ 7	n.s.	*	n.s.	19 $\pm$ 26	n.s.	**	n.s.	15 $\pm$ 23	n.s.	***	n.s.
Corsican pine	5 $\pm$ 36				18 $\pm$ 30				13 $\pm$ 26			
<b>Nitrate-N</b>												
Silver birch	453 $\pm$ 258	***	***	***	460 $\pm$ 165	**	**	n.s.	456 $\pm$ 216	***	***	***
Corsican pine	1466 $\pm$ 558				800 $\pm$ 144				1150 $\pm$ 523			
<b>Potassium</b>												
Silver birch	46 $\pm$ 20	n.s.	n.s.	n.s.	31 $\pm$ 23	n.s.	***	***	38 $\pm$ 22	*	***	***
Corsican pine	59 $\pm$ 31				41 $\pm$ 33				49 $\pm$ 33			
<b>Calcium</b>												
Silver birch	117 $\pm$ 90	***	***	***	45 $\pm$ 33	*	***	n.s.	132 $\pm$ 80	**	***	***
Corsican pine	294 $\pm$ 347				66 $\pm$ 37				299 $\pm$ 267			
<b>Magnesium</b>												
Silver birch	33 $\pm$ 29	**	***	***	45 $\pm$ 33	**	*	**	39 $\pm$ 30	**	***	***
Corsican pine	58 $\pm$ 41				66 $\pm$ 37				62 $\pm$ 41			
<b>Aluminium</b>												
Silver birch	296 $\pm$ 107	**	***	*	229 $\pm$ 78	n.s.	n.s.	n.s.	263 $\pm$ 96	*	***	n.s.
Corsican pine	607 $\pm$ 278				430 $\pm$ 207				515 $\pm$ 256			
<b>Protons</b>												
Silver birch	41 $\pm$ 13	n.s.	***	n.s.	84 $\pm$ 21	n.s.	***	n.s.	60 $\pm$ 17	n.s.	***	n.s.
Corsican pine	32 $\pm$ 13				69 $\pm$ 26				50 $\pm$ 20			

\* $p < 0.05$ ; \*\* $p < 0.01$ ; \*\*\* $p < 0.001$ .

presumably assimilated into the leaf. According to recent research by Stachurski and Zimka (2002),  $\text{NH}_4^+$  is taken up from rainwater as well as from gas/aerosol, whereas  $\text{NO}_3^-$  canopy uptake only occurs from the gas/aerosol fraction.

In both stands, nitrogen input through litterfall was lower than the deposition load. In forests less polluted by atmospheric depositions, input by litterfall is by far the dominant nitrogen source (Ukonmaanaho and Starr, 2002). This emphasizes the high degree of disturbance by external pollution sources in the forests of the present study. Total annual N-input to the forest floor (throughfall deposition and litterfall) was more than 50% lower in

the birch stand than in the pine stand. These differences were mainly due to differences in throughfall deposition.

For both stands, canopies were sources of base cations during the growing and dormant seasons. This can be attributed to the washing off of dry deposition (e.g. soil dust) captured by the canopy, and to element leaching from the leaves. Factors that possibly regulate canopy leaching of base cations are (i) precipitation characteristics such as duration (Lovett and Lindberg, 1984), incidence (Lovett and Schaefer, 1992) and acidity of rain events (Schaefer et al., 1988; Schaefer and Reiners, 1990; Hansen et al., 1994), (ii) canopy characteristics such as above-ground biomass, nutrient amount and concentrations in tree leaves (Lovett et al., 1989) and (iii) forest soil characteristics such as extractable amount of base cations and soil solution characteristics (Lovett and Schaefer, 1992).

According to the canopy budget calculation model, the foliar leaching of potassium accounted for 75% and 53% of throughfall deposition in the silver birch and the Corsican pine stands, respectively. Houle et al. (1999), Van Ek and Draaijers (1994), Parker (1983) and Ragsdale et al. (1992) also reported that the leaching of  $\text{K}^+$  was very important, and generally over 70% of throughfall deposition. Potassium throughfall deposition was significantly higher under birch, which was

Table 5

Element percolation ( $\text{mol ha}^{-1}$  period $^{-1}$ ) at 1 m depth under the Corsican pine and the silver birch stand

	September 1998–August 1999		September 1999–February 2000	
	Birch	Pine	Birch	Pine
Ammonium-N	107	107	0	14
Nitrate-N	1814	4029	443	1600
Potassium	141	159	59	59
Calcium	517	1187	130	431
Magnesium	173	239	21	66
Aluminium	878	1907	341	678
Protons	200	200	100	100

mainly due to higher leaching during the growing season. This can be attributable to thinner cuticles and higher wettability of birch foliage. Houle et al. (1999), Van Ek and Draaijers (1994) and Alcock & Morton (1985) also found that deciduous canopies leached significantly more  $K^+$  than coniferous canopies. These differences were most explicit during the growing season, which can be attributed to the higher physiological activity of both forest types in this season.

No significant difference in calcium throughfall deposition between both stands was found in our case study. According to the canopy budget model,  $Ca^{2+}$  was taken up by both canopies, and mostly by the pine canopy. It is probable that the calculated low uptake rates of Ca should be seen as bias on the filtering approach, since in literature only leaching is reported (Houle et al., 1999; Lovett and Schaefer, 1992). The major assumptions and weakness of the canopy budget model of Ulrich (1983) can be summarized as (i) the use of bulk deposition instead of wet-only deposition, (ii) the assumption that Mg, Ca and K containing particles are deposited with equal efficiency as particles containing Na and (iii) the assumed relation between wet and dry deposition of particles. In spite of these assumptions, Draaijers et al. (1998) stated that canopy exchange and dry deposition of Mg, Ca and K can be estimated reasonably well using throughfall and precipitation measurements in association with the canopy budget model.

Magnesium throughfall deposition was significantly higher under the pine canopy due to the higher calculated dry deposition. The calculated contribution of canopy leaching to total throughfall deposition of  $Mg^{2+}$  (21% for birch and 14% for pine) was between that of  $K^+$  and  $Ca^{2+}$ , and therefore agrees with previous research (Houle et al., 1999; Van Ek and Draaijers, 1994; Ragsdale et al., 1992).

The leaching of potassium and magnesium and the uptake of ammonium were highest for the birch canopy, despite its summergreen character and the lower basal area and stem volume of the birch stand. As the meteorological and soil conditions were similar, the higher canopy leaching for birch might be due to higher nutrient concentrations in the birch leaves compared to pine needles (Johansson, 1995). The concentration gradient between water on the surface and the interior of the leaves modifies the degree of uptake or leaching (Schaefer et al., 1988; Lovett et al., 1989; Schaefer and Reiners, 1990).

#### 4.2. Output of elements

In the birch stand, the percolation of nitrate at 1 m depth was 60% lower than in the pine stand. This can be explained by the lower N input in the birch stand. Macdonald et al. (2002) found that throughfall nitrogen

deposition accounted for 60% of the variation in nitrate percolation for 181 forests throughout Europe. They found that the relationship between throughfall input and percolation was identical to that reported by Dise and Wright (1995).

The difference in nitrate percolation under both forest types might also be related to species-specific nitrogen uptake and retention capacity. For certain broadleaved species, such as ash (*Fraxinus* spp.) and oak (*Quercus* spp.), high ammonium concentrations are a stimulus for nitrate uptake (Stadler et al., 1993), while nitrate uptake by roots of coniferous trees is known to be strongly reduced in the presence of ammonium (Marschner et al., 1991; Rennenberg et al., 1996). No specific literature was found for Corsican pine or silver birch. Gebauer et al. (2000) presume that nitrate is a more important mineral N source for broadleaved trees than for conifers. This supports the scenario that birch might be able to take up nitrate-N from deeper soil horizons, whereas pine trees are only able to consume ammonium available in the upper soil layer. During the dormant season, uptake can be assumed to be low for both species, and nitrogen output will be strongly related to input.

Assuming a steady state in the litter layer, nitrogen uptake by both species can be calculated by subtracting soil nitrogen percolation from nitrogen input. This uptake, expressed per  $m^3$  stem volume, was 116 and 45 mol N for birch and pine, respectively. However, as a consequence of the much higher total stem volume in the pine stand, total root N uptake was much higher in the pine stand.

The capacity of a forest to retain nitrogen depends on the tree species it contains. After a 9-year period of chronic nitrogen addition, Magill et al. (2000) reported an overall N retention of 96% in a hardwood plot and 85% in a coniferous plot. Although the hardwood stand did not show significant increases in nitrate percolation, nitrate losses were measured in the pine stand after the first year of addition. Soil organic matter was suggested to be the major nitrogen pool. Similarly, Gebauer et al. (2000) found that young and mature conifer stands retained less than 10% of throughfall N deposition, while broadleaf trees were able to retain 70% and more.

Although distinct differences in nitrate percolation were apparent in this study, both stands showed clear symptoms of a nitrogen surplus and soil acidification processes. The forest soil is strongly impoverished because nitrate percolation is linked to losses of base cations (Carnol et al., 1997), which reduces the buffering potential of the soil. Soil acidification is reflected in both stands by the loss of base cations, and by the mobilisation of aluminium in particular. The percolation of nitrate, calcium and aluminium over a 1-year period was more than twice as high under Corsican pine than under birch. The species effect on soil percolation

was even stronger during the dormant season. Three mechanisms might be responsible for the observed differences (Meesenburg et al., 1995; Wesselink and Mulder, 1995). First, a higher input of base cations by throughfall and litter can result in a higher output. This cannot explain the present differences in percolation, since the total input of  $Mg^{2+}$  and  $K^+$  to the forest floor was higher in the birch stand. Also the slightly higher total input of  $Ca^{2+}$  in the pine stand cannot explain why  $Ca^{2+}$  percolation was more than twice as high under pine. Secondly, differences in soil acidification due to the different proton input or the different input of base cations could be responsible for percolation differences under the species. Soil acidification will induce displacement of cations by protons, thereby reducing the cation pool and increasing the proportion of Al(III) on the soil-exchange complex. More detailed research and soil analyses are necessary to prove this hypothesis. A third possibility is that the higher percolation of anions under pine involves a higher soluble cation fraction, making total cation percolation a function of total anion percolation (Johnson, 1992).

The conversion of high-density homogeneous pine forests into lower density birch forests could potentially reduce the impact of deposition on forest soils. If the N input to forest soil could be reduced, e.g. by replacing coniferous species with broadleaved species, the output of nitrate and consequently of base and acid cations could drop rapidly (Boxman et al., 1998; Bredemeier et al., 1998; Tietema et al., 1998). As well as reducing the nitrogen load, such a conversion also slows down the acidification rate of forest soils and improves the nitrogen uptake capacity of the ecosystem. However, caution is urged because clear symptoms of nitrogen saturation and soil acidification were also found in the birch stand. The sustainable provision of forest ecosystems' regulation function consequently requires a drastic reduction in emissions on the long term.

## 5. Conclusions

A naturally regenerated stand of silver birch (*B. pendula* Roth) contributes less to acidification and eutrophication of the soil than the homogeneous plantation of Corsican pine (*P. nigra* ssp. *laricio* Maire). The lower nitrogen throughfall deposition in the birch stand was reflected in a 60% lower nitrate leaching at 1 m depth compared with pine. The forest soil is strongly impoverished because nitrate leaching is linked to losses of aluminium and base cations: leaching of calcium and aluminium over a 1-year period was more than twice as high under the Corsican pine stand than under the birch stand. It is concluded that, considering these results, the conversion of high-density homogeneous pine planta-

tions into lower density birch forests could potentially reduce the impact of deposition on forest soils.

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