Nr 190 • 1993

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SOIL ACIDIFICATION AND ELEMENT FLUXES OF A *FAGUS SYLVATICA* FOREST AS INFLUENCED BY SIMULATED NITROGEN DEPOSITION

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(Received July 6, 1991; revised November 8, 1991)

Abstract. The influence of simulated 3- and 9-fold increases in the current N deposition on element fluxes and soil acidification was investigated in a beech forest in S Sweden. Treatments, in three replicates, were 66 and 198 kg N ha⁻¹ yr⁻¹ as NH₄NO₃ during 5.5 yr. All the supplied N could not be accounted for by leaching or soil accumulation, and possible pathways are discussed. Leaching of NO₃⁻ increased greatly upon treatment. Nitrification, also occurring in the control soil, was considerably enhanced by the low-N but not by the high-N treatment. Leaching of NO₃⁻ (added or resulting from nitrification) had a strongly acidifying influence. Soil-solution (cup lysimeters at 30 cm depth) pH was 4.51, 4.07 and 3.81 in the control, low- and high-N plots, respectively. The buffering of protons shifted from the base-cation to the Al buffer range. The base-cation/Al ratio (equivalents) of cation mobilization was 2.7 in the control, 1.1 in low-N and 0.15 in high-N plots. Also Mn and Rb fluxes were drastically increased. The C:N ratio of beech litterfall decreased with increasing N supply but fluxes of elements in litterfall and in throughfall were not influenced. The bulk deposition of N was 18 kg ha⁻¹ yr⁻¹. A moderate further increase in the N deposition to this N-saturated forest would considerably increase NO₃⁻ leaching and soil acidity and cause depletion of base cations and mobilization of Al and Mn.

1. Introduction

Nitrogen has long been considered the nutrient limiting the primary productivity of forests. During the last decades the situation has changed, and atmospheric N deposition now contributes N in quantities well above the demand of forests in much of Europe. Many forest soils are approaching a state of 'N saturation' in which biomass productivity does not respond to increased N supply (Nihlgård, 1985). The current load to SW Sweden, 15 to 20 kg N ha⁻¹ yr⁻¹, is in the upper part of the range of the proposed 'critical load' of N (Nilsson and Grennfelt, 1988). In most forest types, there is little tendency for inorganic N to accumulate in the soil. The NO₃⁻ anion is only weakly adsorbed on soil surfaces and is thus readily leached if not taken up by roots. The NH₄⁺ ion can be retained on cation exchange sites. However, quantities of exchangeable NH₄⁺ on exchange sites in forest soils are usually insignificant due to the high biological demand and to nitrification of NH₄⁺ not incorporated into biological tissue. The balance between N supply and biological N demand generally controls leaching of NO₃⁻ and cations (see e.g. Tamm, 1991).

An increase in the NH₄⁺-N supply to a forest may promote nitrification which is a strongly acidifying process. Soil acidification potentially changes element fluxes not only in the soil but also in the entire ecosystem, including root uptake, throughfall

and litterfall (Reuss and Johnson, 1986; Parker, 1990; Tamm, 1991).

Effects of N additions have been much studied in coniferous forests (see e.g. Tamm, 1991) but little is known about effects on beech-forest ecosystems. The aim of the present study was to quantify the influence of simulated 3- and 9-fold increases in the current N deposition on (1) element fluxes and soil acidification and (2) the fate of N in a beech-forest ecosystem in S Sweden. Granulated NH_4NO_3 was repeatedly supplied during 5.5 yr, and chemical changes in throughfall, litterfall, soil solution and soil were studied during the last experimental year. The study forms part of an integrated experiment where also effects on leaf chemistry, tree growth, performance and nutrient dynamics, vegetation changes and macrofungal fruitbody production were studied (Tyler *et al.*, 1992).

2. Site Description and Experimental Design

The experimental site is situated in a *Fagus sylvatica* forest, Maglehems Ora (55 °35'N, 14 °07'E) in eastern Skåne, southernmost Sweden, 4 km from the Baltic Sea. The elevation of this level site is 76 to 78 m a s l which is well above the highest coast-line. The bed-rock is a fine-grained gneiss which is covered by a delta deposit of glaciofluvial sand, only slightly mixed with moraine. The texture is mainly fine sand (0.06 to 0.6 mm) with a low clay (<10%) and a low boulder content. The site is very well drained and the soil profile is a light Dystric Cambisol, transitional to a Cambic Arenosol with less distinct horizons. The humus type is a weakly aggregated sandy mull, at sites slightly transitional to moder. The soil has a pH-

TABLE I

The pH (0.2 M KCl) of the soils before the experiment (August 1985) and after nearly 5 yr (March/April 1990). Sieved (2 mm mesh) fresh soil. Mean±s.d. n=6 (two samples from each plot). Low N and High N:66 and 198 kg N ha⁻¹ yr⁻¹, respectively. Neither in 1985, nor in 1990 did pH differ significantly between control, low N and high N in any of the horizons (Tukey test; p level 0.05). Significance of the difference between years (in the same treatment and soil horizon): ^a p<0.05, ^b p<0.01, ^c p<0.001 (Student's t test). Data provided by B. Nihlgård (pers. comm.)

		1985	1990
A horizon	Control	3.56±0.11	3.66±0.07
	Low N	3.61±0.13	3.59±0.13
	High N	3.56±0.10	3.59±0.12
AB horizon	Control	3.66±0.04	3.54±0.07 ^b
	Low N	3.68±0.05	3.45±0.12 ^b
	High N	3.70±0.04	3.44±0.10 ^c
B horizon	Control	3.87±0.04	3.80±0.04 ^b
	Low N	3.84±0.03	3.78±0.06 ^a
	High N	3.80±0.07	3.74±0.09

TABLE II

Base (Ca, Mg, K, Na) saturation, exchangeable Al percentage and effective cation exchange capacity in different horizons of the soil before the start of the experiment.
Data provided by B. Nihlgård (pers. comm.)

Horizon	Base saturation %	Exchangeable Al %	Effective CEC $\mu\text{eq g}^{-1} \text{ dw}$
A (0-5 cm)	32.2	57.7	16.1
AB (5-15 cm)	10.1	83.5	17.2
B (15-30 cm)	10.7	84.0	16.7

KCl of 3.6 in the A horizon and 3.8 in the B horizon (Table I). The percentage base saturation and percentage exchangeable Al are given in Table II.

The area has mainly been forested for centuries. The present beech stand is even aged, established after an ample mast production in 1869. No thinning has occurred since 1980 and the canopy is closed. The number of trees (exclusively beech) is ca 200 ha^{-1} , the basal area ca 30 $\text{m}^2 \text{ ha}^{-1}$ and the mean tree height 32 m. The shrub layer is sparse. The field layer, not fully closed, is dominated by *Oxalis acetosella*, *Anemone nemorosa* and *Cardamine bulbifera* with some *Viola riviniana*, *V. reichenbachiana*, *Rubus idaeus* and a few other species.

The climate is temperate, suboceanic and subhumid. The mean temperature of July (warmest month) is 17 °C and that of February (coldest) -1 °C. The years 1985-1987, especially the winters, were colder than average whereas there were positive temperature anomalies in 1989-1990, e.g. +5.5 °C in February 1990. Except for the dry year 1989, the annual precipitation during the 6 yr of experiment was close to or slightly above the long-time (1961-1989) average, 602 mm. Usually, the precipitation is fairly evenly distributed over the year though somewhat lower during the spring months. These data are from the meteorological station at Vitemölla, 7 km away, where the precipitation November 1989-October 1990 was 555 mm which is less than was measured at the experiment site.

In the 1.3-ha experimental area, nine 25 × 25 m plots with 5-m buffer strips in between were laid out in 1985. Controls and two levels of N treatment were randomized in triplicates. The treatment plots were supplied with granulated (1 to 3 mm diam.) 'kalkammonsalpeter', made up of NH_4NO_3 with CaCO_3 and MgCO_3 additions. The granulate, containing 27.7% N (13.9% $\text{NH}_4\text{-N}$ and 13.8% $\text{NO}_3\text{-N}$), 4% CaCO_3 , 2% MgCO_3 and traces of other elements, see Table IV) was distributed by hand on one occasion each month from May to September during 1986 through 1990. The supply amounted to 66 and 198 $\text{kg N ha}^{-1} \text{ yr}^{-1}$ in the low- and high-N treatments, respectively. Prior to this, 16 and 48 $\text{kg N ha}^{-1} \text{ yr}^{-1}$ (divided on two occasions) were supplied to the low- and high-N plots, respectively, in the autumn of 1985. The annual supply during 1986-1990 corresponded to about 3 and 9 times the atmospheric wet deposition of N to the stand.

3. Materials and Methods

Bulk deposition was collected in four samplers (shaded polyethylene, PE, funnels, diam. 20 cm, with PE sieves, connected to 10-L PE bottles) placed 2 m above ground in a clear-cut area 700 m from the plots. In each plot, throughfall was collected in five shaded samplers (funnels as above, 5-L bottles) placed c. 0.5 m above ground and randomly distributed in the area >7.5 m away from the plot border to avoid border effects and >1.5 m away from trunks to diminish the influence from stemflow water coming loose from the trunks, some of which were slightly inclined. Litterfall was also collected in these funnels.

Bulk deposition and throughfall were weighed in the field. Bulk deposition waters were treated as single samples but the five throughfall samples were pooled together. A 1.3-L subsample was brought to the laboratory. The litter from the five funnels was also pooled together.

Soil solution was collected with ceramic cup lysimeters (P80, vacuum 60 to 80 kPa). In a regular pattern in each of the nine forest plots, four lysimeters were installed at 30 cm soil depth (upper part of the B horizon) >5 m away from the plot border. Collecting bottles (Duran glass) were placed in a frigolite-covered PVC container below ground to keep samples dark and cool. To allow lysimeters to equilibrate with the soil solution, the first few lysimeter solutions, obtained within 2 mo of installation, were discarded.

After this equilibration time, soil solution was sampled continuously from 12 July, 1989 to 9 November, 1990. Bulk deposition, throughfall and litterfall were sampled continuously from 8 November, 1989 to 9 November, 1990. Sampling period duration was 12 to 39 days during the vegetation seasons and 40 to 46 days during the defoliated season.

Soil samples were taken in August 1985, before the experiment, and in March/April 1990 from the A (0 to 5 cm), AB (5 to 15 cm) and B (15 to 30 cm) horizons. From each horizon, two bulk samples were attained, each consisting of fifteen subsamples taken out with a thin soil auger along either of the two diagonals.

The aqueous samples were stored at -20°C . After thawing, throughfall samples were filtered (paper OOR, Munktell STORA) whereas bulk precipitation and soil leachate were not. Subsamples were taken out for analysis of NH_4^+ , NO_3^- , SO_4^{2-} , Cl^- and dissolved organic carbon (DOC). Subsamples (700 to 1000 mL) for metal analysis were transferred to a 1-L Erlenmeyer flask which was sealed with a hood of filter paper and evaporated at 105°C . The residue was treated with 10 mL conc. HNO_3 (anal. gr.) to destruct organic matter and redissolve the metals. The volume was reduced by evaporation to c. 2 mL and diluted with deionized water to 25 mL prior to analysis.

The litterfall samples, stored in paper bags at -20°C , were dried at c. 40°C , separated into leaf litter and other litter and weighed. Samples from November 1989 to November 1990 were bulked together. Leaf-litter subsamples (2.5 g) were digested in 30 mL conc. HNO_3 . The samples were evaporated, filtered and diluted.

The soil samples were dried at 85 °C for > 24 hr and sieved (mesh 2 mm). Extracts for NH_4^+ and NO_3^- analyses were obtained using 0.2 M KCl. Extracts for analyses of Na, K, Ca and Mg were obtained using 1M ammonium acetate (pH 7.0). Extracts for analyses of exchangeable acidity and exchangeable Al were obtained using 1M KCl.

The following analytical methods were used for aqueous and/or litter samples: pH was determined electrometrically. NH_4^+ was determined colorimetrically (flow injection analysis, FIA); NO_3^- , SO_4^{2-} and Cl^- by ion chromatography; Rb (with 2000 ppm K in solutions) by flame emission; Cd by AAS; S (litter only), P, Al, Ca, Cu, K, Mg, Mn, Na and Zn by inductively coupled plasma spectrometry. DOC in aqueous samples was analyzed by infrared technique (C analyzer). In litter samples, C was analyzed using infrared technique and total N by Kjeldahl analysis. Soil 0.2 M KCl extracts were analysed for NH_4^+ and NO_3^- using FIA. On soil 1M KCl extracts exchangeable acidity was determined titrimetrically with NaOH after addition of fenolftaleine as an indicator, and exchangeable Al was thereafter determined titrimetrically with HCl on the same sample after addition of NaF.

Element fluxes for the 1-yr period from 8 November, 1989 to 9 November, 1990 were calculated as follows: For bulk precipitation and throughfall, the element fluxes (concentration times water volume) were calculated for each of the eleven sampling occasions. The element fluxes in litterfall were calculated by multiplying the concentrations in the leaf-litter fraction by the dry weight of the pooled 1-yr total litterfall. The soil-solution fluxes were calculated for each of the nine sampling occasions using measured lysimetric soil-solution concentrations and calculated soil water fluxes, assuming the flux of Cl^- in throughfall being equal to the flux of Cl^- in the soil solution of the control plots (Mulder *et al.*, 1989b).

One-year element fluxes were calculated by adding up the fluxes for all sampling occasions. For some elements, fluxes in bulk deposition, throughfall and soil solution were calculated for the winter half year (8 November, 1989–27 April 1990; 'defoliated' period) and the summer half year (27 April–9 November, 1990; 'foliated' period) separately.

The measured water volumes for the 1-yr period was 852 L m⁻² for bulk deposition and 571 L m⁻² for throughfall (average of the control and the two treatments). The volume of soil leachate, as calculated using the Cl^- flux, was 317 L m⁻² as an average.

4. Results

4.1. CHEMISTRY OF THROUGHFALL AND SOIL SOLUTION

The tree canopy has a strong influence on the chemistry of the precipitation. The bulk deposition pH was 4.47 (annual arithmetic mean) whereas the throughfall pH was 5.9 to 6.0 (Table III). Throughfall pH was c. 1.5 units higher during the foliated period (mean value 6.5) than during the defoliated period (mean 5.0). Upon passage of the canopy, the precipitation was enriched in NH_4^+ , NO_3^- , SO_4^{2-} , P,

TABLE III

Mean pH and element concentrations (mg L^{-1}) in bulk deposition, throughfall and soil solution from the upper B horizon. Three replicates. Eleven sampling occasions (8 November 1989-9 November 1990). DOC=dissolved organic carbon. n.a.=not analyzed. Means lacking common letters differ significantly ($p < 0.05$) between treatments (Tukey test). n: 44 for bulk deposition, 33 for throughfall and 55-87 for soil solution

	pH	DOC	$\text{NH}_4^+\text{-N}$	$\text{NO}_3^-\text{-N}$	$\text{SO}_4^{2-}\text{-S}$	P	Cl	Al	Ca	Cd	Cu	K	Mg	Mn	Na	Rb	Zn
Bulk deposition	4.47	n.a.	1.41	1.07	1.52	0.054	4.31	0.04	0.48	0.00014	0.00446	0.44	0.21	0.01	2.00	0.018	0.018
Throughfall																	
Control	5.90 ^a	9.90 ^a	2.08 ^a	1.57 ^a	2.55 ^a	0.651 ^a	9.24 ^a	0.04 ^a	2.24 ^a	0.00012 ^a	0.00493 ^a	4.42 ^a	0.92 ^a	0.35 ^a	3.77 ^a	0.048 ^a	0.015 ^a
Low N	5.98 ^a	10.3 ^a	2.15 ^a	1.46 ^a	2.33 ^a	0.429 ^a	7.85 ^a	0.04 ^a	2.06 ^a	0.00011 ^a	0.00529 ^a	4.10 ^a	0.84 ^a	0.23 ^a	3.18 ^a	0.018 ^a	0.013 ^a
High N	5.94 ^a	10.3 ^a	2.10 ^a	1.68 ^a	2.72 ^a	0.461 ^a	10.0 ^a	0.04 ^a	2.30 ^a	0.00012 ^a	0.00523 ^a	4.34 ^a	0.97 ^a	0.35 ^a	3.97 ^a	0.052 ^a	0.016 ^a
Soil solution																	
Control	4.51 ^a	12.2 ^c	0.69 ^b	2.86 ^c	5.77 ^a	n.a.	17.1 ^a	1.22 ^c	6.92 ^c	0.00102 ^c	0.00069 ^a	0.56 ^a	1.30 ^c	0.41 ^c	6.44 ^a	0.002 ^c	0.074 ^c
Low N	4.07 ^b	18.1 ^b	1.94 ^b	29.0 ^b	4.93 ^a	n.a.	15.6 ^a	7.52 ^b	21.6 ^b	0.00324 ^b	0.00081 ^a	3.55 ^b	4.48 ^b	4.61 ^b	7.72 ^a	0.013 ^b	0.273 ^b
High N	3.81 ^c	24.3 ^a	3.76 ^a	56.3 ^a	5.43 ^a	n.a.	19.7 ^a	17.9 ^a	28.4 ^a	0.00571 ^a	0.00087 ^a	4.17 ^b	8.01 ^a	9.74 ^a	7.72 ^a	0.026 ^a	0.428 ^a

Cl^- , Ca, Mg, Na, Rb and especially K and Mn whereas the concentrations of Al, Cd, Cu and Zn were lower or unaltered in the throughfall compared with the bulk precipitation. None of the elements showed any significant difference in mean throughfall concentrations between the treatments low N, high N and control.

Contrary to the throughfall, the soil solution responded very clearly to the treatments. The overall response was a strong acidification. The mean pH was 0.4 and 0.7 units lower in the soil solution of the low- and high- N plots, respectively, than in the control which had a mean pH of 4.51.

In the low-N soil solution, the mean NH_4^+ concentration was not statistically different from the control but the NO_3^- concentration was increased ten times. In the high-N soil solution, the NH_4^+ concentration was c. five times and NO_3^- c. 20 times the control. Increasing NH_4NO_3 load gave rise to increased leaching of Al, Ca, Cd, K, Mg, Mn, Rb, and Zn. Compared with the control, the concentrations in the high-N soil solutions were increased 4 to 13 times for most elements, 15 times for Al and even 24 times for Mn. Only for Ca, K and Mg can part of this increase be attributed to contribution from the fertilizer.

In the treated plots, the Al concentration in the soil solution from the upper B horizon (30 cm) was very high, up to 17 meq L^{-1} (150 mg L^{-1}) in the high-N treatment (Figure 1). The pH, the NO_3^- concentration and the total Al concentration of the soil solution were closely related. There was a dramatic decrease in soil-solution pH with increasing NO_3^- concentration. Likewise, there was a concomitant increase in the Al solubilization with a decrease in pH. A treatment effect is clearly displayed in the plots; H^+ , Al and NO_3^- concentrations increasing in the order control < low-N < high-N.

Interestingly, also the organic-matter solubilization seems to have been enhanced by the treatment, as shown by significantly increased DOC concentrations (Table III). The concentrations of SO_4^{2-} and Cl^- were not altered by the treatments. These anions are only weakly adsorbed by this Dystric Cambisol (Table IV), and the treatments did not seem to change the adsorption capacity, as the Fe- and Al-oxide content in the B horizon normally is low in this type of soil.

4.2. FLUXES AND SOIL BUDGETS

Bulk deposition of N was rather high, $1.8 \text{ g N m}^{-2} \text{ yr}^{-1}$ (Table IV). The N fluxes in bulk deposition and throughfall were rather similar. The annual fluxes of elements (including N) in throughfall were not obviously influenced by the treatments. Neither were the element fluxes in litterfall, even though the litterfall C/N ratio decreased with increasing N treatment: 43.5, 40.0 and 37.6 in the control, low-N and high-N plots, respectively (difference between control and high-N significant at $p < 0.05$, Tukey test).

In the soil solution, however, the low-N and, even more, the high-N treatment considerably increased the fluxes not only of NH_4^+ and NO_3^- but also of DOC, SO_4^{2-} , Al, Ca, Cd, Cu, K, Mg, Mn, Rb, and Zn.

Using element fluxes, a soil budget can be calculated from the input terms

Fig. 1. pH and concentrations of NO₃⁻ and Al (meq L⁻¹) in the soil solutions. Fifteen sampling occasions from 12 July 1989 to 9 November 1990. +: control; crosses: low-N; quadrats: high-N treatment.

TABLE IV

Fluxes and calculated soil budget of elements ($\text{mg m}^{-2} \text{ yr}^{-1}$). Three replicates. Soil-water fluxes estimated assuming Cl^- input equal to Cl^- output. Nitrogen in litterfall and biomass increment (above-ground biomass incorporation) considered as $\text{NH}_4^+\text{-N}$. In aqueous fluxes, S is in the form of $\text{SO}_4^{2-}\text{-S}$. Biomass increment: average of two old beech stands on Podzol soil in North-Central Skåne (Bergkvist, Folkesson and Olsson, in prep.); S and P: values from Nihlgård (1972). n.a. = not analyzed

	DOC	$\text{NH}_4^+\text{-N}$	$\text{NO}_3^-\text{-N}$	Tot-N	S	P	Al	Ca	Cd	Cu	K	Mg	Mn	Na	Rb	Zn
Bulk deposition (BD)	n.a.	990	783	1770	1230	29	28	332	0.114	3.66	415	237	8	2600	7.74	14.00
Throughfall (TF)																
Control	4110	998	716	1710	1270	211	20	909	0.071	2.19	1650	434	111	2300	12.8	7.53
±St dev	321	235	12	247	9	49	0.2	63	0.004	0.36	194	6	25	90	1.46	0.29
Low N	4140	1040	699	1740	1170	154	19	816	0.060	2.29	1460	390	88	1960	11.2	6.69
±St dev	382	76	31	107	54	27	2	46	0.008	0.34	123	43	13	288	2.85	0.54
High N	4080	953	751	1700	1240	165	20	905	0.065	2.43	1580	443	113	2280	12.5	8.21
±St dev	348	116	63	179	35	28	1	35	0.006	0.44	101	1	7	82	0.48	1.27
Litterfall (LF)																
Control	-	4960	-	4960	493	510	37	4660	0.051	2.01	1310	468	887	157	5.10	13.5
±St dev	-	505	-	505	38	10	2	358	0.007	0.04	85	62	55	10	0.72	1.23
Low N	-	5250	-	5250	504	418	36	4580	0.054	1.71	1330	497	775	153	5.15	12.9
±St dev	-	633	-	633	41	19	0.5	332	0.011	0.25	209	28	78	9	1.24	1.28
High N	-	5750	-	5750	509	428	36	4310	0.048	1.78	1290	502	758	144	5.33	12.1
±St dev	-	441	-	441	29	30	3	207	0.013	0.21	59	46	41	11	0.59	1.34
Fertilizer addition (FA)																
Control	-	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Low N	-	3310	3290	6600	14	25	12	1020	0.004	0.003	65	489	3	17	0.00	0.13
High N	-	9940	9860	19800	41	74	35	3060	0.012	0.008	196	1470	9	50	0.00	0.40
Biomass increment (BI)																
-	-	1500	-	1500	250	n.a.	4	533	0.011	1.20	660	108	92	17	4.00	3.23

Table IV (Continued)

	DOC	NH ₄ ⁺ -N	NO ₃ ⁻ -N	Tot-N	S	P	Al	Ca	Cd	Cu	K	Mg	Mn	Na	Rb	Zn
Soil solution (SS)																
Control	3780	122	1380	1500	1760	n.a.	383	2870	0.368	0.19	208	508	171	1790	0.67	28.2
±St dev	973	82	1260	1340	213		93	1540	0.075	0.04	123	210	106	511	0.24	9.49
Low N	6580	398	6830	7230	2020	n.a.	1610	5110	0.915	0.22	666	977	978	1980	2.68	64.6
±St dev	1770	138	3410	3550	528		1350	2440	0.470	0.15	568	267	505	304	2.32	34.9
High N	8440	793	10700	11500	2100	n.a.	3280	5280	1.15	0.28	863	1600	1830	1730	5.32	76.9
±St dev	3330	426	3990	4420	1150		402	1070	0.204	0.07	333	119	394	482	0.69	37.6
Soil budget (BD+FA-BI-SS)																
Control	-	-632	-597	-1230	-780	-	-358	-3070	-0.266	2.28	-454	-379	-255	793	3.07	-17.4
Low N	-	2400	-2760	-360	-1030	-	-1570	-4290	-0.808	2.25	-846	-359	-1060	620	1.06	-53.7
High N	-	8640	-57	8570	-1080	-	-3220	-2420	-1.04	2.19	-912	-1	-1900	904	-1.58	-65.8

throughfall (TF), litterfall (LF) and fertilizer addition (FA) minus the output terms root uptake (RU) and leaching with soil solution (SS). Only part of the RU will end up in the above-ground biomass (biomass incorporation, BI), the remainder being internally circulated (internal circulation, IC) as TF and LF minus bulk deposition (BD). Since thus $RU=BI+IC$ and $IC=TF+LF-BD$, the soil budget $TF+LF+FA-RU-SS$ can be reduced to $BD+FA-BI-SS$ (Figure 2).

Both wet and dry deposition should be included in the input term since dry deposition can contribute substantial amounts of e.g. N to a forest (Lovett and Lindberg, 1986). Dry deposition of N and S is known to be of much less importance to deciduous than to coniferous forests, however (see Wiman *et al.*, 1990). The fact that there was no difference between bulk-deposition fluxes and throughfall fluxes of NH_4^+ , NO_3^- and SO_4^{2-} indicates dry deposition to be negligible to the stand under study. Such a comparison does not consider the presence of foliar uptake of e.g. N, however (Schaefer and Reiners, 1990). For the other elements studied here, dry deposition is known to be of minor importance. Though possibly somewhat underestimating the input of some elements, the use of bulk deposition as input to the ecosystem can be justified from what is referred above.

In a beech forest, stemflow is of course an important pathway to the soil for many elements. Results from five beech forest localities in Skåne during 3 yr, show that the stemflow contribution to the total amount reaching the ground (as throughfall+stemflow) is c. 5% for each of NH_4^+ and NO_3^- . For Al, Ca, Cd, Mg, and Zn the corresponding percentage is c. 10%, and for SO_4^{2-} , K, Mn and Na c. 15% (Bergkvist, Folkesson and Olsson, in prep.). Stemflow, creating steep flux gradients near trunks (Falkengren-Grerup, 1989), is thus important for the element transport to the ground. These gradients were beyond the aim of the present study, however. Further, stemflow cannot possibly influence the ground beyond 1 m from the trunks, especially not in this sandy soil where lateral water flux can be considered negligible. Inclusion of the area close to trunks would have led to an overestimation of element fluxes to the ground on a stand-area basis. Being based on lysimeters placed well away from trunks, our fluxes should thus be valid for the stand as a whole with the exception of the area within 1 m of the trunks, which was calculated to constitute less than 6% of the stand area.

In the control plots, the calculated soil budget was negative (decreasing stores) for all elements except for Cu, Na and Rb (Table IV). With increasing fertilizer load, the budget became increasingly negative for S and the acidity-sensitive metals Al, Cd, K, Mn, and Zn. The budget of Mg became less negative due to the Mg addition in the treatment. Also Ca was added in amounts high enough to keep pace with the increased leaching in the treatment plots. The Rb budget turned negative in the high-N plots. Copper and Na were not affected by the treatments and showed positive budgets in both control and treatment soils.

Of the N reaching the soil as throughfall, 88% was leached from the rooting zone (30 cm soil depth) in the control plots. The amount of NO_3^- leaching with the soil solution was 190% of the amount deposited with throughfall but the

Fig. 2. Annual flux of Mn in the control, low-N and high-N plots. $\text{mg m}^{-2} \text{yr}^{-1}$. BD=bulk deposition, TF=throughfall, LF=litterfall, FA=fertilizer addition, BI=incorporation into above-ground biomass, IC=internal circulation ($=\text{TF}+\text{LF}-\text{BD}$), RU=root uptake ($=\text{BI}+\text{IC}$), SS=soil solution. BD, TF, LF, FA and SS measured values, BI from a parallel study, others calculated. $\text{BD}+\text{FA}-\text{BI}-\text{SS}$ gives the Mn release from the 0-30 soil compartment. See further Table IV.

corresponding value for NH_4^+ was only 12%, which indicates nitrification to be important also in the untreated beech-forest soil which is rather acidic.

In the control plots, both NH_4^+ and NO_3^- had negative soil budgets. In the low-N plots, the soil budget of (total inorganic) N was slightly negative. Interestingly, however, the greater part of NH_4^+ added with the fertilizer was efficiently transformed to NO_3^- and leached out of the rooting zone—the leaching of NO_3^- increased five times, compared with the control. In the high-N plots, the NO_3^- leaching was enhanced eight times compared with the control. Most of this originated from the NO_3^- added with the treatment, however. All the mineral N supplied was not leached, however, and more than one-third of the added amount seemed to be retained within the system or lost by other pathways, and the N budget appeared to be greatly positive.

The transport of NH_4^+ through the soil profile was about equal in size during the winter (trees defoliated) and summer (foliated) half years (Table V). In the control plots, the leaching of NO_3^- was much higher during the defoliated than the foliated period whereas this difference was small in the treated plots. The bulk deposition of N to the canopy was fairly similar during the defoliated and the foliated period but the fertilizer was distributed (on five occasions) only during the foliated period. The soil-solution pH was lowest and the Al leaching highest during periods with the highest NO_3^- leaching, i.e. the defoliated period in the control (and low-N) plots but the foliated period in the high-N plots. The increase in the leaching of DOC in the N-treated plots indicates an enhanced mineralisation of soil-organic matter with increasing N load. However, also the added CaCO_3

TABLE V

Mean pH and fluxes of dissolved organic C (DOC), NH_4^+ -N, NO_3^- -N and Al in the soil solutions during the period 8 November 1989–27 April 1990 ('defoliated period'), four sampling occasions, and 27 April–9 November 1990 ('foliated period'), seven sampling occasions. Three replicates. $\text{mg m}^{-2} \text{yr}^{-1}$

	pH	DOC	NH_4^+ -N	NO_3^- -N	Al
Defoliated period					
Control	4.24	2630	51	1220	286
±St dev		749	26	1140	70
Low N	4.02	5130	154	4020	840
±St dev		1590	149	1910	609
High N	3.99	6330	384	4930	1140
±St dev		2380	462	2780	304
Foliated period					
Control	4.61	1150	71	158	97
±St dev		291	56	116	25
Low N	4.09	1450	244	2810	772
±St dev		276	141	1520	744
High N	3.75	2110	410	5730	2130
±St dev		969	39	1720	213

(4%) and MgCO_3 (2%) could have some part in the increased solubilization of soil organic matter. The leaching of DOC was highest in the late autumn, especially in the high-N treatment.

4.3. SOIL ACIDIFICATION AND BUFFERING

The annual H^+ load to the soils caused by the acid production of the transport or transformation of H^+ , NH_4^+ , NO_3^- and SO_4^{2-} was calculated according to Mulder *et al.* (1987). Also the alkalizing effect of the CaCO_3 and MgCO_3 added with the fertilizer, must be included in the calculations (expressed in equivalents):

$$\begin{aligned} \text{H}^+ \text{ load} = & (\text{H}_{\text{in}}^+ - \text{H}_{\text{out}}^+) + (\text{NH}_4^+_{\text{in}} - \text{NH}_4^+_{\text{out}}) - (\text{NO}_3^-_{\text{in}} - \text{NO}_3^-_{\text{out}}) - \\ & - (\text{SO}_4^{2-}_{\text{in}} - \text{SO}_4^{2-}_{\text{out}}) - (\text{CO}_3^{2-}_{\text{in}} - \text{CO}_3^{2-}_{\text{out}}). \end{aligned}$$

The subscripts 'in' and 'out' indicate input flux with bulk deposition+fertilizer addition and output flux from the rooting zone at 30 cm soil depth, respectively. For comparison, the mobilization of Al, base cations and Mn was calculated as well (expressed in equivalents):

$$\text{Al mobilization} = \text{Al}_{\text{out}} - \text{Al}_{\text{in}}.$$

$$\begin{aligned} \text{Base cation mobilization} = & (\text{Na}_{\text{out}} - \text{Na}_{\text{in}}) + (\text{K}_{\text{out}} - \text{K}_{\text{in}}) + (\text{Ca}_{\text{out}} - \text{Ca}_{\text{in}}) \\ & + (\text{Mg}_{\text{out}} - \text{Mg}_{\text{in}}). \end{aligned}$$

TABLE VI

Input and output fluxes of H^+ , NH_4^+ , NO_3^- , SO_4^{2-} and CO_3^{2-} , and the H^+ load to the soil. $\text{meq m}^{-2} \text{ yr}^{-1}$

	H^+	NH_4^+	NO_3^-	SO_4^{2-}	CO_3^{2-}	
Input:						
Bulk deposition	35	71	56	77	0	
Fertilizer addition						
Control	–	0	0	0	0	
Low N	–	237	235	1	92	
High N	–	710	705	3	276	
Output:						
Soil solution (30 cm)						
Control	17	9	99	110	0	
Low N	33	28	488	126	0	
High N	40	57	764	131	0	
	H_{in}^+	$\text{NH}_4^+_{\text{in}}$	$\text{NO}_3^-_{\text{out}}$	$\text{SO}_4^{2-}_{\text{out}}$	$\text{CO}_3^{2-}_{\text{out}}$	$\Sigma \text{H}^+ \text{load}$
	$-\text{H}_{\text{out}}^+$	$-\text{NH}_4^+_{\text{out}}$	$-\text{NO}_3^-_{\text{in}}$	$-\text{SO}_4^{2-}_{\text{in}}$	$-\text{CO}_3^{2-}_{\text{in}}$	
H load						
Control	18	62	43	33	0	156
Low N	2	279	197	48	–92	435
High N	–5	723	3	52	–276	498

$$\text{Mn mobilization} = \text{Mn}_{\text{out}} - \text{Mn}_{\text{in}}$$

In the low-N and high-N plots, the soil acidification, regarded as the H^+ load to the soil compartment 0 to 30 cm, was c. three times that of the control plots (Table VI). In the control plots, NH_4^+ accumulation, nitrification and NO_3^- leaching together accounted for 67% of the total acid production. In the low-N plots, these processes correspondingly accounted for as much as 90% of the acid production, most of which was attributable to nitrification. In the high-N plots, however, the greater part of the acid production could be ascribed to NH_4^+ accumulation or uptake, according to the calculations. Only a minor part of the supplied NH_4^+ could be recovered in the high-N soil after extraction, however (Table VII) and other pathways, e.g. denitrification, are probably involved. The acidification attributable to SO_4^{2-} leaching was of limited importance in both treatments.

In the control plots, the main buffering mechanism was leaching of base cations but Al buffering was also of importance (Table VIII, Figure 3). The low-N treatment

TABLE VII

Soil stores (0-30 cm; mmol m^{-2}) of NH_4^+ and NO_3^- . Sampling in March/April 1990 after four years of N treatment

	NH_4^+	NO_3^-	$\Sigma_{\text{inorganic N}}$
Control	151	14	165
Low N	190	56	246
High N	219	59	278

TABLE VIII

Input, output and mobilization of base cations (Ca, Mg, K, Na), Al^{3+} and Mn^{2+} . Mobilization = output with soil solution minus input with bulk deposition and fertilizer, $\text{meq m}^{-2} \text{ yr}^{-1}$

	Base cations	Al	Mn	Σ mobilization
Input:				
Bulk deposition	160	3	0.3	
Fertilizer addition				
Control	0	0	0.0	
Low N	94	1	0.1	
High N	281	4	0.3	
Output				
Soil solution (30 cm)				
Control	268	43	6.0	
Low N	439	179	35.6	
High N	493	364	66.5	
Mobilization				
Control	108	40	5.7	154
Low N	186	175	35.2	395
High N	52	357	65.9	475

Fig. 3. Base-cation, Al and Mn mobilization in the soil (30 cm). Mobilization = output with soil solution minus input with bulk deposition and fertilizer. Base cations: Ca, Mg, K, Na. Circle area proportional to mobilization ($\text{meq m}^{-2} \text{ yr}^{-1}$). See further Table VII.

doubled the base-cation mobilization but the Al mobilization was four times that of the control and the Al buffering became as important as the base-cation buffering. The buffering action in the high-N plots was mainly by Al mobilization which was nine times that of the control plots.

The Al proportion of the total cation mobilization increased from 26% in the control to 44% in the low-N plots and to 75% in the high-N plots where Mn buffering was as important as base-cation buffering (Figure 3). Interestingly, Mn was the element with the most marked increase in mobilization; 12 times in the high-N plots compared with the control. In spite of this considerable mobilization in the soil due to acidification, the great internal Mn circulation in the ecosystem seemed to be unaffected by the fertilizer treatment (Figure 2). Manganese, like K, is characterized by a remarkably efficient internal circulation in the forest ecosystem. Most of the Mn deposition to the ground was mediated by litterfall.

The pH-KCl of the soil tended to be slightly lower in the soils that had been N treated but the difference from the control was not statistically significant (Table I). In the AB horizon of both control and treatment plots, pH was lower in 1990 than in 1985. The exchangeable acidity and the concentration of exchangeable Al were also determined (for 1985 and 1990) but no treatment effects were found.

After 5 yr of treatment, in the spring of 1990, the KCl-extractable soil store (0 to 30 cm) of inorganic N in the low- and high-N plots was only 50 and 70%, respectively, higher than the control (Table VII). After these 5 yr, this excess N in the low- and high-N soils corresponded to only 70 and 17%, respectively, of the 1-yr net addition of N (input with BD plus FA minus output with SS).

5. Discussion

The primary effect of the NH_4NO_3 treatment was a substantial increase in the leaching of NO_3^- ions with the soil solution. This excess NO_3^- , emanating partly from the NO_3^- supplied with the fertilizer and partly from nitrification of the supplied NH_4^+ , leaches through the soil which has a very limited capacity of NO_3^- adsorption. If the percolating NO_3^- stems from nitrification, it gives a considerable H^+ contribution to the soil (see e.g. Reuss and Johnson, 1986). Nitrification is a process giving rise to a H^+ load which is twice that arising from NH_4^+ uptake (Gundersen and Rasmussen, 1990);

Nitrification and leaching of the excess NO_3^- were the main acid-producing mechanisms not only in the low-N treated soil but also in the control soil. It is now well established that nitrification occurs also in acidic forest soils (Kriebitzsch, 1978; Lang, 1986; Gundersen and Rasmussen, 1990). According to the calculation of the H^+ load, the soil acidification in the high-N treated plots was mainly due

to NH_4^+ accumulation in the soil (or root uptake), however. In these plots, the efficiency of the nitrifiers seemed to be insufficient to keep pace with the great addition of NH_4^+ , and the NO_3^- production was lower than in the low-N plots. This elevated NH_4^+ level may have had a toxic influence on the nitrifiers. Ectomycorrhizal fungi have been shown to be intoxicated by very high NH_4^+/K^+ ratios in a laboratory study (Boxman *et al.*, 1986). There are possibly alternative explanations of the low rate of nitrification in the high-N plots.

Results from the soil extraction (KCl) indicate that far from all the N added with the fertilizer to the high-N plots could be accounted for by accumulation in the 0 to 30 cm soil or by leaching. This sandy soil, low in clay content, certainly has a limited ability to accumulate N (Zöttl, 1990). The merely moderate increase in the soil store of inorganic N found 5 yr after the onset of the treatment indicates a situation approaching steady state rather than a continuous N accumulation. Moreover, lower total-N contents (Kjeldahl) in the A horizon (0 to 10 cm) of the low- and especially the high-N soils than in the control were actually found at Maglehem by Tyler *et al.* (1992). Enhanced N uptake and/or nitrate leaching may possibly have occurred as a quick response during the first few years after the onset of treatment.

Part of the N not recovered may be explained by increased biomass uptake of N. The slight but statistically significant decrease in the C:N ratio of the beech-leaf litter supports this probability (Tyler *et al.*, 1992). In a parallel study at the same site, Balsberg Pålsson (1992) likewise found the concentrations of both NO_3^- and total N in the beech leaves to be somewhat higher in the treated plots than in the control. Nitrogen fertilization is often but not always reported to increase the N concentration in leaves (Hüttel, 1990) and litter (Khanna and Ulrich, 1981). On the other hand, an enhanced N uptake into the tree biomass should promote tree growth but according to a stem-increment analysis after four years of treatment, tree growth was not influenced by the treatment (Tyler *et al.*, 1992). An increased N uptake in trees is also contradictory to the supposed N saturation of the ecosystem.

Compared with increased biomass uptake, denitrification could possibly form a quantitatively more important pathway for that amount of added N that was not recovered as leached through the soil or stored within it. Denitrification is known to be favoured by a high pH and shifts between aerobic and anaerobic conditions typical of sites with a fluctuating ground-water table (Haynes, 1986). Such conditions do not apply to the acidic and sandy soil of Maglehem. This does not exclude the presence of microhabitats suitable to denitrifiers, however (Tamm, 1991). Further denitrification is promoted by high concentrations of soil NO_3^- , a condition certainly prevailing in the treated soil. Remaining one of the possible pathways for the excess N in the high-N plots, denitrification would merit experimental investigation.

The difficulty in recovering all added N was not expected but the concordant results from all three replicates strengthen the observation.

The calculation of the H^+ load of the soil presupposes that the NH_4NO_3 was quantitatively dissolved and incorporated into the soil. Soon after supply, no traces of NH_4NO_3 granules were visible on the ground. It cannot be excluded that part of the NH_4NO_3 N might have been lost through ammonia volatilization (Tamm, 1991). (A saturated aqueous solution of the NH_4NO_3 had a pH of 5.5.) Since pK_a is as high as 9.3, NH_3 volatilization of the fertilizer supplied to this acidic soil is highly unlikely, however.

The most thorough going effect of the treatment was the intense acidification as revealed by the highly acidic soil solution. The acidification of the soil was not yet, after five and a half years of treatment, strong enough to be discernible by an extraction analysis, however.

The decrease in the soil-solution pH and the great NO_3^- leaching brought about base-cation depletion and increased Al release in the mineral soil. It is remarkable that the total cation mobilization increased four-fold in the high-N treatment and that 75% of the this mobilization was Al. The possibility that the increase in Al leaching could be an artifact due to Al leaching from the lysimeter material can be ruled out according to tests by Rasmussen *et al.* (1986) as well as in our laboratory. The results clearly indicate that the NH_4NO_3 treatment had driven the Maglehem soil from the base-cation buffer range into the Al buffer range (Ulrich, 1983). As to Mn, it is noteworthy that this heavy metal was mobilized in the high-N plots in greater quantity than were the base cations together.

Soil acidification is connected with increased solubilization of Al-containing solid phases (e.g. gibbsite or organically bound Al; Thomas and Hargrove, 1984; Mulder *et al.*, 1989a). The solubility of Al, Ca, Cd, K, Mg, Mn, Na, Rb, and Zn is very susceptible to changes in soil acidity, as demonstrated by experimental acidification of lysimeter soils (Bergkvist, 1986). A minor pH decrease within the measured pH range of the soils in the present study will raise the soil-solution concentration of these metals considerably (Bergkvist, 1987a; Tyler *et al.*, 1987). The origin of the solubilized base cations and Cd is cation exchange complexes (Berggren, 1992a). Since Cu is mainly mobilized in the form of complexes with organic matter, the increased Cu flux in the treated soils is probably more a result of increased leaching of organic matter than of acidification (Berggren, 1992b).

The strongly enhanced NO_3^- leaching in the treated soils indicates that the vast majority of the added N was not taken up by the roots. This feature is characteristic of N saturation (Nihlgård, 1985). In combination with a high bulk deposition of N, the lack of N interception by the canopy forms another sign of N saturation of this beech-forest soil. In less N-loaded environments, the N flux is commonly lower in throughfall than in bulk deposition (Helmisaari and Mälkönen, 1989; Parker, 1990).

Most of the leaching of NO_3^- and NH_4^+ commonly occurs during spring when plant uptake and soil biological activity are low (Mälkönen, 1990). This was so also in the untreated plots of Maglehem. In the treated plots, the N leaching was of about the same size during the foliated and the defoliated half year, however.

This is further evidence of N saturation.

This suggested incapability of the tree roots to take up the excess N in the treated plots may be related to the deterioration of the mycorrhizal efficiency that is indicated by the disappearance of fruitbodies of ectomycorrhizal macrofungi. This was in contrast to the litter-decomposing species where a drastic increase in the number of fruitbodies was found (Rühling and Tyler, 1991). The increased DOC concentrations in the soil solution also point towards enhancement of the organic-matter mineralization rate in the N-treated soil, but the Ca- and Mg-carbonate addition with the fertilizer may also partly increase the solubilization of organic matter. Further evidence for an enhanced mineralization rate is provided by the slightly but significantly decreased C content of the humus layer of both the low- and high-N plots (Tyler *et al.*, 1992). In contrast, Söderström *et al.*, (1983) found N amendments to decrease microbial activity in Podzol soils.

Fertilizer amendments are often reported to result in raised concentrations of various elements in throughfall soon after supply (Khanna and Ulrich, 1981; Parker, 1990). This was not so in the present experiment with repeated treatment, not even for NO_3^- or NH_4^+ . This lack of response in the throughfall is remarkable with regard to the substantial increase in soil-solution concentrations of most elements analysed.

Despite the location of this forest far away from pollution sources, N input with bulk deposition was as high as $1.8 \text{ g m}^{-2} \text{ yr}^{-1}$ which is at the same level as that found in another local study (Bergkvist *et al.*, 1987). Nitrogen deposition twice as high, 3.0 to $6.0 \text{ g m}^{-2} \text{ yr}^{-1}$, is reported from deciduous and coniferous forests in Denmark and central Europe where at the same time the N output was in the range 1.1 to $8.7 \text{ g m}^{-2} \text{ yr}^{-1}$ (van Breemen *et al.*, 1982, 1987; Bredemeier, 1985; Mulder *et al.*, 1987; Rasmussen, 1988). Compared to this, the N output in our control plots ($1.2 \text{ g m}^{-2} \text{ yr}^{-1}$) is rather low.

In input/output studies in a number of coniferous forests in northern and central Europe, it has been shown that NO_3^- -N outputs are less than $0.1 \text{ g N m}^{-2} \text{ yr}^{-1}$ where inputs are less than $1.0 \text{ g N m}^{-2} \text{ yr}^{-1}$ (Grennfelt and Hultberg, 1986). The NO_3^- -N output increases drastically as input exceeds $1.0 \text{ g N m}^{-2} \text{ yr}^{-1}$, and at inputs of $2.4 \text{ g N m}^{-2} \text{ yr}^{-1}$, the NO_3^- -N output is almost $2.0 \text{ g N m}^{-2} \text{ yr}^{-1}$. In the present study, NO_3^- -N output from the upper 30 cm increased drastically upon N treatment. This beech forest soil thus seems to fit into the pattern of increased N leaching from forest soils with increasing N inputs.

The soil-solution concentration of total Al is commonly reported to be in the range of 0.5 to 36 mg L^{-1} in studies of deciduous and coniferous forests on Cambisols and Podzols of Scandinavia, central Europe and the USA (Heinrichs and Mayer, 1977; Matzner and Hetsch, 1981; Lochman, 1983; David and Driscoll, 1984; Driscoll *et al.*, 1985; Rost-Siebert, 1985; Bergkvist 1987a, b; Mulder *et al.*, 1987, 1989b; Neitzke and Runge, 1987). Compared to this range, the Al concentrations in our control, low- and high-N soil solutions were in the lower, middle and middle/upper part, respectively.

The leaching of base cations, Al and NO_3^- in this acidic soil clearly indicates that the current load of N is, not only in the long run, above the sustainable, or 'critical' load of N (Nilsson and Grennfelt, 1988).

6. Conclusions

In conclusion, the repeated treatment during five and a half years of the Maglehem beech forest with granulated NH_4NO_3 gave the following main results:

- Soil NO_3^- leaching increased considerably with increasing N treatment.
- Nitrification, also occurring in the control soil, was greatly enhanced in the low-N plots. In the high-N plots, however, nitrification seemed to be inhibited, and the great NO_3^- leaching only slightly exceeded the amount of NO_3^- added with the treatment.
- The leaching of NO_3^- , whether arising from nitrification, NO_3^- addition or both, had a strongly acidifying influence.
- All N added to the high-N plots could not be recovered in soil or soil leachate. Denitrification or incorporation into below- or above-ground tree biomass could be tentative pathways of this excess N.
- The treatment brought about a shift from the base-cation to the aluminium buffer range.
- The soil acidification drastically increased the fluxes of cations, especially Al, Mn and Rb.
- The treatment promoted the mineralization of organic carbon.
- Beech litterfall C:N decreased with increasing N supply.
- Fluxes of elements, including N, in litterfall of throughfall were not influenced by the treatment.
- This forest is N saturated. A moderate further increase in the N deposition, as NO_3^- or NH_4^+ , would soon lead to a considerable increase in NO_3^- leaching and soil acidity as well as depletion of base cations and mobilization of Al and Mn.

Acknowledgments

Our gratitude is due to Bengt Nihlgård for providing soil samples, to Anders Jonshagen for field work, to Anita Balogh for chemical analyses and to Dan Berggren for constructive criticism of the manuscript. The study was financially supported by the Swedish Environmental Protection Agency.

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