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Flow of deposited inorganic N in two Gleysol-dominated mountain catchments traced with $^{15}NO_3^-$ and $^{15}NH_4^+$

ISABELLE PROVIDOLI[1], HARALD BUGMANN[2], ROLF SIEGWOLF[3], NINA BUCHMANN[4] and PATRICK SCHLEPPI[1,*]

*[1]Swiss Federal Institute for Forest, Snow and Landscape Research (WSL), CH-8903 Birmensdorf, Switzerland; [2]Forest Ecology, Swiss Fed. Inst. of Technology, ETH-Zentrum, CH-8092 Zurich, Switzerland; [3]Laboratory of Atmospheric Chemistry, Paul Scherrer Institute, CH-5232 Villigen-PSI, Switzerland; [4]Institute of Plant Sciences, Swiss Fed. Inst. of Technology, ETH Zentrum, CH-8092 Zurich, Switzerland; *Author for correspondence (e-mail: patrick.schleppi@wsl.ch; phone: +41-44-739-24-22; fax: +41-44-739-22-15)*

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Abstract. In two mountain ecosystems at the Alptal research site in central Switzerland, pulses of $^{15}NO_3$ and $^{15}NH_4$ were separately applied to trace deposited inorganic N. One forested and one litter meadow catchment, each approximately 1600 m^2, were delimited by trenches in the Gleysols. $K^{15}NO_3$ was applied weekly or fortnightly over one year with a backpack sprayer, thus labelling the atmospheric nitrate deposition. After the sampling and a one-year break, $^{15}NH_4Cl$ was applied as a second one-year pulse, followed by a second sampling campaign. Trees (needles, branches and bole wood), ground vegetation, litter layer and soil (LF, A and B horizon) were sampled at the end of each labelling period. Extractable inorganic N, microbial N, and immobilised soil N were analysed in the LF and A horizons. During the whole labelling period, the runoff water was sampled as well. Most of the added tracer remained in both ecosystems. More NO_3^- than NH_4^+ tracer was retained, especially in the forest. The highest recovery was in the soil, mainly in the organic horizon, and in the ground vegetation, especially in the mosses. Event-based runoff analyses showed an immediate response of $^{15}NO_3^-$ in runoff, with sharp ^{15}N peaks corresponding to discharge peaks. NO_3^- leaching showed a clear seasonal pattern, being highest in spring during snowmelt. The high capacity of N retention in these ecosystems leads to the assumption that deposited N accumulates in the soil organic matter, causing a progressive decline of its C:N ratio.

Introduction

During the last few decades, human activities have increased the production of reactive nitrogen through intensive agriculture and fossil fuel combustion (Galloway et al. 1995; Vitousek et al. 1997). The anthropogenic creation of reactive nitrogen increased between 1860 and 2000 from approximately 15 Tg N a^{-1} to 165 Tg N a^{-1}, with about five times more reactive nitrogen coming from losses resulting from food production than from energy production (Galloway et al. 2002). These various reactive nitrogen molecules cycle by

biochemical pathways and are easily distributed by hydrological and atmospheric transport processes over long distances and move from one environmental system to another (Galloway et al. 2002). This phenomenon is called the nitrogen cascade (Tietema et al. 1995; Galloway 1998). Within the nitrogen cascade, temperate forests and grasslands can be major reservoirs and a short to long-term sink for the reactive nitrogen. Given the N-limited nature of both ecosystem types there is a large potential for reactive nitrogen accumulation with a residence time of years to centuries causing a slow eutrophication (Galloway et al. 2003).

In several studies, the N deposition of forest ecosystems was manipulated to evaluate the complex interactions of processes in the N cycle and to measure N cycling within these ecosystems (Emmett et al. 1998; Tietema et al. 1998; Wright and Rasmussen 1998; Nadelhoffer et al. 1999a, b; Lamontagne et al. 2000). In the European research project NITREX, all sites showed an immediate response of nitrate leaching within the first year of either N addition or N exclusion (roof experiment). In these coniferous forests the responses of vegetation and soil were delayed (Gundersen and Rasmussen 1995; Moldan et al. 1995; Gundersen et al. 1998). The Alptal site in Switzerland was part of the NITREX project, representing N-limited mountain ecosystems. Similarly to the other NITREX sites, nitrate leaching increased at the Alptal site within a few weeks of N addition. According to Schleppi et al. (2004), NO_3^- is released at the Alptal site by three different mechanisms: (1) NO_3^- from precipitation bypassing the soil by preferential flow path, (2) flushing of older precipitation NO_3^- temporarily stored in the soil pores and (3) flushing of NO_3^- produced by nitrification. NO_3^- leaching occurred even though the trees were still slightly deficient in N (Schleppi et al. 1999b, 2004; Hagedorn et al. 2001). However, over 60% of the added $^{15}NH_4{}^{15}NO_3$ tracer was recovered in the soil pool, and only approximately 10% was leached as NO_3^- (Schleppi et al. 1999a). Despite considerable information on various aspects of the nitrogen cycling on the Alptal site, a lot about the nitrogen accumulation in the ecosystems is still unclear. To understand the mechanisms influencing the nitrogen flow through the ecosystems, there is need for a deeper understanding of the single processes and flows, particularly in the soil pool as well as in the leaching from the catchment.

In this study, we applied, in contrast to the study of Schleppi et al. (1999a), the NH_4^+ and the NO_3^- tracers separately, on a forest and a litter meadow catchment (each 1600 m^2) to trace deposited inorganic N. Therefore, we were able (1) to quantify after one year the ^{15}N recovery of both N forms through the ecosystem among trees, understory vegetation, litter layer, soil, roots; and (2) to follow the flow of $^{15}NO_3^-$ in runoff (in part event-based) continuously over one year. This way, we were able (3) to infer about the accumulation and absorption capacity of nitrogen in both ecosystems.

Material and methods

Study area

The research site is located in the Alptal valley, on the northern edge of the Alps of central Switzerland (47°03′ N, 8°43′ E), at 1200 m asl. It lies within the Erlenbach headwater catchment, which covers 0.7 km^2 and consists of 40% naturally regenerating forest and 60% litter meadow, both never artificially fertilised. The *Picea abies* stand has a relatively low leaf area index of 3.8 (Schleppi et al. 1999b) with trees up to 250 years old. Bulk atmospheric deposition of inorganic N is 12 kg ha^{-1} a^{-1}, with equal contributions of NO_3^- and NH_4^+ (Schleppi et al. 1999a). The climate is cool and wet, with a mean annual temperature of 6 °C and a mean annual precipitation of 2300 mm, reaching a maximum in June (270 mm) and a minimum in October (135 mm). Usually, soils are covered with snow from mid-November to April and the vegetation period lasts from June to September. The parent rock material is Flysch, which is composed of sedimentary conglomerates with clay-rich shists, and the major soil types are very heavy Gleysols of low permeability with the water table close to the surface throughout the year (Hagedorn et al. 1999). Slope is about 20% with a west aspect. Generally, the soil profile consists of a LF, an A and a B horizon. In the forest, vegetation and soil types form a mosaic pattern closely related to microtopography. Trees grow on mounds, were the water table is at a depth below 40 cm and umbric Gleysols are abundant with raw humus (LFH), Ah and oxidised or partly oxidised Bg or Br horizons. On mounds, dominating plant species are *Picea abies*, with 15% *Abies alba* and *Vaccinium myrtillus*. In depressions, the water table frequently reaches the surface, leading to mollic Gleysols with a thin LF horizon, an anmoor topsoil (Aa) and an almost permanently reduced Bg or Br horizon. The waterlogged depressions are too wet for tree growth, and ground vegetation is dominated by *Caltha palustris*, *Petasites albus, Poa trivialis* and *Carex ferruginea* (Muller 1997). The meadow was formerly used for litter production, i.e. for animal bedding. In contrast to the forest, it has no distinct microtopography and the soil is characterised by an anmoor topsoil (mollic Gleysol) , like the depressions in the forest, and an almost permanently reduced Bg or Br horizon. The vegetation consists mainly of a small sedge meadows (*Molinietum*), with a few small trees (*Picea abies* and *Alnus incana*). Since a few decades, societal and economic changes in forestry and agriculture have resulted in different land use patterns and large parts of the litter meadows lie fallow since 20 years.

Experimental design

One forested and one meadow catchment, each approximately 1600 m^2, were delimited by trenches (Figure 1). Due to the impermeable gleyic sub-soil, the

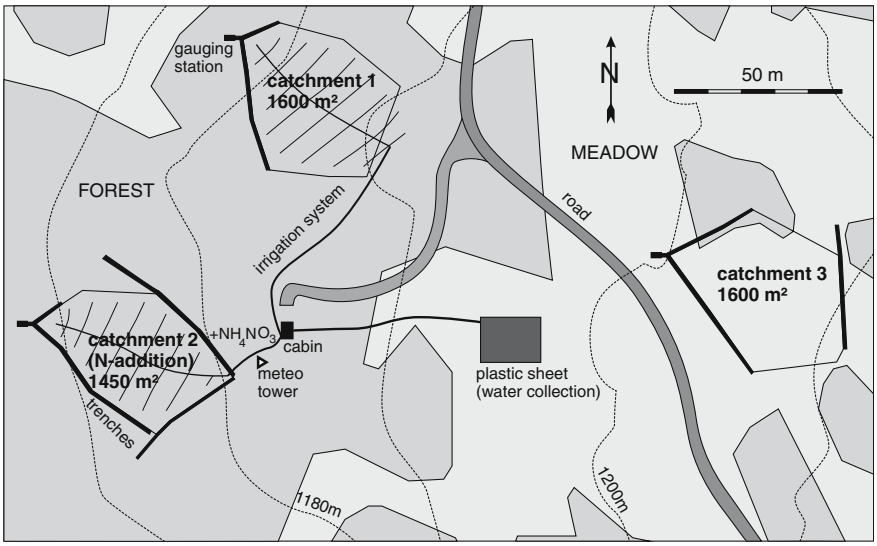

Figure 1. Experimental set-up of the forest (1) and meadow (3) catchment in Alptal, Switzerland.

water is believed not to infiltrate below the depth of the trenches (80 cm), and thus the water budget is approximately balanced (Schleppi et al. 1998). $K^{15}NO_3$, resp. $^{15}NH_4Cl$ tracers (both 99 atom% ^{15}N), dissolved in deionised water (10 l per catchment and application), were applied as two successive labellings with a backpack sprayer on each catchments directly above the ground vegetation to label the atmospheric deposition of inorganic N. The application occurred in minimal but frequent doses during 1 year, weekly during the vegetation period and fortnightly in winter. The seasonality of the ambient deposition rates was thereby mimicked approximately. The first labelling ($K^{15}NO_3$) started in summer 2000, and the second labelling ($^{15}NH_4Cl$) started in summer 2002 after a one-year chase period. The second labelling was stronger to mask the residual effects of the application of the $K^{15}NO_3$ 2 years earlier. Due to the high isotopic enrichment, the amount of tracer applied was only 0.17 mmol m^{-2} $K^{15}NO_3$, resp. 0.7 mmol m^{-2} $^{15}NH_4Cl$, so that the associated fertilisation effect was negligible compared to ambient deposition. The catchments were equipped for proportional sampling of runoff water. Runoff was continuously measured at a gauging station with a V-notch weir (Schleppi et al. 1998). Immediately after the first application of the NO_3^- tracer, the runoff of the first two rain events was sampled with a high temporal resolution (30 runoff samples in five days). Based on previous research (Schleppi et al. 1999a), 10 to 20% of the labelled nitrate was expected to be leached by preferential flow paths already during the first event. After this event-based analysis, the flux of ^{15}N as nitrate in runoff water was measured throughout the experiment, using samples taken weekly and pooled proportionally over

2 weeks, 1 month and later over 3 month periods. Additionally, soil, trees (needles, branches and bole wood), and ground vegetation were sampled in September 2001, three months after the end of the first, and in 2003, 3 months after the end of the second labelling period. Soil samples were taken in a grid of 8×8 m with a soil corer (5 cm inner diameter, 25 cm depth) reaching into the B horizon. These soil cores were immediately put on ice in the field, transported to the laboratory and processed within one day. Due to the microtopography and the mosaic pattern of the soil, the soil cores were grouped according to the soil types (mollic or umbric Gleysol) to obtain sufficient soil for the analysis. Out of 21 cores in the meadow, five soil groups were formed. Each group formed one composite sample of four or five single cores. Similarly, in the forest, 24 cores were split up into four soil groups of six single soil cores. In this case, each group was further split up into two subgroups with three single soil cores from the upper or the lower part of the catchment to form one composite sample. The soil cores consisted of litter layer, LF horizon (approx. 5–0 cm), A horizon (approx. 0–5 cm) and B horizon (5–20 cm). The above-ground vegetation was sampled separately. For this purpose, the ground vegetation was clipped at a place (18.5 cm $\times$ 20 cm) close to the soil sampling points and pooled like the soil samples into composite samples. Each vegetation sample was kept refrigerated and processed in the laboratory within one day. The trees were sampled in winter 2000/2001 and in winter 2002/2003. Sampling was done as described previously in Schleppi et al. (1999a).

Laboratory analyses

The soil samples were separated into surface litter and into the soil horizons LF, A and B. From the LF and the A horizon material, the roots were picked out by hand with tweezers, rinsed free of soil with deionised water and separated into the diameter classes of > 4 mm and < 4 mm. Large roots (> 15 mm diameter) were removed. At least 200 g field-moist soil per sample was freed from roots (work time $\sim$4 h) and used for total N and ^{15}N analyses as well as for extractions. Extractable inorganic N and ^{15}N (NH_4^+-N, NO_3^--N) and total extractable N and ^{15}N were determined by adding 40 g of soil material from the LF horizon or 60 g from the A horizon to 160 ml of 0.5 M K_2SO_4, then shaking the mixture for 1.5 h and filtering it through folded filters ($790^{1/2}$, diameter 185 mm, Schleicher & Schuell, Dassel, Germany) into PET bottles. The extract was stored in the freezer until further processing. As a next step, half of the extract was lyophilised using a freeze dryer (Christ, BETA 1–8, Osterode am Harz, Germany) to measure total extractable N and ^{15}N using mass spectrometry. With the other half of the extract, extractable $^{15}NO_3^-$ and $^{15}NH_4^+$ concentrations were analysed using the diffusion method (adapted from Downs et al. 1999). About 80 ml of the extract were put into 100 ml PE bottles, together with a glass microfibre filter (GF/F 25 mm, Whatman, Maidstone, England; 5 mm $\times$ 12 mm, calcinated in a muffle furnace (Naber,

Type No. 7, Zurich, Switzerland) for 6 h at 450 °C), wetted with 30 μl H_2SO_4 2 M and wrapped in PTFE band. Then 1.5 mg l^{-1} MgO were added and the bottle was tightly closed. NH_4^+ was thus converted to NH_3, diffused through the PTFE band and finally captured on the microfibre filter. The bottles were shaken (40 min^{-1}) for a week, whereupon the filter was removed, dried in an evacuated desiccator in the presence of concentrated sulphuric acid, and then unpacked from the PTFE band. The filter was stored in a glass vial and packed in a silver cup prior to mass spectrometric analysis. The remaining extract was further processed by adding a new microfibre filter and 0.5 g Devarda's alloy. Thereby, NO_3^- was reduced to NH_4^+, and the same chemical procedure was repeated as with NH_4^+.

N and ^{15}N in the microbial biomass were determined by chloroform fumigation and extraction (Brookes et al. 1985). Soil material of 20 g from the LF horizon or 30 g from the A horizon were fumigated with $CHCl_3$ in an evacuated desiccator in the dark for 24 h and afterwards extracted with 80 ml of 0.5 M K_2SO_4. The extract was stored in the freezer and then lyophilised entirely to determine the total N concentration and the $^{15}N/^{14}N$ isotope ratio. The remaining bulk soil and all other solid samples (above and below-ground biomass, litter) were dried to constant weight at 65 °C and the dry matter content was calculated. All samples were ground, and the total N and ^{15}N concentrations were determined by an elemental analyser coupled to an isotope-ratio mass spectrometer (Delta S, Finnigan, Bremen, Germany).

The soil samples were ground with a vibratory mill (0.25 mm), the above and below-ground biomass materials with a centrifuge grinder (0.5 mm), and the litter layer with a coffee grinder. The vegetation was first separated into five groups: monocotyledons, dicotyledon herbs, dicotyledon shrubs and mosses. The C:N ratio of all three soil horizons was measured with a C + N analyser (NC 2500, CE instruments Thermoquest, Milano, Italy).

Calculation of recovery rates

The N pools of the ecosystem were calculated from dry masses and N concentrations. The isotopic values are presented in the δ notation (Equation 1):

$$\delta^{15}N = R_{sample}/R_{standard} - 1 \quad (1)$$

where R is the molar fraction $^{15}N/^{14}N$. $\delta^{15}N$ is usually expressed in ‰ (times 1000). The atmospheric N_2 is used as a standard with $R_{standard} = 0.0036765$. The molar fraction R_{sample} is thus calculated as (Equation 2):

$$R_{sample} = \left(\delta^{15}N + 1\right) \cdot R_{standard} = {}^{15}N/{}^{14}N \quad (2)$$

The fractional abundance of ^{15}N in the sample is defined by (Equation 3):

$$F_{sample} = R_{sample}/\left(R_{sample} + 1\right) = {}^{15}N/\left({}^{15}N + {}^{14}N\right) \quad (3)$$

The tracer fraction X_{sample} defined as the molar ratio of tracer N to total N content of a pool can be calculated as (Equation 4):

$$X_{\text{sample}} = \left(F_{\text{sample}} - F_{\text{reference}}\right)/(F_{\text{tracer}} - F_{\text{reference}}) \quad (4)$$

where $F_{\text{reference}}$ is the fractional abundance of ^{15}N in the pre- or non-labelled sample and F_{tracer} is the fractional abundance of the applied tracer (atom%). For the NO_3^- tracer experiment, the control (nonlabelled) samples were used as $F_{\text{reference}}$; the F_{sample} from the NO_3^- tracer (prelabelled and sampled in autumn 2001) was then used as $F_{\text{reference}}$ for the NH_4^+ tracer (sampled in autumn 2003). By doing this subtraction, we have to keep in mind that the ^{15}N recovery of the NO_3^- tracer is not constant over this time period and therefore it is just an approximation of the actual values. Studies by Providoli et al. (submitted) showed that changes were relatively small from 1 month onwards.

The tracer recovery in an ecosystem pool (Z_{pool}) was calculated as a fraction of added tracer (Equation 5):

$$Z_{\text{pool}} = X_{\text{sample}} \cdot N_{\text{pool}}/N_{\text{tracer}} \quad (5)$$

where N_{pool} is mass of the N pool [mol] and N_{tracer} is mass of applied tracer [mol], both expressed on the same basis (per plot or per unit area). Z_{pool} is usually expressed in % (times 100).

For soil microbial N, the recovered ^{15}N was calculated as the difference between the recovered ^{15}N in the extract of fumigated soil and that in the non-fumigated soil (no correction factor was used for extraction efficiency). The extractable $DO^{15}N$ was calculated as total extractable ^{15}N minus extractable $^{15}NO_3^-$-N and $^{15}NH_4^+$-N. The recovery in soil minus the recoveries in soil microbial ^{15}N and extractable ^{15}N was considered as immobilised soil N (ISN). According to Schleppi et al. (1998), the water budget of the small catchments is not influenced by unknown water ways. Therefore, quantitative N budgets were calculated from measurements of deposition inputs and leaching outputs. The tracer concentration in runoff [$\Delta\gamma^{15}N$] was calculated as $\delta^{15}N$ of the sampled runoff minus $\delta^{15}N$ of the runoff before the tracer application. Background levels of denitrification were estimated based on previous studies on the manipulated forest catchment on this site (Mohn et al. 2000). These measurements were used to assess denitrification on the control catchments. For the meadow, measurements on the anmoor topsoil in the forest were used for an estimation. According to Hagedorn et al. (1999), flow paths in the soil had the highest NO_3^- supply and were the locations with the highest denitrification activity. Therefore, we assumed that the denitrification had the same $\delta^{15}N$ as the runoff, and tracer recovery was estimated.

Due to logistical constraints, the experiment within the forest and the meadow was not replicated. Therefore, the differences between these ecosystems could not be tested statistically. Comparisons between the forest and the meadow were thus made only descriptively.

Results

Event-based runoff for NO_3^- tracer

The first two rain events after the first NO_3^- tracer application were sampled with a high temporal resolution for both catchments starting on July 3 and July 7, 2000 (Figures 2 and 3). During the first event, it rained 31 mm and in the second 42 mm. Discharge responded rapidly to storm events and the highest runoff peak reached in both rain events, in the forest as well as in the meadow, 3.4 mm h^{-1}. In the forest, the NO_3^- concentration was highest during the first discharge peak in both rain events. By the end of the runoff peak, the concentration declined to values close to those measured before the rain. In the meadow, the NO_3^- concentration in runoff was lower than in the forest and the peaks were less pronounced (Figures 2 and 3).

In the forest, $\Delta\gamma^{15}$N enrichment was highest at the first discharge peak on July 3 and had a second sharp peak at the second discharge peak on July 3

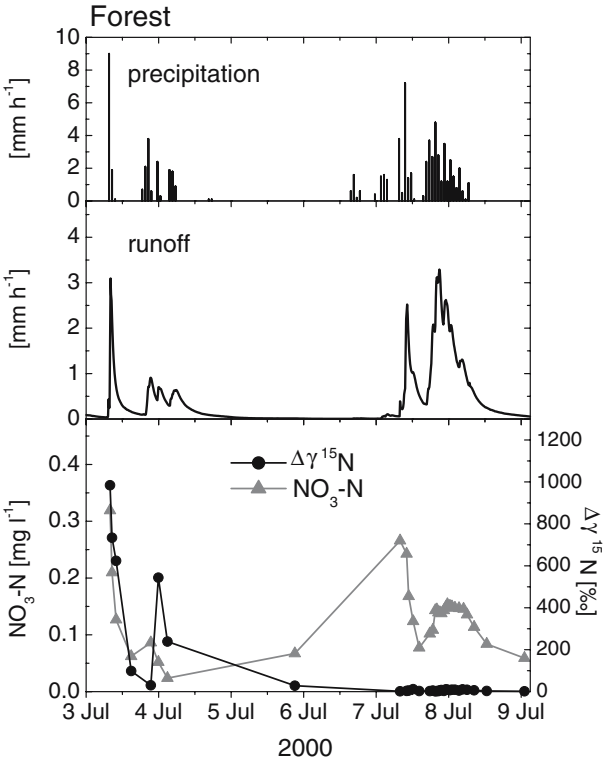

Figure 2. Runoff from the experimental forest catchment, its concentration in NO_3^- and $\Delta\gamma^{15}$N during two rainfall events in July 2000.

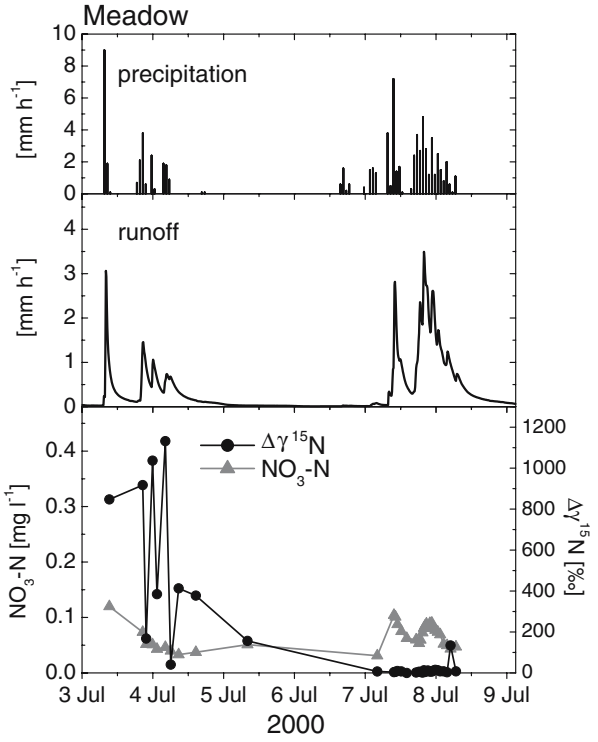

Figure 3. Runoff from the experimental meadow catchment, its concentration in NO_3^- and $\Delta\gamma^{15}N$ during two rainfall events in July 2000.

(Figure 2). In the second rain event, on July 7, the $\Delta\gamma^{15}N$ remained close to zero. In the meadow, in the first rain event, $\Delta\gamma^{15}N$ showed sharp peaks coinciding with the discharge peaks (Figure 3). As in the forest, the $\Delta\gamma^{15}N$ was at a low level for the second rain event, showing only one peak at the end of the rain event. For both ecosystems, the $\Delta\gamma^{15}N$ was dynamic, whereas the NO_3^- concentration was less dynamic than $\Delta\gamma^{15}N$.

Weekly runoff for NO_3^- and NH_4^+ tracers

Over the 1 year of tracer application, runoff was sampled for both tracers. From 9 months on, the NO_3^- leaching for the $^{15}NO_3$ labelling was higher in the meadow than in the forest (Figure 4). For both ecosystems, a strong NO_3^- leaching started at the beginning of snowmelt in March 2001. The $^{15}NO_3^-$ leaching followed the same seasonal pattern. Especially in winter 2001 and in spring 2001, ^{15}N in runoff was two and four times higher in the meadow than in the forest, respectively. After the cessation of the $^{15}NO_3^-$ labelling in June 2001,

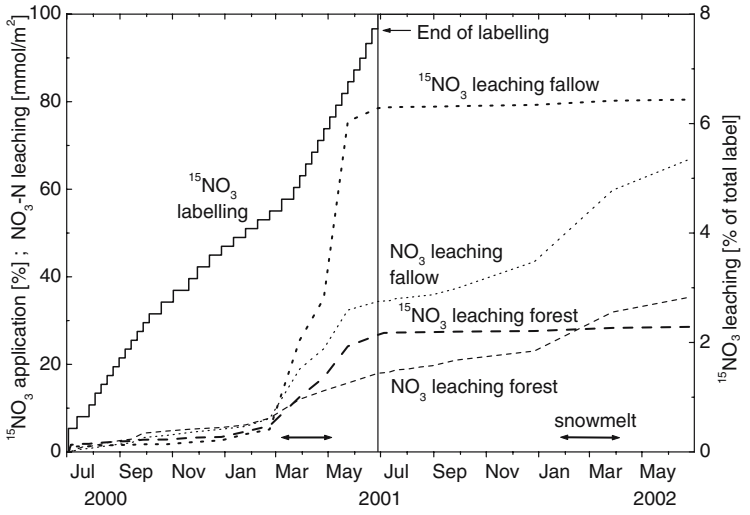

Figure 4. NO_3^- tracer addition and nitrate concentration in discharge-proportional runoff samples from the experimental forest and meadow catchment. $^{15}NO_3^-$ of total label and NO_3^- leaching concentrations from July 2000 to May 2002.

NO_3^- leaching was still going on and had again the highest values at snowmelt in 2002, which started a bit earlier than the year before, i.e. in January. However, the $^{15}NO_3^-$ leaching stopped in both ecosystems after the cessation of the labelling.

For the NH_4^+ tracer, the total ^{15}N flow in runoff was less than 0.2% in the forest over 1 year (June 2002–June 2003) (Table 3). Due to the low NH_4^+ leaching an analogous figure to the Figure 3 for the NO_3^- tracer is missing and we represent the results in Table 3. The recovery in the meadow was a slightly higher, reaching 0.6%. The highest ^{15}N leaching was in spring 2003 during snowmelt.

Total recovery of ^{15}N for the NO_3^- tracer in 2001

Total ^{15}N recovery was higher in the forest (81%) than in the meadow (67%) (Table 1, Figure 5). In the forest, 25% of the tracer was recovered in the above-ground biomass. The highest ^{15}N sink was the moss layer (14%), and 5% were recovered in the herbaceous ground vegetation. Both, the tracer fraction and the pool size were high for the mosses, whereas the tracer fraction as well as pool size were lower for the monocotyledons, dicotyledon herbs and dicotyledon shrubs. The trees recovered 6% of the tracer.

In the meadow, the above-ground biomass (including a few trees) recovered 13%, whereas the monocotyledons had the highest recovery (9.6%). The tracer fraction was higher for the dicotyledon herbs than for the monocotyledons, but

Table 1. Partitioning of the added ^{15}N labelled nitrogen for the NO_3^- tracer in trees, ground vegetation, litter layer, soil horizons, roots and runoff in forest and meadow, as means.

Forest: NO_3^- tracer					Tracer	
Pool		Pool size [kg/m^2]	N conc. [g/kg]	N pool [g/m^2]	Fraction [μmol/mol]	Recovery [%]
Vegetation (above-ground)	Trees	18.1	2.2	39.4	2.3	5.6
	Mosses	0.06	17.8	1.1	321	13.6
	Monocotyledons	0.03	14.6	0.4	100	1.5
	Dicotyledon herbs	0.02	15.1	0.3	122	1.6
	Dicotyledon shrubs	0.03	11.1	0.3	87	1.1
	Other species	0.04	15.2	0.6	53	1.2
Litter layer	Litter	0.5	13.4	6.8	53	14.4
Roots	LF roots	0.8	14.2	6.5	23	5.9
	A roots	0.3	15.7	2.9	10	1.1
Soil	LF (5–0 cm)	4.9	12.2	59.7	5	11.2
	A (0–5 cm)	19.9	8.5	169.4	0	2.8
	B (5–20 cm)	68.0	2.9	195.2	2	16.8
Runoff		3276	0.00015	0.496	173	2.3
Denitrification[a]						1.7
Recovery						80.8

Table 1. Continued.

Forest: NO_3^- tracer					Tracer	
Pool		Pool size [kg/m^2]	N conc. [g/kg]	N pool [g/m^2]	Fraction [μmol/mol]	Recovery [%]
Meadow: NO_3^- tracer Pool						
Vegetation (above-ground)	Trees[b]	1.3	3.1	4.1	3.2	0.4
	Mosses	0.003	23.6	0.1	181	0.4
	Monocotyledons	0.26	11.0	2.9	105	9.6
	Dicotyledon herbs	0.04	10.7	0.4	185	2.3
Litter layer	Litter	0.2	15.4	3.7	92	10.9
Roots	LF roots	0.5	11.7	4.7	30	4.5
	A roots	0.5	17.5	4.9	25	3.9
Soil	LF (5–0 cm)	8.8	10.4	91.7	7	19.1
	A (0–5 cm)	37.0	7.3	270.2	1	7.1
	B (5–20 cm)	55.5	5.5	303.6	0	0.0
Runoff		4442	0.0002	0.9	297	6.4
Denitrification[a]						2.7
Recovery						67.1

Pool sizes as dry matter (except runoff), N concentrations, N pools, tracer fractions and tracer recoveries.

[a]Calculation mentioned in material and methods.

[b]Small trees growing in the fallow meadow.

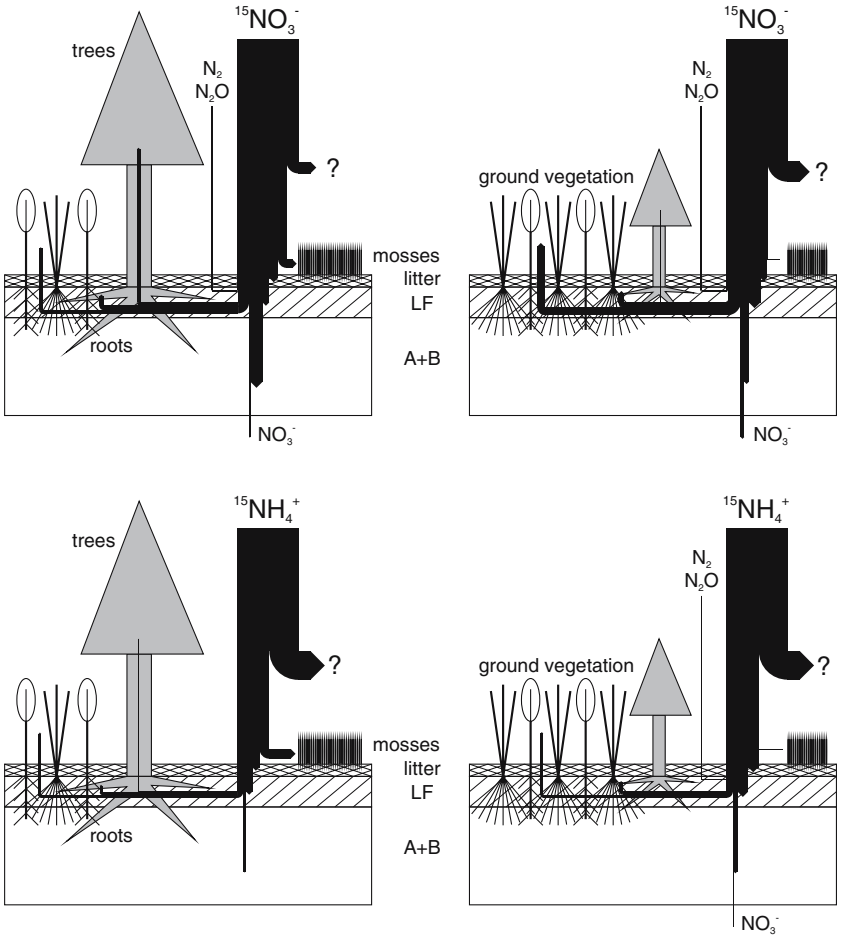

Figure 5. Flow and fate of the NO_3^- and the NH_4^+ tracers in the forest (left) and in the meadow (right) catchments. From left to right: ground vegetation, roots, trees, denitrification, nitrate leaching, soil mineral horizons (A + B), organic horizon (LF), litter layer, mosses, unrecovered. Tracer recoveries $< 0.2\%$ are not displayed (see Table 1 and 3).

the difference in pool size explained the higher recovery for the monocotyledons. Almost no mosses were present.

The recovery in the litter layer was higher for the forest than for the meadow. The tracer fraction of this layer was higher for the meadow, but the litter pool size in the forest was two and a half times larger leading therefore to a higher recovery.

In the LF and A horizons, the recovery in the meadow was higher, whereas the recovery in the B horizon was higher in the forest. The soil horizons were further partitioned in ^{15}N compartments (Figure 6).

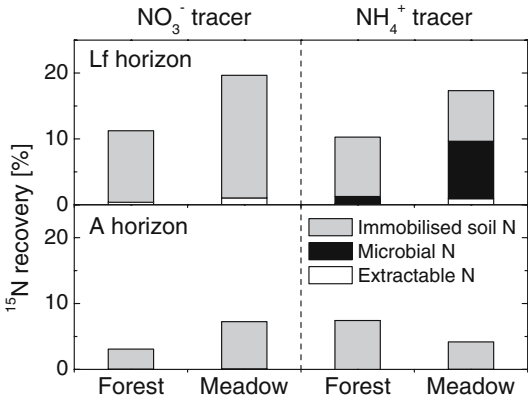

Figure 6. Total ^{15}N recovery of added NO_3^- and NH_4^+ tracers in the soil pool fractions: immobilised soil N, microbial N, and extractable N in LF and A horizons in the experimental forest and meadow catchment in 2001 and in 2003. Extractable N is mainly DON with a negligible amount of extractable NO_3^- and NH_4^+.

Total extractable N and microbial N were almost not detectable in the LF and the A horizons for both ecosystems. The highest recovery within the soil was in immobilised soil N for both horizons. For all three soil horizons (LF, A and B), the C and N concentrations were measured in both ecosystems (Table 2).

The forest had a higher C:N ratio than the meadow, whereas especially the C concentrations were much higher in the forest.

For both ecosystems, the recovery in the roots was higher for the LF roots than for the A roots. The recovery in runoff was higher for the meadow (6%) than for the forest (2%), and some tracer must have been lost due to denitrification.

Total recovery of ^{15}N for the NH_4^+ tracer in 2003

Total ^{15}N recovery was similar for the forest and the meadow (53%) (Table 3, Figure 5). In the forest, 22% of the tracer was recovered in the above-ground

Table 2. N and C concentration and C:N ratio for the soil horizons LF, A and B for the forest and the meadow in 2001, as means.

Pool		N conc. [%]	C conc. [%]	C:N
Forest	Lf (5–0 cm)	1.42	30.35	20.96
	A (0–5 cm)	1.01	17.98	17.10
	B (5–20 cm)	0.33	5.84	17.69
Meadow	Lf (5–0 cm)	1.11	18.18	16.05
	A (0–5 cm)	0.93	12.46	13.39
	B (5–20 cm)	0.65	9.87	15.09

Table 3. Partitioning of the added ^{15}N labelled nitrogen for the NH_4^+ tracer in trees, ground vegetation, litter layer, soil horizons, roots and runoff in forest and meadow, as means.

Forest: NO_3^- tracer					Tracer	
Pool		Pool size [kg/m^2]	N conc. [g/kg]	N pool [g/m^2]	Fraction [μmol/mol]	Recovery [%]
Vegetation (above-ground)	Trees	18.1	2.2	39.4	2.2	1.0
	Mosses	0.12	13.9	1.7	915	16.1
	Monocotyledons	0.02	15.6	0.3	189	0.6
	Dicotyledon herbs	0.03	16.4	0.6	279	1.6
	Dicotyledon shrubs	0.03	11.5	0.3	352	1.2
	Other species	0.04	14.5	0.6	231	1.3
Litter layer	Litter	0.4	13.7	6.1	186	11.6
Roots	LF roots	0.6	16.5	5.3	75	4.1
	A roots	0.5	14.4	3.6	14	0.5
Soil	LF (5–0 cm)	6.1	11.7	71.0	14	10.2
	A (0–5 cm)	19.5	8.6	168.7	3	4.3
	B (5–20 cm)	61.2	3.6	219.2	0	0.0
Runoff		2140	0.0001	0.2	66	0.2
Denitrification[a]						0.0
Recovery						52.8

Table 3. Continued.

Forest: NO_3^- tracer					Tracer	
Pool		Pool size [kg/m^2]	N conc. [g/kg]	N pool [g/m^2]	Fraction [μmol/mol]	Recovery [%]
Meadow: NH_4^+ tracer Pool						
Vegetation (above-ground)	Trees[b]	1.3	3.1	4.1	3.2	0.1
	Mosses	0.00	20.21	0.06	879	0.5
	Monocotyledons	0.39	12.12	4.67	85	3.2
	Dicotyledon herbs	0.06	11.69	0.66	274	1.5
Litter layer	Litter	0.4	13.3	4.9	413	16.7
Roots	LF roots	0.7	24.6	7.9	76	4.9
	A roots	0.2	14.0	1.9	22	0.3
Soil	LF (5–0 cm)	10.0	10.5	105.3	20	17.3
	A (0–5 cm)	22.0	8.2	181.1	2	2.3
	B (5–20 cm)	52.3	5.7	298.7	2	4.8
Runoff		3266	0.00017	0.571	129	0.6
Denitrification[a]						0.7
Recovery						53.1

Pool sizes as dry matter (except runoff), N concentrations, N pools, tracer fractions and tracer recoveries.
[a]Calculation mentioned in material and methods.
[b]Small trees growing in the fallow meadow.

biomass. Again, the highest ^{15}N sink was the moss layer (16%), and the herbaceous ground vegetation recovered 5%. The trees had a recovery of 1%. In the meadow, the above-ground biomass (including a few trees) recovered 5.3%. The ground vegetation recovered 5.2%, and the monocotyledons had a higher recovery than the dicotyledon herbs. The tracer fraction was higher for the dicotyledon herbs than for the monocotyledons, but the monocotyledons had a six and a half times higher pool size, thus leading to higher ^{15}N recovery.

The recovery in the litter layer and in the LF horizon was higher for the meadow. In the A horizon, the recovery was slightly higher in the forest, whereas in the B horizon the recovery was again higher for the meadow. The soil horizons were further partitioned in ^{15}N compartments (Figure 6). The total extractable ^{15}N was almost not detectable in the LF and the A horizon. The microbial ^{15}N could be determined for the LF horizon, and resulted in higher values in the meadow (9%) than in the forest (1%). In the forest, the highest recovery for both horizons was in immobilised soil N. In the meadow, microbial ^{15}N had the highest recovery in the LF horizon, whereas in the A horizon immobilised soil N had the highest recovery. For both ecosystems, recovery in the roots was higher for the LF horizon than for the A horizon, and the recovery in runoff was slightly higher for the meadow (0.6%) than for the forest (0.2%). An almost negligible amount of tracer was lost due to denitrification.

Discussion

Total ^{15}N recovery

The results showed that both ecosystems generally had a high capacity to retain more than 50% of the N added as tracer. More NO_3^- than NH_4^+ tracer was retained in general, especially in the forest. Similar to other tracer studies (Buchmann et al. 1996; Nadelhoffer et al. 1999b; Gebauer et al. 2000; Lamontagne et al. 2000; Zak et al. 2004) not all of the added tracer was recovered. Total recoveries are often between 60 and 80%, as shown in an ongoing synthesis of ^{15}N applications to natural ecosystems in the temperate or arctic zones (P. Templer, Univ. of California, Berkley & M. Mack, Univ. of Florida, Gainesville, personal communication). Denitrification could be a significant loss only if its tracer fraction was much higher than in NO_3^- leaching, which is unlikely. Volatilisation of NH_3 may partly explain the lower recovery of the NH_4^+ tracer compared to NO_3^-. According to Lamontagne et al. (2000), woody detritus could be an important ^{15}N sink due to their potential to immobilise N during decomposition. This pool was not measured in the present study. After a $^{15}NH_4{}^{15}NO_3$ application (Schleppi et al. 1999a), its ^{15}N concentration was approximately the same as in living wood. The amount of woody debris, however, is much smaller in this forest with selective harvests that in a natural system; therefore, it cannot be expected to receive more than 1% of the applied tracer. Some NO_3^- can be leached into soil

horizons below the sampled depth. This, however, is practically limited to the mounds in the forest, because in other places the water table is high, inducing a lateral flow towards the runoff rather than a deep infiltration. Deeper soil horizons thus probably contain less tracer than the sampled part of the gleyic B horizon. Taken together, and along with the natural variability of soil and vegetation, these different factors may sum up to the missing ^{15}N, even if no single one appears to explain large losses.

Event-based and annual flow of $^{15}NO_3^-$ in runoff

The event-based runoff analyses shortly after the NO_3^- tracer application showed an immediate response of ^{15}N in runoff for both ecosystems (Figures 2 and 3). Sharp $\Delta\gamma^{15}N$ peaks and lower NO_3^- peaks were detected, indicating an immediate leaching of the added N tracer. Creed and Band (1998) showed that NO_3^- peaks indicate flushing from the soil layers, when they become saturated with water. Likewise, Schleppi et al. (2004) demonstrated on the Alptal site that NO_3^- peaks usually correspond to the rising water table during rain events, and they include 'old' NO_3^-, which was already in the soil before the rain event started, as well as 'new' labelled $^{15}NO_3$. According to Schleppi et al. (2004), the flushed NO_3^- in runoff was mainly from recently deposited N. Hagedorn et al. (2001) showed at the same site that a large proportion of the runoff had limited contact with the subsoil and originated from the precipitation and the topsoil.

In the meadow, the flushing of the ^{15}N was more dynamic than in the forest. This could be due to stronger water-table dynamics, which led to stronger flushing dynamics, or it could be an artefact of the sampling frequency, which was unevenly distributed for the two catchments. However, the total NO_3^- concentration was more dynamic in the forest than in the meadow.

The annual pattern of NO_3^- in runoff showed a clear seasonal pattern (Figure 4). For both ecosystems, the highest NO_3^- leaching occurred in late winter and in spring at snowmelt events. After the cessation of the NO_3^- tracer application the total NO_3^- leaching was still going on and had the same seasonal pattern as in the first year, showing high leaching at snowmelt events. However, the $^{15}NO_3^-$ leaching stopped. This emphasizes that the applied $^{15}NO_3^-$ was either leached out of the system or immobilised directly in the soil or the biomass within a few weeks or months as shown by Providoli et al. (submitted) at the same site. Therefore, the delayed leaching of $^{15}NO_3^-$ was very minute. In the meadow, $^{15}NO_3^-$ leaching was much higher than in the forest. This could be explained by the higher N retention in the forest by trees, which are missing in the meadow, by the higher N retention of the ground vegetation in the forest, and by the much higher C content in the forest, which absorbs more N by direct immobilisation.

In the forest, tracer recovery in runoff was much lower compared to the N manipulation study by Schleppi et al. (1999a) on the same site. Schleppi et al.

(1999a) recovered 10% of the $^{15}NH_4{}^{15}NO_3$ tracer in runoff, whereas we recovered in the forest only about 1% for both tracers. This large difference can be explained by the different tracer application. In the study of Schleppi et al. (1999a), the tracer was applied during rain events by rotating sprinklers, thus mimicking wet deposition, and being flushed immediately by the preferential flow paths. However, in our study, tracer application was performed not only during but also after rain events with a backpack sprayer. Thus we were mimicking wet as well as dry deposition. The tracer applied as dry deposition does not enter the preferential flow paths in the soil immediately, and is therefore stored in the system for a longer time.

^{15}N recovery in pools

The sink strength of the above-ground vegetation, which consisted mostly of perennial species was higher in the forest than in the meadow for both tracers. This was especially due to the much higher moss biomass in the forest. As already described by Oechel and Van Cleve (1986) and DeLuca et al. (2002) and confirmed for our site by Providoli et al. (2005), the moss layer acted as a very efficient filter, absorbing nutrients that arrive on their surface in rainfall or throughfall. Nutrients taken up by mosses are generally not available to the vascular plants until the mosses die and undergo slow decomposition. As already shown for this site (Providoli 2005), mosses take up more NH_4^+ than NO_3^-, which may be due to the higher energy requirement for NO_3^- reduction. For the NO_3^- tracer, the herb and shrub layer (especially the monocotyledons), had a two times higher recovery in the meadow than in the forest. In a previous study on the same site, Providoli et al. (submitted) showed that vascular plants had a higher uptake for NO_3^- than for NH_4^+. The present study confirms those results.

The recovery in the below-ground biomass was not very high for both ecosystems and both tracers ($< 6\%$). Tracer uptake by the tree roots partly resulted in tracer recovery in the above-ground tree biomass, which was much higher for the NO_3^- tracer than for the NH_4^+ tracer. This indicates, that the trees preferred the NO_3^- tracer as did the vascular plants of the ground vegetation. This tracer recovery is comparable with a study on the same site (Schleppi et al. 1999a), trees recovered 8% of the tracer, and with a study by Buchmann et al. (1996) and May et al. (1996) in a *Picea Abies* forest, tree uptake was below 10% of the tracer.

The litter layer retained more than 10% of both tracers in both ecosystems and was therefore quite an important sink. The recovery can be due to abiotic absorption of the litter, to direct immobilisation into the litter during decomposition, but also through new litter input.

The soil was the most important sink in both ecosystems, with an exception for the NH_4^+ tracer in the forest. Other tracer studies, e.g. by Buchmann et al. (1996), May et al. (1996), Nadelhoffer et al. (1999a) or Lamontagne et al.

(2000) showed that the soil was the primary tracer sink. In these studies, the recovery was decreasing downwards the soil profile and was higher in the organic than in the mineral horizon. Similar recoveries were calculated on our site and have already been reported by Schleppi et al. (1999a) and Providoli et al. (submitted). However, in the actual study in the forest, the recovery in the B horizon was higher than in the upper two horizons for the NO_3^- tracer (17%), whereas the recovery in the meadow was below the detection limit. This is a remarkable difference between the two ecosystems, and it is not consistent with the studies mentioned before, showing that the recovery was decreasing downwards the soil profile. This fact could be due to a leaching process along macropores, i.e. cracks originating during the dry winter period in 2001. During snowmelt, the ^{15}N may have been leached in the B horizon, and after that the soil cracks may have closed again, so that the ^{15}N was stored in the impermeable soil layer.

On our site, in both ecosystems, both tracers were recovered particularly as immobilised soil N (ISN). This was already shown in previous short-term tracer studies on small plots on the same site (Providoli et al. submitted), where the tracer was immobilised either through biotic (by microbial biomass) or abiotic (via chemical processes) immobilisation. Also in other studies, ISN was recovered in the longer term (a few months after tracer application) (Perakis et al. 2001 and Hedin and Hart et al. 1993). However, these results are in contrast to a study by Zak et al. (2004) in a sugar maple-dominated hardwood forest on a sandy soil. In this study, there was after one year no recovery in the soil organic matter (SOM), which corresponds to the ISN on our site. ^{15}N was only recovered in the SOM within hours after tracer application, and was released after time steps longer than a month and shorter than a year.

On our site, the recovery in the microbial N was only detectable for the NH_4^+ tracer in the LF horizon, especially in the meadow. These results correspond well to the previous short-term study on small plots (Providoli et al. submitted), where especially the NH_4^+ tracer was favoured by the microbial biomass. However, it is surprising that the recovery is still so high after one year. Compared to other studies the recovery in microbial N is especially known to act in the short-term (Hart et al. 1993; Zogg et al. 2000; Perakis and Hedin 2001).

Contrary to tracer experiments on small plots (Providoli et al. submitted), it was not possible to replicate the present catchment study. An absolute comparison of the forest and meadow ecosystems or a quantitative extrapolation of our results over the landscape are therefore not possible. However, the average NO_3^- concentrations measured in the runoff are close to those of the surrounding Erlenbach catchment, and the N retention was also found to be similar compared to a site in Bavaria with higher N deposition but with similar geology and soils (Schleppi et al. 1998). In spite of the lack of replications, our small catchments can therefore be viewed as representative of a broad range of natural ecosystems on soils with hindered permeability in the temperate climate zone. The measured retention and partitioning of ^{15}N thus provide valuable

information about the fate of N from atmospheric deposition in these types of ecosystems.

Conclusion

Our tracer study showed that both ecosystems had the capacity to retain most of the added N tracer. The model of the N cascade, where the reactive N moves from one environmental system to another, is useful to interpret our results. The primary sink of the deposited N is the organic soil horizon. The ground vegetation (including the mosses) can be regarded as a temporary sink, from which N is entirely recycled to the soil on the time scale of years. Assuming that half of the ambient N deposition (12 kg ha^{-1} a^{-1} bulk, 17 kg ha^{-1} a^{-1} throughfall), remains in the soil at a decadal time scale, it would take 20–30 years (meadow resp. forest) to lower the soil C:N ratio by 1 unit. After 7 years of N addition ($+25$ kg ha^{-} a^{-1}) at the Alptal site, Schleppi et al. (2004) showed a decrease of the C:N ratio from 20 to 18 in the organic horizon of the forest soil, accompanied by increased NO_3^- leaching. This can be interpreted as a progressive saturation of the ecosystem in the long-term N cascade: atmosphere $\rightarrow$ terrestrial ecosystem $\rightarrow$ water. The very fast leaching observed in this study, in turn, can be considered as an incomplete interaction with the soil, thus as a shortcut atmosphere $\rightarrow$ water in the cascade.

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