# Nitrate leaching from a forest ecosystem with simulated increased N deposition

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#### 1 Introduction

Deposition of inorganic nitrogen (N) to natural or semi-natural ecosystems has increased over the past decades due to emissions by human activities. Inputs of ammonium NH<sub>4</sub><sup>+</sup> and nitrate NO<sub>3</sub><sup>-</sup> can induce an eutrophication of previously N-limited systems and finally cause their "nitrogen saturation" (Ågren & Bosatta, 1988; Aber et al., 1989). This happens when the deposition rate is larger than the retention capacity of the soil and vegetation, in other words if the ecosystem's "critical load" for nitrogen (Downing et al., 1993; Reynolds et al., 1998) is exceeded. Nitrate leaching is usually regarded as the main symptom of N saturation, at least in forests, where this form of N loss is more important than in nonforested natural or semi-natural ecosystems (van Breemen & van Dijk, 1988). Possible negative effects on the vegetation include nutritional disbalances, increased sensitivity to biotic and abiotic stresses, reduced root growth and altered competition between species (Ortloff & Schlaepfer, 1996).

As a non-point source, forests contribute to the eutrophication of streams, rivers and seas with N. Critical loads of N are exceeded in large tracts of Swiss forests (Kurz & Rihm, 1997). Nitrate leaching into surface and ground water is therefore a matter of concern. Leaching rates are obviously low compared to agricultural cropland, but forest areas are large enough to make them a considerable N source for Swiss water bodies (Braun et al., 1991).

Nitrogen deposition may have more impact on montane ecosystems because their N cycle is naturally tighter than in lower altitude ecosystems (McNulty & Aber, 1993). As part of the European research project NITREX (nitrogen saturation experiments: Wright & Rasmussen, 1998), we therefore studied the effects of an increased N deposition on a montane forest at Alptal, in the Swiss Prealps.

## 2 Material and methods

#### 2.1 Site description

The Alptal valley is located in central Switzerland. The research site has an altitude of 1200 m and a cool, wet climate (6°C mean temperature and 2300 mm precipitation per year). It is moderately affected by atmospheric nitrate and ammonium deposition: 12 and 17 kg N ha<sup>-1</sup> year<sup>-1</sup> in bulk and throughfall deposition, respectively (Schleppi et al., 1998b). Umbric Gleysols occur atop a Flysch substratum (calcareous sandstones with clay-rich shists). The mean slope is about 20 % with a west aspect. Dependent on the micro-topography, the soil bears different humus types: mor (raw humus) on the mounds and anmoor (muck humus) in the depressions, where the water table is high and reducing conditions frequent.

The trees are predominantly Norway spruce (*Picea abies*) with 15% silver fir (*Abies alba*) and mainly grow on the mounds. The stand is naturally regenerated, with trees up to 250 years old. With a leaf area index of 3.8, the density of the canopy is low; the basal area is 41 m<sup>2</sup> ha<sup>-1</sup> for 430 stems ha<sup>-1</sup> (>10 cm in diameter). The ground vegetation is well developed; different botanical associations form patches according to humus types and light conditions (Schleppi et al., 1999b).

## 2.2 Experimental catchments and N addition

Two forested catchments (approx.  $1500 \text{ m}^2$  each) have been delimited by trenches (Schleppi et al., 1998b). Nitrogen (as NH<sub>4</sub>NO<sub>3</sub>) was added to rainwater during precipitation events and applied by sprinklers to one of the catchments. This simulated a deposition increase of 30 kg N ha<sup>-1</sup> year<sup>-1</sup>. The water used as a vector for the N addition corresponded to a supplementary precipitation of approximately 130 mm per year. During the winter, the automatic irrigation was replaced by the occasional application of a concentrated NH<sub>4</sub>NO<sub>3</sub> solution on the snow with a backpack-sprayer. The effects of the treatment were compared with a control catchment receiving only unaltered rainwater and with one year of pre-treatment measurements on both catchments (Schleppi et al., 1998a).

#### 2.3 Sampling and analyses

Water discharge was measured with V-notch weirs. Runoff proportional samples, bulk deposition and throughfall were collected weekly. Soil solution was collected from plots in a replicated design with the same treatments as the catchments. Suction plates were used to sample at 5 and 10 cm depth, suction cups for 30 cm (Gr horizon). Water analyses included ICP-AES or ICP-MS (cations + P), IC (anions) and FIAS ( $NH_4^+$ ) (Schleppi et al., 1998b). Total dissolved N (TDN) was measured as nitrate after persulfate digestion: samples (10 ml) were autoclaved for 45 minutes at 130°C with 15 ml of potassium persulfate (20 g l<sup>-1</sup>) and NaOH (6.67 g l<sup>-1</sup>). Flasks (50 ml) were tightly closed for autoclavation, as this was found to reduce blank N values. Dissolved organic N (DON) was calculated as TDN-NO<sub>3</sub><sup>-</sup>-NH<sub>4</sub><sup>+</sup>.

# 2.4 <sup>15</sup>N labelling

The added nitrogen was labelled with <sup>15</sup>NH<sub>4</sub><sup>15</sup>NO<sub>3</sub> during the first treatment year (Schleppi et al., 1999a). Quarterly-pooled water samples were concentrated over exchange resins. Anions and cations were eluted separately and nitrate was reduced to ammonium with Devarda's alloy. Ammonium was converted to ammonia and captured in fibreglass filters enclosed in teflon membranes (adapted from Downs et al., 1998). The filters were analysed

by mass spectrometry. Vegetation and soil pools were analysed during the treatment year following labelling.

## **3** Results and discussion

#### 3.1 Water budget

Yearly water budgets (August to July) were calculated by estimating evapotranspiration according to Wendling (1975), but using the measured reflected radiation instead of a fixed albedo. Differences between inputs and outputs were within 7% (Fig. 1). Water budgets were therefore considered closed and element budgets of these artificially delimited catchments could be calculated from precipitation and runoff samples.



Figure 1: Water budget of the forested experimental catchments at Alptal

## 3.2 Nitrate leaching

Both catchments were very similar during the pre-treatment calibration year (Schleppi et al., 1998b). Afterwards, the elevated N deposition caused a strong increase in nitrate concentrations in the runoff water and topsoil solution (Fig. 2). There was, however, no effect in the soil solution collected below 10 cm. The very low nitrate concentrations in the gleyic horizon at the 30 cm depth were due to the reducing conditions, resulting in considerable losses via denitrification (Mohn, 1999). Comparison of the catchment runoff with the soil solution shows a close coupling between the topsoil solution and the runoff. Seasonal variations and absolute concentrations of nitrate at the 5 cm depth corresponded to the catchment runoff. This was confirmed in other chemical compounds as well (Hagedorn et al., 1997). In contrast, the soil solution at the 30 cm depth appeared to be completely uncoupled from the runoff generation.



Figure 2: Nitrate concentration in the soil solution (mean  $\pm$  standard error, n=5) and in the runoff.

The close linkage between soil water from the topsoil and the catchment runoff indicates the dominance of near-surface flow or preferential solute transport. This is supported by the fast response of surface and subsurface runoff to precipitation events (Feyen et al., 1996). As a consequence, water residence times and contact with the soil matrix are insufficient for a complete biological immobilisation of nitrate. Ammonium, on the contrary, is effectively retained by cation exchange. Net nitrification occurs only after a period of drying in small aerobic areas in the soil and enhances nitrate leaching during the next rainfall event (Hagedorn et al., 1999). Correspondingly, denitrification rates were increased on the treated plots only shortly after rain events with N addition (Mohn, 1999).

Higher nitrate concentrations were observed when the soil was dry and aerobic (June - July 1996, August - September 1997, November 1997), and during snowmelt events (February - April 1997), which led to considerable leaching. Nitrate leaching was about 4 kg N ha<sup>-1</sup> year<sup>-1</sup> in the control catchment. It increased by 3.2 and 4.7 kg N in the first and second treatment years respectively, corresponding to 11% and 16% of the N addition (Fig. 3).

#### 3.3 Nitrogen budget

Compared to other coniferous forests with manipulated N inputs (Bredemeier et al., 1998), Alptal has to be considered as a site with both moderate inputs and outputs of inorganic N (tab. 1). The response of the ecosystem to the manipulation is also intermediate between unsaturated sites in Scandinavia and highly impacted sites in the Netherlands or Germany. In our case, however, it is not possible to relate the nitrate leaching to a nitrogen saturation of the ecosystem, because a large proportion of the soil is bypassed, which limits the biological immobilisation of nitrate. The micro-topography is a further heterogenic factor affecting water flow and N cycle. A lateral migration of nitrate (and other nutrients) from the mounds to the depressions probably explains the differences observed in the nutrient indicator values of the vegetation (Schleppi et al., 1998b). This also explains the apparent contradiction between nitrate leaching and the slight N deficiency in the trees.

		control		N addition	
		$(\text{kg ha}^{-1} \text{ year}^{-1})$	(% of inputs)	$(kg ha^{-1} year^{-1})$	(% of inputs)
bulk deposition:	NO <sub>3</sub> <sup>-</sup>	6.0	31	6.0	13
	$\mathrm{NH_4}^+$	6.6	34	6.6	14
	DON	2.5	13	2.5	5
dry deposition <sup>1</sup>		4.6	23	4.6	10
N addition:	NO <sub>3</sub> <sup>-</sup>			13.9	29
	$\mathrm{NH_4}^+$			13.9	29
total inputs		19.7	100	47.4	100
runoff:	NO <sub>3</sub> <sup>-</sup>	3.7	19	7.3	15
	$\mathrm{NH_4}^+$	0.1	1	0.2	0
	DON	5.6	28	6.1	13
denitrification <sup>2</sup>		1.7	9	2.9	6
total outputs		11.1	57	16.5	35
apparent retention		8.6	43	30.9	65

Table 1: Nitrogen inputs and outputs of the forested experimental catchments at Alptal. <sup>1</sup>according to Fischer-Riedmann (1995); <sup>2</sup>according to Mohn (1999).

Dissolved organic nitrogen (DON) is an important form of nitrogen loss from unpolluted forests (Hedin et al., 1995). This was also the case in our moderately impacted site since more DON was leached than nitrate (tab. 1). So far, however, there was no effect of the treatment on DON losses. After 7 years of N additions (up to 150 kg ha<sup>-1</sup> year<sup>-1</sup>) to both a deciduous and a coniferous forest, Currie et al. (1996) reported an increased production of DON in the forest floor. They, however, found no increase in the amount exported from the soil. This indicates that effects of an elevated N deposition can only be expected in the long term, as a consequence of a lowered C/N ratio in the soil organic matter (McDowell et al., 1998). DON also accounted for a considerable flux in the bulk deposition. The measured concentrations were in the range of the few data published so far (Qualls et al., 1991; Cornell et al., 1995; Michalzik et al., 1997).

Quantifying the N fluxes of the forest ecosystem showed that, despite the increased nitrate leaching and denitrification due to the simulated increased deposition, the system retained most of the added N. Isotope labelling allowed to show where this supplementary N was incorporated.



Figure 3: Nitrate leaching from the forested experimental catchments as affected by a deposition increase of 30 kg N ha<sup>-1</sup> year<sup>-1</sup> (redrawn from Schleppi et al., 1998a).



Figure 4: Partitioning of the added <sup>15</sup>N-labelled  $NH_4NO_3$  into ecosystem pools and outputs (data from Schleppi et al., 1999a). Shaded segment: tracer not recovered.

## 3.4 <sup>15</sup>N partitioning

Neither the ground vegetation, nor the needles, bark or wood of the trees showed any increase in their nitrogen content (Schleppi et al., 1999b). Analyses of <sup>15</sup>N confirmed that the vegetation contained only small amounts of labelled N in both the year of application and the following year (Schleppi et al., 1999a).

On average, only 4% of the N in the ground vegetation was labelled with <sup>15</sup>N, corresponding to 5% of the N added to the system (Fig. 4). Current-year needles contained 1.1% N. Only 2% of it was from labelled N the year following the label application. The <sup>15</sup>N content of older needles was about 1/3 less. These results show that the uptake from current deposition is very small compared to the redistribution of N within the mature trees. This redistribution also takes place in the sapwood of the trunks, where labelled N could be recovered from at least 14 annual tree rings. A total of 8 % of the labelled N was recovered in the above-ground parts of the trees. Compared to other whole-ecosystem applications of <sup>15</sup>N (Nadelhoffer et al., 1999), this value is among the lowest. The relative small N-uptake by the vegetation is probably due to other ecological factors limiting plant and root growth at Alptal, especially phosphorus, anaerobic soil conditions and a short vegetation period (Schleppi et al., 1999b).

Most <sup>15</sup>N was recovered in the soil: 13 % in litter and roots; 63 % in the fine earth (<2 mm). The <sup>15</sup>N labelling decreased markedly with increasing horizon depth. This corresponds to the chemical analyses (Fig. 2) but is in contradiction with the downward migration of <sup>15</sup>N from <sup>15</sup>NH<sub>4</sub> and especially from <sup>15</sup>NO<sub>3</sub> found by Buchmann et al. (1996). This discrepancy can certainly be ascribed to the reducing conditions of the Alptal soil and to the hindered permeability of its gley horizon.

Nitrate leaching into runoff water accounted for 10% of the added N, corresponding to 2.8 kg N ha<sup>-1</sup> for the year of <sup>15</sup>NH<sub>4</sub><sup>15</sup>NO<sub>3</sub> application. This amount is close to the increase in nitrate leaching measured by chemical analyses during the same period (3.2 kg N ha<sup>-1</sup>) and indicates that most of the leached nitrate came directly from the treatment, without interaction with the soil N pool.

#### 4 Conclusions

The Alptal forest is subjected to moderate N deposition rates. Under ambient conditions, this montane ecosystem exhibits nitrate and DON leaching and so loses half the amount of deposited N into runoff water.  $NH_4NO_3$ , experimentally added to the throughfall, increased nitrate losses by leaching and by denitrification. These effects, however, were less than proportional to the simulated deposition increase. Analyses of runoff processes and soil solution chemistry as well as a <sup>15</sup>N tracer application showed that the leaching of nitrate was to a large extent hydrologically driven. In spite of the rapid water flow, the limited contact with the soil matrix was sufficient for ammonium to be removed by cation exchange, but not for a complete biological immobilisation of nitrate. Nitrate leaching is therefore better explained by precipitation or snowmelt water bypassing the soil rather than by a soil-internal N surplus.

Most of the deposited N is retained in the soil and relatively small amounts are taken up by the vegetation. For this reason, the risk of the N deposition affecting tree health or plant biodiversity is quite small in the short term. However, long-term effects can be expected due to a continuous build up of the N content in the soil and in the perennial species.

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