NITRATE LEACHING FROM A MOUNTAIN FOREST ECOSYSTEM WITH GLEYSOLS SUBJECTED TO EXPERIMENTALLY INCREASED N DEPOSITION

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(Received 20 August 2002; accepted 10 May 2003)

Abstract. Nitrate leaching was measured over seven years of nitrogen (N) addition in a paired-catchment experiment in Alptal, central Switzerland (altitude: 1200 m, bulk N deposition: 12 kg ha⁻¹ a⁻¹). Two forested catchments (1500 m² each) dominated by *Picea abies* were delimited by trenches in the Gleysols. NH₄NO₃ was added to one of the catchments using sprinklers. During the first year, the N addition was labelled with ¹⁵N. Additionally, soil N transformations were studied in replicated plots. Pre-treatment NO₃⁻-N leaching was 4 kg ha⁻¹ a⁻¹ from both catchments, and remained between 2.5 and 4.8 kg ha⁻¹ a⁻¹ in the control catchment. The first year of treatment induced an additional leaching of 3.1 kg ha⁻¹, almost 90% of which was labelled with ¹⁵N, indicating that it did not cycle through the large N pools of the ecosystem (soil organic matter and plants). These losses partly correspond to NO₃⁻ from precipitation bypassing the soil due to preferential flow. During rain or snowmelt events, NO₃⁻ concentration peaks as the water table is rising, indicating flushing from the soil. Nitrification occurs temporarily along the water flow paths in the soil and can be the source of NO₃⁻ flushing. Its isotopic signature however, shows that this release mainly affects recently applied N, stored only between runoff events or up to a few weeks. At first, the ecosystem retained 90% of the added N (2/3 in the soil), but NO₃⁻ losses increased from 10 to 30% within 7 yr, indicating that the ecosystem became progressively N saturated.

Keywords: forest ecosystem, Gleysols, nitrate leaching, nitrogen deposition, paired-catchment experiment

1. Introduction

Deposition of inorganic nitrogen (N) to terrestrial ecosystems has increased over the past decades due to emissions associated with human activities. Increased inputs of ammonium (NH₄⁺) and nitrate (NO₃⁻) can induce eutrophication of previously N-limited systems, including forests, and finally cause their 'nitrogen saturation' (Ågren and Bosatta, 1988; Aber *et al.*, 1989). According to these authors, increases in plant N uptake and N mineralisation occur before saturation is reached. Then, at saturation, there is a sharp increase in nitrification. Nitrate leaching, finally, increases as the N saturation continues. Nitrate leaching is thus

Water, Air, and Soil Pollution: Focus **4**: 453–467, 2004.

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usually regarded as the main symptom of N saturation in forests (Gundersen *et al.*, 1998).

As part of the European research project NITREX (Wright and Rasmussen, 1998; Emmett *et al.*, 1998), we studied the effects of increased N deposition on a mountain forest in Alptal, Switzerland (Schleppi *et al.*, 1998). Nitrogen was added underneath the tree crowns both in a paired-catchment experiment and in a replicated plot design. We found that under ambient conditions as well as with increased N deposition, NO_3^- leaching from the Gleysols of this site did not fit into the concept of N saturation by Aber *et al.* (1989). Specifically, NO_3^- leaching occurred even if the trees were still slightly deficient in N (1.1% N in needles) and if no effects of the N addition were observed on the vegetation (Schleppi *et al.*, 1999b). Further, neither the inorganic N pools in the soil (extractable NO_3^- and NH_4^+) nor the net nitrification were affected by two years of N addition, but NO_3^- leaching clearly increased (Hagedorn *et al.*, 2001).

In the model of N saturation by Stoddard (1994), some of the deposited NO_3^- may bypass the ecosystem during winter and spring before any N saturation. Concentrations measured at Alptal during the dormant season would fit to this 'stage 0'. Nitrate leaching, however, also happens regularly during the summer, which would point to advanced (stage 2) saturation. It is therefore not possible to classify our site on Stoddard's scale.

In most cases, NO_3^- leaching from forest ecosystems is studied on upland soils without waterlogging. Leaching itself is often calculated from concentrations in the soil solution collected below the rooting depth. Results from such experiments should not be extrapolated to the relatively impermeable Gleysols of our site. There, dye-tracer experiments showed a fast, preferential flow of infiltrating precipitation towards drainage trenches (Feyen *et al.*, 1999), and water flow paths with increased NO_3^- concentrations could indeed be detected by microsuction cups (Hagedorn *et al.*, 1999). We therefore proposed that NO_3^- in the runoff from this forest largely corresponds to NO_3^- from rain or snowmelt bypassing the soil matrix. This hypothesis is supported by the high proportion of ^{15}N -labelled NO_3^- leached from the treated catchment, indicating that it comes mainly from the applied N and not from an increased net mineralization of (unlabelled) soil N (Schleppi *et al.*, 1999a).

An end-member mixing analysis (EMMA) also showed that, at peak discharge, more than half of the runoff water in the experimental catchments came directly from precipitation (Hagedorn *et al.*, 2001). For a whole rainfall event, however, this direct contribution appeared to be only approximately 20%. The EMMA model was further able to predict NO_3^- leaching with a high coefficient of determination, but these predictions were systematically too low, reaching approximately 60% of the measured values. Even when the model clearly indicates some direct NO_3^- leaching due to a lack of interaction with the soil ('new' N), the analysis suggests a considerable contribution of pre-event ('old') N. This contradicts the minimal proportion of unlabelled N in the additional leaching induced by the treatment.

In this paper, we present both short- and long-term patterns of NO₃⁻ leaching with the aim of reconciling these previous findings on the relative contributions of 'new' and 'old' N.

2. Material and Methods

2.1. SITE DESCRIPTION

The Alptal valley is located in central Switzerland. The research site is at an altitude of 1200 m and has a cool, wet climate (6 °C mean temperature and 2300 mm precipitation per year, of which 800 mm is as snow). Atmospheric NO₃⁻ and NH₄⁺ deposition is moderate: 12 and 17 kg N ha⁻¹ a⁻¹ in bulk and throughfall respectively (Schleppi *et al.*, 1998). Umbric Gleysols occur atop a Flysch substratum (calcareous sandstones with clay-rich schists, a typical formation along the northern edge of the Alps). The slope is about 20% with a west aspect. Depending on the microtopography, 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 common.

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 yr old. With a leaf area index of 3.8, the density of the canopy is low; the basal area is 41 m² ha⁻¹ for 430 stems ha⁻¹ (>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, each approximately 1500 m² in size, were delimited by trenches (Schleppi *et al.*, 1998). The yearly water budgets for each catchment were found to be approximately balanced, and discharge and water chemistry are comparable with the surrounding, natural catchment of the Erlenbach stream (Schleppi *et al.*, 1998). Element budgets of the artificially delimited catchments can thus be calculated from precipitation and runoff samples. The depth of the water table is automatically measured in two piezometers, one on a mound, one in a slight depression on the slope.

Nitrogen (as NH₄NO₃) was added to rainwater during precipitation events and applied by sprinklers to one of the catchments. This simulated an increased deposition of 25 kg N ha⁻¹ a⁻¹ to the ground vegetation and the soil. The water used as a vector for the N addition corresponded to a supplementary precipitation of approximately 110 mm per year. During the winter, the automatic irrigation was replaced by the occasional application of a concentrated NH₄NO₃ solution on the snow with a backpack-sprayer. The seasonality of the ambient deposition rates was thereby approximately mimicked. The effects of the treatment were compared

with a control catchment receiving only unaltered rainwater and with one year of pre-treatment measurements made from both catchments.

2.3. SAMPLING AND ANALYSES

Bulk deposition and throughfall samples were collected weekly. In both catchments, water discharge was continuously measured with V-notch weirs. Runoff samples were collected proportionally to the discharge and bulked weekly (Schleppi *et al.*, 1998). Additionally, runoff from several rainfall and snowmelt events was monitored with a high temporal resolution (Hagedorn *et al.*, 2000). Soil solution samples were collected from plots (20 m² each, 5 replications) located near the catchments and subjected to the same N treatments. Suction plates were used to sample water at 5 and 10 cm depth, suction cups for 30 cm (reduced Bg horizon) (Hagedorn *et al.*, 2000). Anions in water samples were analysed by ion chromatography (Schleppi *et al.*, 1998). Soil cores were taken after 2 and 7 yr on 20 grid points of the treated catchment and analysed for carbon (C) and N contents with a C + N analyser.

2.4. ¹⁵N LABELLING

The added N was labelled with ¹⁵NH₄¹⁵NO₃ during the first treatment year (Schleppi *et al.*, 1999a). Water samples, pooled quarterly, were concentrated over exchange resins. Anions and cations were eluted separately and NO₃⁻ was reduced to NH₄⁺ with Devarda's alloy. Ammonium was converted to NH₃ and captured in fibreglass filters enclosed in teflon membranes (adapted from Downs *et al.*, 1999). The filters were analysed by mass spectrometry.

3. Results

3.1. EVENT-BASED ANALYSES OF NO₃⁻ LEACHING

An initial event-based sampling of runoff water was done in August 1994, during the pre-treatment year and at the end of a wet week (60 mm precipitation in the 6 previous days). During the sampled 12 h rainfall event, NO₃⁻ concentrations were highest during the first discharge peak (Figure 1). By the end of the runoff peak, these concentrations had progressively declined to values close to those measured before the rain. During this event, NO₃⁻ concentrations in the runoff remained within the range measured in the topsoil solution (NO₃⁻-N: 0.05–0.7 mg L⁻¹, Hagedorn *et al.*, 2001). They were also similar to concentrations in the rain itself (0.22 mg L⁻¹ NO₃⁻-N). In the first samples from each catchment, the NO₃⁻-N/Cl⁻ ratios were 0.6 and 1.4, respectively. From the second to the last samples, the ratios declined from 0.5 down to below 0.4 in both catchments.

Figure 1. Runoff from the experimental catchments and its concentrations in NO_3^- and chloride during a summer rainfall event, before the N addition started.

Figure 2. Runoff from the control catchment, its concentrations in NO_3^- and Cl^- , and depth of the water table during a summer rainfall event.

In samples collected in July 2000 from the first runoff peak of another long rainfall event (Figure 2), NO_3^- concentrations increased above values measured in the soil solution, or in the precipitation of this event (NO_3^- -N: 0.31 mg L^{-1}). The water table in the depression piezometer also rose sharply during this first runoff peak. During the second part of the event, the water table remained high in the depression, and slowly increased in the mound piezometer. At the same time, a broad NO_3^- peak was measured in the runoff. During the first discharge peak, the ratio NO_3^- -N/ Cl^- dropped from 1 to 0.6. Then, during the second peak, it increased up to 1.2, before decreasing again in the last three samples down to 0.9.

The first two days of the main snowmelt period were also sampled in April 1995, before the N treatment began (Figure 3). These samples showed a general decrease in NO_3^- concentrations, except for a short peak on the second day. In each catchment, this NO_3^- peak corresponded to the maximum runoff, and also approximately to the maximum water table levels.

The analysis of one rainfall event in July 1998, during the N addition, has previously been presented in Hagedorn *et al.* (2001). There was a NO_3^- peak at the beginning of the event, and NO_3^- concentrations were elevated in the runoff of the treated catchment throughout the event.

Runoff water was further sampled in April 1999 during the 6th and 7th days of a long snowmelt period (Figure 4). Compared to the event sampled in April 1995, the NO_3^- and Cl^- concentrations were low and stable in the runoff from the control catchment. In the N-treated catchment, Cl^- was also low, but NO_3^- concentrations were elevated during the whole event.

3.2. WEEKLY ANALYSES OF NO_3^- LEACHING

Over the seven years of weekly runoff analyses, low NO_3^- concentrations were usually measured in discharge-proportional samples collected during the summer. Higher concentrations were observed when the soil was relatively dry, like in autumn 1995, or during snowmelt, as in winter 1996 (Figure 5). During snowmelt, high concentrations are sometimes coupled to high discharges, leading to considerable NO_3^- fluxes. Over 7 yr, 45% of the losses from the control catchment occurred during three months of the year, between March and May. Based on these observations, a multiple regression analysis was calculated to explain NO_3^- concentrations in weekly runoff samples from the control catchment. The concentration data were first transformed to their logarithms in order to obtain a normal distribution of the residues. Independent variables were: the discharge (as logarithm), the season (as sine and cosine functions with a one year period), the variations of the water table (as the sum of hourly increases), and the bulk N deposition during the week (after subtracting the proportion of snow). The regression model had a coefficient of determination $r^2 = 0.43$ for the control catchment and each factor was highly significant ($p < 0.001$), also in a stepwise model definition. The effect of the discharge was evident but relatively weak: NO_3^- concentrations changed by a factor 10

Figure 3. Runoff from the experimental catchments, its concentrations in NO_3^- and Cl^- , and depth of the water table during two days at the beginning of a snowmelt period.

while the discharge changed by more than 4 orders of magnitude. The maximum of the seasonality curve was in April, 2.5 times as high as the minimum in October. Nitrate concentrations further increased by 2% for each cm that the water table rose during the week. In the treated catchment, the effect of the discharge remained, but the seasonality was damped, with a ratio of 1.6 between spring and autumn concentrations. With the addition of N, the effect of the water table fluctuations was no longer significant.

Figure 4. Runoff from the experimental catchments, its concentrations in NO_3^- and Cl^- , and depth of the water table during a longer snowmelt period. In the treated catchment, the water delivered by the irrigation system contained 17 ml L^{-1} of N.

Figure 5. Nitrate concentrations in discharge-proportional runoff samples from the experimental catchments, with the addition of N beginning in the treated catchment after one year of pre-treatment measurements.

The weekly discharge-proportional samples from both catchments were very similar during the pre-treatment year (Figure 5). Within weeks of beginning the N application, the NO_3^- concentrations in runoff doubled in the treated catchment compared to the control catchment. Thereby, the shape of the concentration curve remained almost the same (correlation: $r = 0.73$, $p < 0.001$), only its magnitude was increased.

3.3. LONG-TERM TRENDS

Nitrate-N leaching from the control catchment varied from year to year between 2.5 and 4.8 kg ha⁻¹ (Figure 6). It was significantly correlated with the yearly bulk N deposition ($r^2 = 0.63$, $p = 0.02$).

In the first treatment year, NO_3^- leaching was 3.1 kg N higher in the treated catchment than in the control, corresponding to 11% of the N addition. Out of these 3.1 kg, 2.8 ± 0.3 were labelled N. In the following years, the difference between the catchments increased up to 7 kg N, or 30% of the addition. The ^{15}N signal in runoff NO_3^- , however, disappeared within three months from the end of the labelling. The difference between total ^{15}N in lyophilised samples and $^{15}\text{NO}_3^-$ did not indicate any labelled dissolved organic N, nor was any ^{15}N signal detected in throughfall after labelling.

Figure 6. Yearly NO_3^- leaching from the forested experimental catchments during a pre-treatment year (0 davidson = 1994–1995) followed by seven years of N addition (at $25 \text{ kg ha}^{-1} \text{ a}^{-1}$, with ^{15}N labelling in year 1). C/N mass ratios ($\pm$ std. error) in the forest floor of the treated catchment after 2 and 7 yr of N addition.

Between 1997 and 2002, the C/N ratio of the forest floor decreased significantly ($p = 0.02$) from 20.2 to 17.8%. If the C content is assumed constant, this corresponds to an accumulation of $23 \pm 10 \text{ kg N ha}^{-1} \text{ a}^{-1}$.

4. Discussion

In winter, the occasional application of NH_4NO_3 to the snow appears to have a sustained effect on NO_3^- concentrations in runoff water. During runoff events, the single N additions by the sprinklers had no distinguishable effect on the concentration curves of NO_3^- in runoff. Finally, the treatment clearly increased weekly NO_3^- concentrations, but did not change the shape of this time-series compared to the control. These observations show that the applications were frequent enough to simulate wet N deposition, even on our site with rapid runoff. This is important since the same amount of nutrient can induce different effects depending on the rate and frequency of application (Persson, 1981), which can then alter the partitioning between plant species, soil, soil microorganisms, and leaching (Johnson, 1992). Our treatment by sprinklers does not allow for a foliar uptake by the trees. This

process, however, was shown to be small in the case of wet deposition (Wilson and Tiley, 1998).

Using natural abundances of ^{15}N and ^{18}O from riverine nitrate, Mayer *et al.* (2002) established that, in large forested catchments, NO_3^- leaching was mainly from nitrification, with only a small possibility of a direct contribution from atmospheric deposition. Our ^{15}N labelling gives opposite results, with 90% of the additional leaching being labelled. If there are interactions between added N and pre-existing ecosystem pools, they must be limited to small N pools and to a relatively short time span (since the ^{15}N signal disappeared from the runoff within three months).

Event-based analyses, including the already published end-member mixing analysis (Hagedorn *et al.*, 2001), show a different pattern. Nitrate peaks usually correspond to a rising water table. This indicates flushing from those soil layers, which become saturated with water (Creed and Band, 1998). Because the water table always rises first in the depressions and only later under the mounds, NO_3^- flushing can also occur as successive NO_3^- peaks during a hydrological event, like in July 2000. This effect is similar to the 'variable source area dynamics' described by Creed and Band (1998). Nitrate peaks observed during the early snowmelt in April 1995 also corresponded to the water table reaching a previously unsaturated soil layer. The highest NO_3^- peak, however, occurred simultaneously with a Cl^- peak. These high concentrations indicate hydrological causes (which affect both anions), without a contribution of net nitrification (which would increase the $\text{NO}_3^-/\text{Cl}^-$ ratio). During the snowmelt sampled in April 1999, concentrations were consistently low in the control catchment. Concentrations declining during the early snowmelt and staying low through the later snowmelt phase are best explained by the ionic fractionation within the melting snowpack (Waldner *et al.*, 2000). The higher concentrations observed in the control catchment in April 1995 can be explained by the same mechanisms since its snowmelt is always slightly later than in the treated catchment. The absence of a clear NO_3^- peak in April 1999 is probably due, as in the rainfall event of August 1994, to previous flushings, which already depleted NO_3^- from the soil zones reached by the water table.

During some events, like in July 2000, NO_3^- peaks correspond to increased $\text{NO}_3^-/\text{Cl}^-$ ratios. In this case, some NO_3^- appears to be produced by nitrification. Even if no net nitrification was measurable in the bulk soil of this forest site, Hagedorn *et al.* (1999) showed that some nitrification takes place along the macropores as they drain out between hydrological events. In summary, NO_3^- is released by the Gleysols at the test catchments by three different mechanisms: (1) NO_3^- from precipitation bypassing the soil by fast preferential and sub-surface-flow, (2) flushing of NO_3^- from previous precipitation events temporarily stored in the soil pores, and (3) flushing of NO_3^- produced by nitrification (in the macropores of the unsaturated soil). In any case, NO_3^- flushed as the water table rises is mainly from recently deposited N, as shown by its strong ^{15}N signal. From the data obtained so far, it is not possible to distinguish between the nitrification of deposited NH_4^+ and

nitrification occurring after the mineralisation of assimilated NO₃⁻. Because the ¹⁵N signal disappears within three months, these release mechanisms are effective on a limited time-scale only. This explains why event-based and long-term contributions of 'new' vs. 'old' N appeared to be in contradiction. Likewise, the discrepancy observed in correlations between NO₃⁻ and discharge is also a matter of the time-scale: mostly positive within a single event, but negative between weekly samples. The effect of flushing, however, can still be observed in the weekly analyses as NO₃⁻ concentrations increase with the cumulative movements of the water table during the sampling time. To describe this quantitatively and in more details, it would be necessary to draw a model taking into account the whole dynamics of the water table across the variable zones, which contribute to runoff (mounds vs. depressions).

The observed NO₃⁻ leaching mechanisms occur preferentially along water-flow paths in the soil (macropores). Because soil heterogeneity is not considered in the N saturation models by Ågren and Bosatta, (1988) or by Aber *et al.* (1989), our site cannot be described within these concepts. A further limitation of these models was the fact that N inputs to the stable soil organic matter are only possible via litterfall. In their newer hypotheses, Aber *et al.* (1998) also consider direct immobilisation in the soil, which is clearly what happened with the Alptal forest. Even if it was detectable by ¹⁵N analyses, the uptake of deposited N by the trees was insignificant for their nutritional status (Schleppi *et al.*, 1999b) and thus for the return of N as litterfall. Foliar leaching did not play a role either as there was no ¹⁵N signal in throughfall after labelling. About 2/3 of the ¹⁵N were recovered in the soil (Schleppi *et al.*, 1999a), and the immobilisation in the forest floor was very important (about 15 kg ha⁻¹ of labelled N in litter and forest floor). This immobilisation may be abiotic, especially since reducing conditions are frequent in this soil and could enable the redox cycles recently hypothesised by Davidson *et al.* (2003). The decline in the C/N ratio measured within five years is strong but still in agreement with the ¹⁵N partitioning. Comparing different sites, Gundersen *et al.* (1998) and Emmett *et al.* (1998) showed the importance of the C/N ratio in the forest floor for its ability to retain N deposition. In our experiment, we observed a progressive increase in NO₃⁻ leaching within seven years of N addition. According to its disappearing ¹⁵N signal, this increased leaching is due to a reduced ability to retain further N inputs rather than to a delayed release of N accumulated in the previous years. We therefore hypothesise that the declining C/N ratio is more and more limiting the ability of the soil to immobilise N from atmospheric deposition.

Acknowledgments

This study received financial support from the Swiss Federal Office of Science and Education and from the Swiss National Fund for Scientific Research. The chemical analyses were done at the central laboratory of the Swiss Federal Institute for

Forest, Snow and Landscape Research (WSL), Birmensdorf (D. Pezzotta). Analyses of ^{15}N were conducted at the Paul Scherrer Institute (PSI), Villigen (Dr. R. Siegwolf, Dr. M. Saurer). Thanks to Melissa Swartz for the English corrections.

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