



Experimental addition of nitrogen to a whole forest ecosystem at Gårdsjön, Sweden (NITREX): Nitrate leaching during 26 years of treatment[☆]

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ABSTRACT

Chronic high deposition of nitrogen (N) to forest ecosystems can lead to increased leaching of inorganic N to surface waters, enhancing acidification and eutrophication. For 26 years nitrogen has been added as ammonium nitrate (NH_4NO_3) at $40 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ to a whole forested catchment ecosystem at Gårdsjön, Sweden, to experimentally simulate the transition from a N-limited to N-rich state. Over the first 10 years of treatment there was an increasing amount of nitrate (NO_3^-) and to a lesser extent ammonium (NH_4^+) lost in runoff, but then N leaching stabilised, and for the subsequent 16 years the fraction of N added lost in runoff remained at 9%. NO_3^- concentrations in runoff were low in the summer during the first years of treatment, but now are high throughout the year. High frequency sampling showed that peaks in NO_3^- concentrations generally occurred with high discharge, and were enhanced if high discharge coincided with occasions of N addition. Approximately 50% of the added N has gone to the soil. The added N is equivalent to 140 years of ambient N deposition. At current ambient levels of N deposition there thus appears to be no immediate risk of N saturation at this coniferous forest ecosystem, and by inference to other such N-limited forests in Scandinavia.

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1. Introduction

Chronic excess deposition of nitrogen (N) leads to adverse effects in forest ecosystems such as changes in growth, altered species composition and abundance of ground vegetation (Bobbink et al., 2010; Boxman et al., 1998) and can result in leaching of inorganic N to surface waters (Dise and Wright, 1995) exacerbating freshwater acidification and contributing to nutrient loading to marine ecosystems (Pearl, 1999). Synoptic surveys of European coniferous-forest ecosystems receiving differing amounts of N deposition show that leaching of inorganic N is related to the carbon to nitrogen (C/N) ratio in soil (Gundersen et al., 1998;

MacDonald et al., 2002). Forests with C/N ratio above about 25 g g^{-1} leach little NO_3^- whereas those with C/N ratio below 25 leach an appreciable fraction of the input N. The implication is that chronic increased N deposition results in progressively decreasing C/N ratio in soil, and ultimately to increased concentrations of inorganic N in runoff. The synoptic data, however, do not tell how much N is needed and how long it will take to reach the threshold for further increased N leaching.

Concern over the potential risk posed by N deposition has led to extensive research on the subject over the past 30 years. One of the approaches was to conduct large-scale whole-ecosystem experiments with N addition to induce the transition from N-limited to N-rich system. The hypothesis is that the added N would reduce the C/N ratio of soil, cross the threshold of C/N 25 and induce increased leaching of inorganic N. In Europe, several of these experiments were gathered under the umbrella of the EU project NITREX (Nitrogen Saturation Experiments) (Wright and van Breemen, 1995). The concept behind NITREX is that in forests that do not leak appreciable amounts of inorganic N, increased N deposition will

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cause progressively increased leaching of N to runoff, and, conversely, in forests that leak a large amounts of inorganic N, decreased N deposition will result in decreased leaching of N.

One of the NITREX sites is located at Gårdsjön, Sweden (Fig. 1). The G2 NITREX catchment is 0.5 ha of N-poor coniferous forest with C/N ratio in soil (organic) of about 30 g g^{-1} and no appreciable loss of inorganic N in runoff before the start of N additions. The adjacent F1 CONTROL catchment served as untreated reference. Beginning in April 1991 at G2 NITREX the ambient N deposition of about $9 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ has been augmented uninterrupted until the present by experimental additions of $40 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ as NH_4NO_3 . This total N deposition of about $50 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ was chosen to reflect ambient levels in forests of central and western Europe that have extensive symptoms of N saturation. The Gårdsjön NITREX experiment has been run without a break for 26 years and the results reported regularly, most recently by Moldan and Wright (2011). Over the 26 years the cumulative dose of N added and incoming from the atmosphere has been $1252 \text{ kg N ha}^{-1}$. In 1991 the N pools in the catchment were 573 kg N ha^{-1} in standing biomass with average C/N of 92 g g^{-1} (Kjonaas and Stuanes, 2008), $2400 \text{ kg N ha}^{-1}$ in the organic soil with C/N of 30 g g^{-1} and $3200 \text{ kg N ha}^{-1}$ in the mineral soil with C/N of 28 g g^{-1} (Moldan et al., 2006). Ecosystem studies included throughfall, litterfall, mineralisation and N processes in soil, soil chemistry, soil solution chemistry, tree biomass and chemical composition, biodiversity of ground vegetation, and mycorrhiza (Brandrud and Timmermann, 1998; Kjonaas and Stuanes, 2008; Kjonaas et al., 1998; Stuanes and Kjonaas, 1998). Data from the Gårdsjön NITREX experiment have been used to evaluate and further develop process-oriented biogeochemical models (Oulehle et al., 2015).

While N deposition at Gårdsjön G2 NITREX was artificially increased, ambient regional sulphur (S) deposition was decreasing (Karlsson et al., 2011). The declining S deposition resulted in recovery from acidification in surface waters (Garmo et al., 2014); increased NO_3^- leaching could offset this recovery. Another observed regional trend was the increasing DOC concentrations in surface waters (Hruška et al., 2009; Garmo et al., 2014). The increasing DOC concentrations are probably related to declining acid deposition and consequently decreasing ionic strength (Hruška et al., 2009).

Here we assess the progression of the site from N-limited to N-rich by means of data from the 26 years of experimental N additions. Further we identify the changes in factors leading to short-term peaks in NO_3^- in runoff by means of high-frequency samples of runoff chemistry collected during the 25th year of treatment. We also analyse the runoff chemistry record with respect to recovery from acidification and trends in runoff DOC and DON.

2. Materials and methods

2.1. Site description

The Gårdsjön research station is located on the west coast of Sweden approximately 15 km inland. The area has historically received large amounts of acid deposition and has been recovering from acidification of soils and waters since the 1990s. The deposition of sulphur (sea-salt corrected, measured as a throughfall) has decreased significantly in the area from an average of $17 \text{ kg S ha}^{-1} \text{ yr}^{-1}$ in 1989–90 to $1.5 \text{ kg S ha}^{-1} \text{ yr}^{-1}$ in 2015–16. Deposition of N, on the other hand, has shown no significant decrease during this time period and was on average $9 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ between 1989 and 2016 (sum NO_3^- and NH_4^+ , measured in open field precipitation).

Vegetation consists mostly of a mixed coniferous forest dominated by Norway spruce. The forest is mature, and there has been no thinning or harvesting during the last 50 years. The bedrock is

siliceous igneous and metamorphic rocks overlain by thin and patchy moraine. The dominant soil is podsol. The average long-term temperature and precipitation are 7.4°C and 1071 mm yr^{-1} , respectively.

2.2. Experimental design and N additions

The NITREX experiment in G2 started in 1991. To achieve an increase in N deposition, medical grade NH_4NO_3 dissolved in de-ionised water was sprinkled over the forest floor across the whole 5250 m^2 G2 catchment area. From 1991 to spring 2010, the dispersion of the dissolved NH_4NO_3 was done on average twice a month via a ground-level sprinkler system. Beginning in 2010 dispersion has been done monthly by a backpack manual pump sprayer. Manual application was preferentially done during rainy days to avoid damage to plant surfaces due to direct exposure to highly concentrated NH_4NO_3 solution. The yearly additions of N were between 28.9 and $51.6 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ (average 39.5), with applications proportional to the amount of ambient precipitation (Table S1).

2.3. Sampling and data sources

Meteorology Meteorological parameters have been monitored at the weather station mast at the Gårdsjön field station, located about 1 km from the G2 NITREX catchment. The weather station recorded wind speed and direction, precipitation, air temperature, relative humidity and global radiation with a resolution of 1 h. Soil temperature has been monitored in the humus layer at 5 cm depth at F1 CONTROL.

Deposition Precipitation was measured close to catchment F1 CONTROL, where also open field deposition was collected and analysed monthly. Throughfall was collected monthly at F1 CONTROL and from 1989 to 1998 also at G2 NITREX.

Runoff Discharge from G2 NITREX 1990–2011 was measured by continuous metering of all the water passing the catchment outlet. At the F1 CONTROL site the discharge has been monitored continuously since 1979 at the wier installed at the catchment outlet. From 2011-present discharge at G2 NITREX was calculated from the discharge in catchment F1 CONTROL using a regression made from the years of measurements of discharge from both catchments (1990–2011). Samples of runoff for chemical analysis were taken by either volume-weighted (from 1989 to 2011) or time-integrated procedures using an ISCO automatic sampler. The time-integration (from 2011-present) entailed sampling every 6 h and bulking 6 samples to give a 36-h sample. These in turn were bulked to give fortnightly or monthly samples that were then analysed for chemical constituents. In 2013 the ISCO system was replaced and during the first half of 2013 one monthly sample was collected directly in the runoff. In addition, over the period 1 September 2015–4 October 2016 a more intensive analytical procedure was used, and the 36-h samples were all analysed individually (total 205 samples, December 2015 missing due to failure of the ISCO sampler). The routine analysis of the fortnightly or monthly samples was conducted by the IVL laboratory; parameters included major ions, N species, and DOC. The intensive samples were analysed at the Czech Geological Survey for the NO_3^- , NH_4^+ , Cl^- and SO_4^{2-} . Chemical analysis of precipitation, throughfall and runoff were performed at IVL Swedish Environmental Research Institute using routine accredited methods. Samples were filtered with 0.45 mm filter. pH was determined by electrode, base cations and anions by ion chromatography, DOC by high-temperature combustion, and NH_4^+ colourimetrically by flow injection analysis. Aluminium was determined by ICP-OES (Inductive Coupled Plasma-Optical Emission Spectrometry) at the Swedish University

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