Ecological Indicators 82 (2017) 451–462

Contents lists available at ScienceDirect

Ecological Indicators

journal homepage: www.elsevier.com/locate/ecolind

Multiple nitrogen saturation indicators yield contradicting conclusions on improving nitrogen status of temperate forests

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ARTICLE INFO

Keywords:
Nitrogen status
Soil solution
DON:TDN ratio
DOC:NO_3^- Ratio
Foliar N:P ratio
Foliar Bc:N ratio
ICP Forests

ABSTRACT

Nitrogen (N) depositions in Europe are decreasing, but this could not explain faster than expected improvement of N saturation indicators in temperate forests. Alongside there were local signs of initial recovery from acidification during the past three decades and enhanced leaching of dissolved organic carbon and nitrogen (DOC, DON). These two global change processes both affect total dissolved nitrogen (TDN) levels and often occur simultaneously, hence complicating mechanistic explanations for changing European forest N status. We aimed to test the hypothesis that forest N status in northwest Europe has started to improve. If this hypothesis is confirmed, we wanted to investigate to what extent such improvement is due to reduced N deposition. We evaluated the evolution of multiple N saturation indicators in five ICP Forests Level II plots in northern Belgium, using long-term soil solution and foliage datasets. The DON:TDN ratio (molar) in soil solution increased overall in the O horizon (mean 0.279–0.463, slope 0.023–0.037 yr^{-1}) and in the mineral soil (mean 0.134–0.78, slope 0.007–0.051 yr^{-1}) between 2005 and 2014. The DOC:NO_3^- ratio (molar) in soil solution increased in three plots in the O horizon (mean 6.84–22.15, slope 0.58–1.92 yr^{-1}) and in four plots in the mineral soil (mean 2.07–25.32, slope −0.06–5.76 yr^{-1}) between 2002 and 2014. The ratio of N and phosphorus (P) concentrations in foliage (mg g^{-1}) and the ratio of base cations (Bc = Ca + K + Mg) and N concentrations in foliage (molar) remained unaltered between 1999 and 2013. Changes in the soil solution chemical composition thus confirmed an improvement in forest N status, despite sustained high NO_3^- concentrations, but biotic recovery appeared to be lagging behind. This demonstrates that insight in forest recovery from N saturation requires a multiple indicator approach, and further monitoring of tree nutritional status alongside soil processes is needed to monitor the evolution of European forest N status.

1. Introduction

Atmospheric deposition of inorganic nitrogen (N) and sulphate (SO_4^{2-}) caused an accelerated acidification and N saturation of temperate forest soils and surface waters in large parts of Europe and the US mainly during the second half of the 20th century (van Breemen et al., 1984; Aber et al., 1989). In temperate forests, soil acidification often depleted base cations (calcium, potassium, magnesium), increased soil solution aluminium (Al^{3+}) concentrations and nitrate (NO_3^-) leaching, and disrupted dissolved organic matter cycling (Kalbitz et al., 2000; Aber et al., 2003; McDowell et al., 2004; Pregitzer et al., 2004; Monteith et al., 2007).

Despite a substantial decrease of inorganic N depositions in large regions of Europe during the past decade (Waldner et al., 2014), critical loads and limits, i.e. the level below which significant harmful effects do not occur according to present knowledge (Nilsson and Grennfelt, 1988) for inorganic N are still frequently exceeded in many European forests (Iost et al., 2012; Waldner et al., 2015). Despite publication of several papers on this matter it is still unclear how long N saturated forests will take to recover. Indeed, many factors, including management, SO_4^{2-} deposition and natural succession, change alongside inorganic N deposition, and individual compartments of the forest ecosystem (e.g., vegetation, below-ground communities, soil and soil solution) react with varying speed to changes in N availability (Stevens, 2016). Nitrogen availability also depends on forest size, forest type, soil type and sampling time and the complex interplay between biotic and abiotic processes (Pastor and Post, 1986; Callesen et al., 1999). In the present study we evaluate the evolution of forest N saturation in Flanders, a region in West-Europe where both inorganic N and SO_4^{2-} depositions strongly decreased, using a selection of indicators based on

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http://dx.doi.org/10.1016/j.ecolind.2017.07.034
Received 22 December 2016; Received in revised form 28 May 2017; Accepted 13 July 2017
Available online 21 July 2017
1470-160X/

long-term data of the elemental concentrations in soil solution and tree mineral nutrition at intensive monitoring sites with varying soil types and tree species.

Long-term data on the concentrations of N species in soil solution collected at intensive forest monitoring plots yield crucial information about N availability and N saturation in forests. While unpolluted forests generally exhibit very limited N losses, consisting almost entirely of dissolved organic nitrogen (DON), N saturated forests with low C:N ratio in organic layers typically show elevated NO_3^- leaching below the rooting zone (Aber et al., 1989, 2003; Perakis and Hedin, 2002; Perakis and Sinkhorn, 2011). The ratio between DON and dissolved inorganic nitrogen (DIN) in soil solution, DON:DIN, has therefore been used as an indicator for N saturation in forests (Williams et al., 2001, 2004; Park and Matzner, 2006). Similarly, low ratios of DON to total dissolved nitrogen (TDN) in soil solution, DON:TDN, and of dissolved organic carbon (DOC) to NO_3^-, DOC:NO_3^-, are often used as indicators of soil N saturation (Currie et al., 1996; Sleutel et al., 2009; Taylor and Townsend, 2010).

The evaluation of forest N status may, however, be confounded when DOC and DON concentrations change alongside TDN and NO_3^- concentrations. Recovery from acidification was indeed found to mobilize DOC and DON (Verstraeten et al., 2016) in five ICP Forests Level II plots in northwest Europe. The concomitant evolutions in N deposition and recovery from acidification, both affecting DON mobility, therefore leads us to question the true share of reduced N depositions in a possible improving N status of these forests. The prime aim of this research was thus to assess recent evolutions in the N status of these five ICP Forests Level II plots in Flanders, northern Belgium. We monitored the concentrations of DON and TDN (2005–2014) in the deposition and soil solution and of DOC and NO_3^- (2002–2014) in the soil solution and critically assessed trends in classic molecular-ratio based indicators. Because throughfall DIN deposition in the plots decreased from 42.1 kg ha^{-1} yr^{-1} to 20.2 kg ha^{-1} yr^{-1} during the period 1994 to 2010 (Verstraeten et al., 2012), we hypothesized that the DON:TDN ratio (molar) and thus the DOC:NO_3^- ratio (molar) increased over the past decade. We expect, however, that increased DON and DOC mobilization due to concomitant recovery from acidification renders these shifting ratios only partly indicative for the actual improvement in forest N status. Moreover, concentrations of DOC, DON, TDN and NO_3^- in throughfall and soil solution obviously are not representative of the tree biological status. Instead, the N status of forests can alternatively be derived from the foliar concentrations of N and phosphorus (P) and their ratio, N:P, in relation to tree species specific critical limits (Mellert and Göttlein, 2012; Veresoglou et al., 2014). A similar indicator is the ratio of the foliar concentrations of base cations (Bc = Ca + K + Mg) and N, Bc:N (Meesenburg et al., 2016). Tree nutritional status provides an integrative criterion for the assessment of site conditions and environmental factors (e.g. soil acidification, N saturation, climate change) and is important to control the success of restoration measures and to follow the natural recovery of forest ecosystems from former anthropogenic impacts (Mellert and Göttlein, 2012). To more broadly track the impact of reduced N deposition on forest N status, we extended the evaluation with the 1999–2013 trends in the foliar N:P ratios and Bc:N ratios and hypothesized that these had decreased and increased, respectively, as a consequence of lowered soil mineral N availability.

2. Material and methods

2.1. Study area

Five plots of the ICP Forests intensive monitoring network (Level II) in Flanders (northern part of Belgium) were included in this study. Flanders has a moderate Atlantic climate with a mean annual precipitation of 852 mm and mean temperature of 10.5 °C (long-term averages for 1981–2010 for the meteorological station of Uccle, www.meteo.be). A detailed description of these plots can be found in Verstraeten et al. (2012). Detailed soil characteristics (C:N ratio, pH-$CaCl_2$, cation exchange capacity, base saturation and soil texture per morphogenetic horizon) were published in Verstraeten et al. (2016). Two plots are located in coniferous forest: *Pinus sylvestris* L. (BRA) and *Pinus nigra* ssp. *laricio* var. *Corsicana* Loud. (RAV). Three other plots are located in deciduous forest: *Fagus sylvatica* L. (WIJ, HOE) and a mixture of *F. sylvatica* L. and *Quercus robur* L. (GON). The soil texture of the plots at RAV and BRA was sand, at WIJ loamy sand and at GON and HOE loam (USDA textural triangle) (Verstraeten et al., 2012). The stands have an age of 82–108 years, a basal area of 29.2–44.9 m^2 ha^{-1} and a very low pH-$CaCl_2$ of 2.5–4.1 in the mineral soil. The five plots are part of the LTER-Europe network (Long-term Ecosystem Research Network).

2.2. Sample collection and measurements

Samples of deposition and soil solution were collected fortnightly from January 2005 till December 2014, according to the guidelines of the ICP Forests manual, part XI and XIV (Clarke et al., 2016; Nieminen et al., 2016). A detailed description of the procedures used for deposition and soil solution sampling can be found in Verstraeten et al. (2012, 2016).

Samples of fresh tree foliage were collected biennially from 1999 till 2013 by professional tree climbers. Samples were always collected from the same five dominant trees in each plot and from the upper third of the crown (needles or leaves that developed in light), according to the guidelines of the ICP Forests manual, part XII (Rautio et al., 2016).

2.3. Chemical analysis

Samples were treated and analysed as prescribed by the ICP Forests manual, part XI, XII and XIV (Clarke et al., 2016; Nieminen et al., 2016; Rautio et al., 2016). Quality control included the analysis of control samples (blanks, reference material, replicates) and participation in the ICP Forests water and foliar ring tests, according to the guidelines of the ICP Forests manual, part XVI (König et al., 2016).

Water samples (500-mL subsamples of each collected fraction) were kept cool during transportation, filtered (0.45 µm), stored in darkness at 4 °C, and analysed within 48 h after sampling. Concentrations of Total Kjeldahl Nitrogen (TKN) (mg L^{-1}) were determined using the continuous flow method (Skalar, limit of quantification, LOQ = 0.5 mg L^{-1}). Concentrations of ammonium (NH_4^+), NO_3^- and nitrite (NO_2^-) (mg L^{-1}) were determined using ion chromatography (Dionex ICS-3000, LOQ = 0.1 mg L^{-1}). Concentrations of DOC were determined using a TOC-analyser (Shimadzu TOC 5050A, LOQ = 0.1 mg L^{-1}).

Foliage samples were dried in an oven at 40 °C until constant weight and pulverised with a suitable mill (Retsch SM 2000). For each of the sampled trees a homogenized subsample was analysed at every sampling event. Foliar N concentrations (mg g^{-1} dry weight at 105 °C) were determined using the Kjeldahl method with NH_4^+-back titration (Gerhardt KB8S, LOQ = 1 mg kg^{-1}). Foliar concentrations of P, Ca, K and Mg (mg g^{-1} dry weight at 105 °C) were determined using ICP-AES (Varian Liberty Series II, LOQ = 50 mg kg^{-1}) after microwave digestion with HNO_3/H_2O_2.

2.4. Data handling

Concentrations of DOC and DON for 2005–2013 were taken from previous studies (Verstraeten et al., 2014, 2016), and were supplemented with new data for 2014. Concentrations of DON were calculated as TKN − NH_4^+. Concentrations of TDN were calculated as TKN + NO_3^- + NO_2^-. The ratio of DON and TDN concentrations (molar), DON:TDN, and the ratio of DOC and NO_3^- concentrations (molar), DOC:NO_3^-, were calculated for each sample for which both concentrations were measured. Deposition fluxes of DIN (kg ha^{-1}) were