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Relevance of canopy drip for the accumulation of nitrogen in moss used as biomonitors for atmospheric nitrogen deposition in Europe

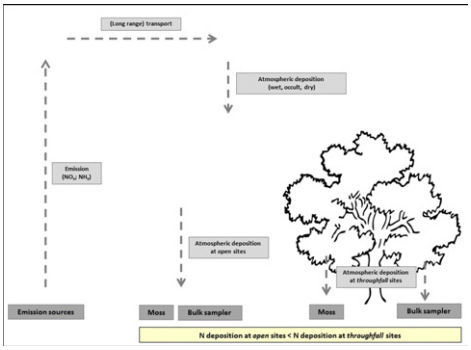
Michaela Meyer [a,*], Winfried Schröder [a], Stefan Nickel [a], Sébastien Leblond [b], Antti-Jussi Lindroos [c], Karsten Mohr [d], Jarmo Poikolainen [e], Jesus Miguel Santamaria [f], Mitja Skudnik [g], Lotti Thöni [h], Burkhard Beudert [i], Helga Dieffenbach-Fries [j], Hubert Schulte-Bisping [k], Harald G. Zechmeister [l]

[a] University of Vechta, Driverstraße 22, 49377 Vechta, Germany
[b] Muséum National d'Histoire Naturelle, 57 rue Cuvier, Case 39, 75005 Paris, France
[c] Natural Resources Institute Finland, P.O. Box 18, FI-01301 Vantaa, Finland
[d] Landwirtschaftskammer Niedersachsen, Mars-la-Tour Str. 1-13, 26121 Oldenburg, Germany
[e] Natural Resources Institute Finland, P.O. Box 413, FI-90014, University of Oulu, Finland
[f] University of Navarra, Irunlarrea No 1, 31008 Pamplona, Spain
[g] Slovenian Forestry Institute, Večna pot 2, 1000 Ljubljana, Slovenia
[h] FUB Research Group for Environmental Monitoring, Alte Jonasstraße 83, CH-8640 Rapperswil, Switzerland
[i] Nationalparkverwaltung Bayerischer Wald, Freyunger Straße 2, 94481 Grafenau, Germany
[j] Umweltbundesamt Deutschland, Paul-Ehrlich-Straße 29, 63225 Langen, Germany
[k] Georg-August University, Büsgenweg 2, 37077 Göttingen, Germany
[l] University of Vienna, CVL, Rennweg 14, 1020 Vienna, Austria

HIGHLIGHTS

- Investigation of nitrogen deposition in forests and (neighbouring) open fields
- Estimation of N contents in moss for non-sampled sites
- Integration of environmental factors potentially influencing N content in moss
- Consideration of spatial variances in vegetation stands in future monitorings

GRAPHICAL ABSTRACT



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ABSTRACT

High atmospheric deposition of nitrogen (N) impacts functions and structures of N limited ecosystems. Due to filtering and related canopy drip effects forests are particularly exposed to N deposition. Up to now, this was proved by many studies using technical deposition samplers but there are only some few studies analysing the canopy drip effect on the accumulation of N in moss and related small scale atmospheric deposition patterns. Therefore, we investigated N deposition and related accumulation of N in forests and in (neighbouring) open fields by use of moss sampled across seven European countries. Sampling and chemical analyses were conducted according to the experimental protocol of the European Moss Survey. The ratios between the measured N content

* Corresponding author.
E-mail addresses: michaela.meyer@uni-vechta.de (M. Meyer), winfried.schroeder@uni-vechta.de (W. Schröder), stefan.nickel@uni-vechta.de (S. Nickel), sleblond@mnhn.fr (S. Leblond), antti.lindroos@luke.fi (A.-J. Lindroos), Karsten.Mohr@LWK-Niedersachsen.de (K. Mohr), jarmo.poikolainen@luke.fi (J. Poikolainen), chusmi@unav.es (J.M. Santamaria), mitja.skudnik@gozdis.si (M. Skudnik), fub@fub-ag.ch (L. Thöni), Burkhard.Beudert@npv-bw.bayern.de (B. Beudert), helga.fries@uba.de (H. Dieffenbach-Fries), Hubert.SchulteBisping@forst.uni-goettingen.de (H. Schulte-Bisping), Harald.Zechmeister@univie.ac.at (H.G. Zechmeister).

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in moss sampled inside and outside of forests were computed and used to calculate estimates for non-sampled sites. Potentially influencing environmental factors were integrated in order to detect their relationships to the N content in moss. The overall average N content measured in moss was 20.0 mg g^{-1} inside and 11.9 mg g^{-1} outside of forests with highest N values in Germany inside of forests. Explaining more than 70% of the variance, the multivariate analyses confirmed that the sampling site category (site with/without canopy drip) showed the strongest correlation with the N content in moss. Spatial variances due to enhanced dry deposition in vegetation stands should be considered in future monitoring and modelling of atmospheric N deposition.

1. Introduction

High atmospheric nitrogen (N) pollution has significant impacts on terrestrial ecosystems. N oxides (NO_x) and ammonia (NH_3^+) are the most important reactive N species (Erisman et al., 2007). Agriculture is largely responsible for the emission of NH_3 while households, road traffic and industry are responsible for the emission of NO_x (EEA, 2014). After emission, air pollutants are transported through the atmosphere and potentially transformed, depending on substance related characteristics, such as particle size, vapour pressure or pH value, and also on meteorological conditions such as wind speed and direction and receptor-dependent structures (Hill, 2010). The subsequent deposition of airborne pollutants takes place via wet (rain, snow), occult (cloud, fog) or dry (particles, gases) deposition (Gauger, 2010; Gauger et al., 2008; Kalina et al., 2002; Unsworth and Wilshaw, 1989). The total deposition includes wet, occult and dry deposition (Schultz-Wildelau and Rost, 1995; Ulrich and Sumner, 1991; VDI, 1983; VDI, 2010). The relationship between dry and wet deposition changes depending on the distance between emission sources and receptors. While dry deposition accounts for most of the total deposition near emission sources, its proportion decreases with increasing distance. In contrast, the amount of wet deposition increases with increasing distance resulting in negative effects even for more remote locations (Hill, 2010).

Ecological effects of atmospheric N pollution are, amongst others, acidification of soil and of waters, eutrophication of ecosystems, climate warming as well as changes in biodiversity and ecosystem integrity (Brandes, 1999; Rockström et al., 2009; Schröder et al., 2015; Sutton et al., 2011; UBA, 2013). According to de Schrijver et al. (2008) and Gauger (2010), forest canopies constitute a considerable sink for atmospheric particles and gases due to their large volume-specific and exposed leaf and woody area. For this reason, N deposition into forest ecosystems should be partitioned into canopy uptake and throughfall deposition onto forest soil (Aber et al., 1998; Wuyts et al., 2015). Tree crowns adsorb gaseous and particulate air pollutants. After admission in the canopy, the deposited material can be removed by rain. Furthermore, nutrients enriched in the leaves or needles can be leached implying an increased N deposition on the forest floor resp. N accumulation in the understory vegetation depending on the forest structure and microclimatic characteristics (de Schrijver et al., 2007; de Schrijver et al., 2008; Reich et al., 2005). Moreover, litterfall provides additional N input to the forest floor (Reich et al., 2005). Consequently, N deposition loads are larger in forests, i.e. beneath the crowns, than in smaller or less structured vegetation units.

Multi-layered ecological interactions and effects of pollutants on the environment can impede the analysis of the amount of atmospheric pollutants and their temporal and spatial distribution (Bobbink et al., 2010; Bobbink and Hettelingh, 2011; Mohr et al., 2005; Pitcairn et al., 2006; UBA, 2011; WGE, 2004). For the evaluation of potential atmospheric pollution and related effects on the environment, the establishment of monitoring networks is of great importance (Harmens et al., 2013; Nagajyoti et al., 2010; Stankovic et al., 2014). The most common technique for estimating atmospheric N fluxes is analysing precipitation sampled outside of and within forests (Draaijers et al., 1997). Precipitation water of open land samplers (bulk sampler collecting wet as well as receptor-specific occult and dry deposition; wet-only sampler) is enriched with elements which are deposited after rain- and washout processes in the atmosphere. The composition of precipitation sampled in forests stands (so-called throughfall) differs from wet deposition (Lindberg et al., 1986). Dry deposited particles on surfaces rinsed by rain or leaching from leaves, needles or microbiota increases element concentrations in rainfall water. On the other hand, adsorption and assimilation of nutrients such as ammonium (NH_4^+) and nitrate (NO_3^-) decreases these elements depending on the nutrient status of the forest stand (Michaelis et al., 1992).

Apart from monitoring networks measuring atmospheric deposition and air quality with physico-chemical methods, the assessment of pollutants is also realized using biological indicators such as moss and lichens (Blagnyté and Paliulis, 2010; Boltersdorf et al., 2014; Frahm, 1998; Harmens et al., 2011; Kaltz et al., 2010; Mohr, 1999). As shown in a large number of studies (Harmens et al., 2014a; Mohr, 2007; Pitcairn et al., 2006; Schröder et al., 2010; Steinnes and Berg, 1997; Stevens et al., 2011; Zechmeister et al., 2008), moss can be used as biomonitors reliably mirroring atmospheric N deposition, including wet, occult and dry deposition due to particular physiological characteristics. However, in contrast to measurements with technical deposition samplers (Adriaenssens et al., 2013; Draaijers et al., 1997; Lindberg et al., 1986; Schmitt et al., 2005), there are only few peer reviewed studies analysing canopy drip effects on the accumulation of N in moss and how this could influence the evaluation of atmospheric deposition patterns (Mohr, 1999, 2007; Skudnik et al., 2014, 2015). Since spatial dense deposition monitoring is rare (Tørseth et al., 2012) but feasible with moss (Blagnyté and Paulis, 2010; Harmens et al., 2013; Tyler, 1990), the key objectives of our study were the following:

Objective 1: Analyses of the relevance of canopy drip for the accumulation of N in moss and for the N concentration in precipitation (dealt with in Sections 2.1, 2.3, 3.2).

Objective 2: Derivation of N contents in moss at non-sampled sites by calculating the N content ratio between open sites and sites influenced by canopy drip (Sections 2.1, 2.3, 3.2).

Objective 3: Analyses the relationship between the modelled N deposition and, both, the measured N content in moss and the calculated N deposition rates based on the measured concentrations in precipitation, respectively (Sections 2.1, 2.3, 2.4.3, 3.4).

Objective 4: Identification of the relevant environmental factors explaining most of the variability in N contents in moss (Sections 2.2, 2.4.4, 3.5).

2. Material and methods

2.1. Moss sampling and chemical analyses

Moss samplings were realised in seven European countries, including Austria (AT), Switzerland (CH), Germany (DE), Spain (ES), Finland (FI), France (FR), and Slovenia (SI) (Fig. 1). In case of DE, the most samplings took place in the northwestern part, with intensively agricultural used and polluted areas (indicated as 'DE-NI' for Lower Saxony). Those