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Nitrogen deposition to forest ecosystems with focus on its different forms

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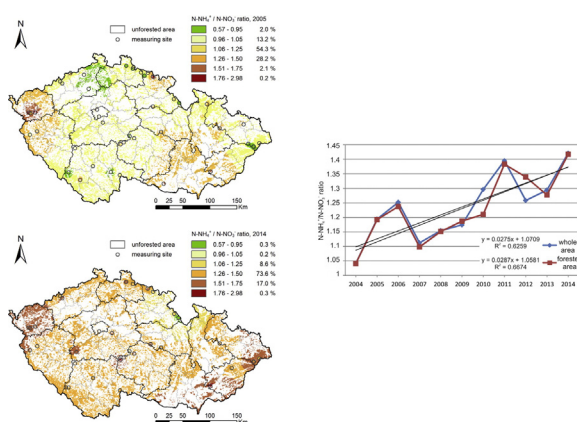
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HIGHLIGHTS

- Ratio of reduced and oxidized N forms in precipitation in Czech forests was studied.
- $\text{N-NH}_4^+/\text{N-NO}_3^-$ ratio in precipitation and wet deposition increases steadily.
- Increased $\text{N-NH}_4^+/\text{N-NO}_3^-$ ratios reflect less reduced emissions of NH_3 than NO_x .
- Increasing $\text{N-NH}_4^+/\text{N-NO}_3^-$ ratios may negatively affect ecosystems.

GRAPHICAL ABSTRACT



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ABSTRACT

Time trends and spatial patterns for wet nitrogen deposition in view of its reduced and oxidized forms for Czech forests are presented. Time trends in NH_4^+ , NO_3^- , and $\text{N-NH}_4^+/\text{N-NO}_3^-$ ratio at 15 sites in 1990–2014 were analysed using the Mann-Kendal test. Changes in spatial patterns derived from results of 43 sites in 2005–2014 were also assessed. Our results indicated a slow but convincing change in favour of NH_4^+ . The $\text{N-NH}_4^+/\text{N-NO}_3^-$ ratio, both in precipitation and wet deposition, has been increasing steadily, reflecting the changes in reduced and oxidized forms of nitrogen emissions. A similar change has been observed in some other regions and might have negative impacts on ecosystems.

Capsule: $\text{N-NH}_4^+/\text{N-NO}_3^-$ ratio in precipitation and deposition changes in favour of NH_4^+ . This reflects changes in emissions and might have implications for ecosystems.

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1. Introduction

Enhanced nitrogen (N) deposition represents one of the most important environmental threads. It is inorganic N deposition, which is usually assessed, though organic N deposition becomes increasingly recognized. In this paper we focus on inorganic N exclusively.

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Inorganic N in the atmosphere is found in oxidized and reduced forms. Oxidized compounds originate mostly from combustion processes, reduced compounds from agriculture, with a non-negligible contribution of natural processes (Erisman et al., 2007). The biogeochemical cycle of N has substantially changed due to human activities in recent decades (Elser, 2011; Fowler et al., 2013; Galloway et al., 2008) with harmful impacts, and negative consequences are anticipated also in the future (Canfield et al., 2010). Currently the contribution to the release of reactive N by humans is comparable to natural (Erisman et al., 2013; Sutton et al., 2011). Though N as an essential nutrient is indispensable for correct functioning of a living cell (Bruce et al., 2014), its elevated deposition is harmful for the environment (Eugster and Haeni, 2013). Sensitivity towards N differs in diverse ecosystems. In a decreasing order of sensitivity to excess N are: native vegetation > forests > agricultural crops (Krupa, 2003). Nitrogen overload might result in important implications for ecosystems, such as eutrophication, acidification, and loss of biodiversity, as has been shown by numerous authors (Bobbink et al., 2010; Dimböck et al., 2014). Though a certain decrease in N deposition has been reported (EEA, 2015), critical loads for N are still exceeded at many sites in Europe (Lorenz et al., 2008). Interestingly, recent papers have reported that it is not just the N load, but rather its form, whether reduced or oxidized, that matters (Britto and Kronzucker, 2013; van den Berg et al., 2016). A shift in favour of reduced N has been reported from different regions (Du et al., 2014; Li et al., 2015; Li et al., 2016) as a consequence of legislation applied recently to improve ambient air quality (EC, 2008). Emission sources regulation resulted in changes in atmospheric chemistry in Europe and elsewhere (Fagerli and Aas, 2008; Hertel et al., 2011; Monks et al., 2009; Rodhe et al., 1981).

Precipitation chemistry has been monitored worldwide since the 1970s in the context of the acidification of ecosystems. Global BAPMON (Background Air Pollution Monitoring Network), regional EMEP (European Monitoring and Evaluation Programme), US NADP/NTN (National Atmospheric Deposition Program/National Trends Network), and CANSAP Canadian Network for Sampling Precipitation, as well as national monitoring networks run by individual countries, provide precipitation chemistry data at different scales.

In the Czech Republic, precipitation chemistry has been measured since the 1970s, and nation-wide monitoring network was established in the 1980s. Some 20 parameters including NH_4^+ and NO_3^- from an ambient air quality monitoring network are regularly analysed. The general time trends and spatial patterns of atmospheric deposition of N have been presented by Hůnová et al. (2004, 2014, 2016a, b). In this work we focus specifically on changes in different forms of N, either reduced or oxidized, and their changing contribution to wet N deposition in a scale of one country for the entire period of monitoring. This information is interesting in particular in view of the possible impacts on ecosystems, as according to the present knowledge, these might be more related to the relative share of N in precipitation than to N load (e.g. BassiriRad, 2015).

For our analysis we used the available data on NH_4^+ and NO_3^- concentrations in precipitation, and studied changes in the $\text{N-NH}_4^+/\text{N-NO}_3^-$ ratio. On the one hand, the $\text{N-NH}_4^+/\text{N-NO}_3^-$ ratio in precipitation reflects the changes in both the absolute quantity and relative contribution of emission sources, pollution climate, and atmospheric chemistry of a specific region, but on the other hand it might also show the impact on ecosystems. The aim of our paper was to explore the long-term data on precipitation chemistry of a region highly polluted in the past and to answer the question of what the time trends and spatial changes are in the $\text{N-NH}_4^+/\text{N-NO}_3^-$ ratio in precipitation and wet deposition in Central Europe, in particular over the Czech forests.

2. Methods

2.1. Data set

The study domain is a Czech forested area, accounting for 34% (26,664 km²) of the Czech Republic territory, dominated by Norway

spruce (*Picea abies*) – at 51% of the forested area, followed by pine trees (*Pinus* spp.) – 17%, beech (*Fagus sylvatica*) – 8%, and oak (*Quercus* spp.) – 7% as the most common species (Ministry of Agriculture, 2015). The data on precipitation chemistry were retrieved from the Ambient Air Quality Database with nation-wide scope run by the Czech Hydro-meteorological Institute (CHMI). For the time trend analysis, we used all wet-only sample results available for the CR, measured at the CHMI sites (Table 1) in the week to month resolution. NO_3^- were analysed by ion chromatography, NH_4^+ by spectrophotometry (flow injection analysis with indophenol, Berthelot reaction) according to CHMU (2015).

2.2. Time trends

Time trends at individual measuring sites were assessed using the Mann-Kendal non-parametric test, recommended by the WMO for this kind of data and used previously in similar studies (Hůnová et al., 2014; Salmi et al., 2002; Sicard et al., 2007). We used software prepared by the Finnish Meteorological Institute (Määttä et al., 2002).

2.3. Spatial patterns

We generated the national map based on two GIS layers: 1: ion concentration in precipitation, 2: precipitation amounts. The final map was obtained by multiplication of these layers.

NH_4^+ and NO_3^- spatial patterns were prepared from 43 sites in total (39 classified as rural and 4 as suburban sites), 37 of these were Czech (CHMI, Czech Geological Survey, Forestry and Game Management Research Institute), and 6 were Polish and German supporting the interpolation in border regions. As only the CHMI sites measure wet-only, while other institutes use bulk samplers, we corrected the bulk data by empirical factors of 0.85 for NO_3^- and 0.74 for NH_4^+ derived from collocated measurements. Maps of ion concentrations were prepared using the Inverse Distance Weighted (IDW) spatial interpolation technique (e.g. Isaaks and Srivastava, 1989). As the precipitation chemistry network was sparse for interpolating purposes taking into account the area of the CR, we run the spatial interpolation in two steps. The values resulting from the first run were extracted to the regular 20 × 20 km point network. In the second run, the interpolation was carried out on newly generated regular network of 1 × 1 km for smoothing purposes. We have already used this approach earlier (Hůnová et al., 2014, 2016b). The map of precipitation amounts was based on measurements of 750 precipitation gauging sites run by the CHMI, using universal

Table 1

Measuring sites for precipitation chemistry run by the CHMI (listed according to increasing altitude).

Site	Region	Classification	Altitude [m a.s.l.]
Ostrava-Poruba	Moravskoslezský	B/S/R	242
Hradec Králové-observatoř	Královéhradecký	B/S/R	276
Praha 4-Libuš	Hl.m. Praha	B/S/R	301
Kuchařovice	Jihomoravský	B/R/A	334
Ústí nad Labem-Kočkov	Ústecký	B/S/RN	367
Kocelovice	Jihočeský	B/R/A	519
Košetice	Vysočina	B/R/N-REG	535
Svratouch	Pardubický	B/R/N-REG	735
Přimda	Plzeňský	B/R/N-REG	740
Červená	Moravsko-slezský	B/R/N-REG	749
Souš	Liberecký	B/R/N-REG	771
Rudolice v Horách	Ústecký	B/R/N-REG	840
Luisino údolí	Královéhradecký	B/R/N-REG	875
Bílý Kříž	Královéhradecký	B/R/N-REG	890
Krkonoše-Rýchory	Královéhradecký	B/R/N-REG	1001

Note: Sites are classified according to EC Decision EoI 97/101/EC (EC 1997): B/S/R - background/suburban/residential; B/S/RN - background/suburban/residential, natural; B/R/A - background/rural/agricultural; B/R/N-REG - background/rural/natural-regional.

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