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Trends in atmospheric deposition fluxes of sulphur and nitrogen in Czech forests

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ABSTRACT

We present the temporal trends and spatial changes of deposition of sulphur and nitrogen in Czech forests based on records from long-term monitoring. A statistically significant trend for sulphur was detected at most of the sites measuring for wet, dry, and total deposition fluxes and at many of these the trend was also present for the period after 2000. The spatial pattern of the changes in sulphur deposition flux between 1995 and 2011 shows the decrease over the entire forested area in a wide range of 18.1–0.2 g m^{-2} $year^{-1}$ with the most pronounced improvement in formerly most impacted regions. Nitrogen still represents a considerable stress in many areas. The value of nitrogen deposition flux of 1 g m^{-2} $year^{-1}$ is exceeded over a significant portion of the country. On an equivalent basis, the ion ratios of NO_3^-/SO_4^{2-} and NH_4^+/SO_4^{2-} in precipitation show significantly increasing trends in time similarly to those of pH.

1. Introduction

Atmospheric deposition has decreased in the recent two decades in Europe (EEA, 2011) substantially due to a combination of several reasons as being:

- More stringent legislation with consequent implementation of effective countermeasures at large sources and decreasing emission of sulphur dioxide (SO_2) and to lesser extent of oxides of nitrogen (NO_x),
- Restructuring of industry and economic activities after profound political and economic changes in the 1990's, and
- Economic crises of recent years.

Nevertheless, the deposition values in many European regions still remain far from satisfactory (EEA, 2011). The long-term monitoring of precipitation chemistry and ambient air pollution is essential for quantification of both wet and dry deposition and revealing the time trends and spatial patterns under major environmental and climate change, and to link these with potential environmental impacts (Skeffington and Hill, 2012). In contrast to many other European countries, the national network for monitoring of ambient air quality and chemical composition of precipitation over the CR has operated for a fairly long time (Hůnová, 2001). The reason is that ambient air quality belonged to the most prominent environmental issues in the former Czechoslovakia and was negatively perceived by the public (Moldan and Schnoor, 1992). It was considered an important factor contributing essentially to forest dieback (Fanta, 1997), and also had the most negative consequences for human health which manifested, for example, in life expectancy decrease (Bobak and Leon, 1992).

Sulphur dioxide (SO_2) as the first pollutant started to be measured in ambient air in the CR in 1960s. The oldest records of precipitation chemistry date back to 1978. The major emission source of SO_2 was the combustion of poor local lignite with high sulphur content. The daily mean concentrations of SO_2 in the Czech part of the so called "Black Triangle" reached very high values, e.g. 1600 μg m^{-3} in January 1982 (CHMI unpublished data). Annual means reached hundreds of μg m^{-3} in the most polluted areas (Bridgman et al., 2002).

Severe air pollution, ranking the former communist Czechoslovakia in the most polluted European countries (Moldan and Schnoor, 1992), has decreased substantially. Profound socio-economic changes in Central Europe in 1989 resulted in significant improvement of many environmental indicators, including industrial emissions. The trend in SO_2, NO_x and NH_3 emissions from the Czech sources is shown in Fig. 1.

The aim of the paper is to present the temporal trends and spatial patterns of sulphur and nitrogen deposition fluxes in Czech mountain forests for the entire period between measurement commencement

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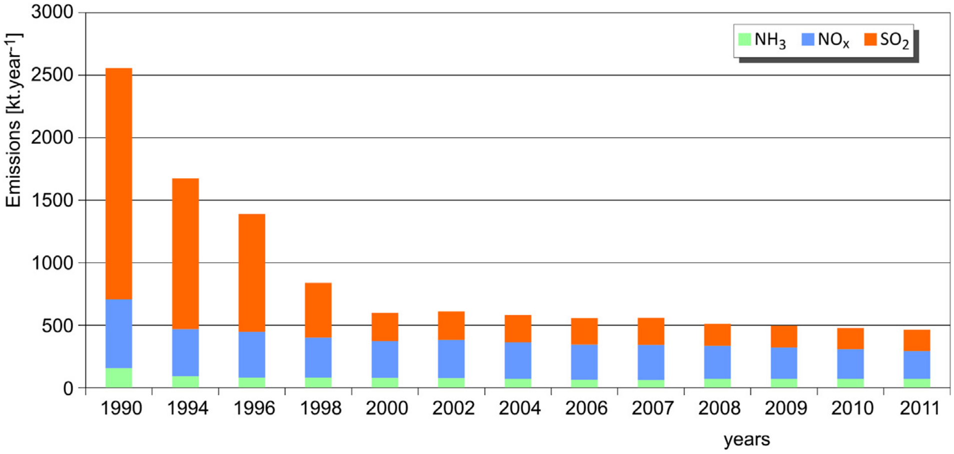

Fig. 1. SO_2, NO_x and NH_3 emissions from Czech emission sources, 1990–2011.

and 2011. The trends of sulphur and nitrogen deposition for values measured until 2001 at selected Czech rural sites operated by the CHMI have already been published (Hůnová et al., 2004). Nevertheless, generally high inter-annual variations in meteorology can confound attempts to detect trends in deposition through measurement. Long term data series of wet deposition of some two decades are therefore required to measure deposition trends (Matejko et al., 2009). For approximation of the most recent deposition trends, however, we also separately checked the data after 2000.

2. Methods

2.1. Atmospheric deposition of sulphur and nitrogen

For our analysis, we used the data from 15 Czech sites (Table 1) operated by the Czech Hydrometeorological Institute collected in a nation-wide ambient air quality database (Ostatnická, 2012). The sites are situated as far away as possible and practical from local sources to meet a demand for regional representativeness. Total deposition was calculated as a sum of wet and dry deposition fluxes. The evolution of the atmospheric deposition measurements over the Czech rural area, the beginning of precipitation composition measurements at individual sites and accessibility of ambient air pollution concentration for dry deposition flux estimation is summarized in Table 2.

Wet deposition was calculated based on automated wet-only samples on weekly basis, analysed by standard methods with comprehensive QA/QC procedures, described in detail by Hůnová et al. (2011). Dry deposition was estimated using the inferential method, combining measurements and modelling. This method, recommended by Fowler et al. (2009), is based on the assumption of a steady-state relationship $F = c.\ v_d$, where the dry deposition flux (F) is a product of the mean concentration (c) and of the dry deposition velocity (v_d). Dry deposition velocities used were as follows: for SO_2: $v_d = 0.7$ cm s^{-1} for forest (0.35 cm s^{-1} for areas outside the forest), for NO_x: $v_d = 0.4$ cm s^{-1} for forest (0.1 cm s^{-1} for areas outside the forest), for NH_3: $v_d = 0.66$ cm s^{-1} for forest (0.44 cm s^{-1} for areas outside the forest). Deposition velocities for SO_2, NO_x and NH_3 were calculated using the resistance analogy based on meteorological and land use data for the CR (Dvorakova et al., 1995; Fiala et al., 2001). Annual mean concentrations of SO_2, NO_x and NH_3 were used as recorded at the sites. If these were not available, we used an expert estimation based on the measurements from the neighbouring sites of the same type according to EoI classification (EC, 1997). For SO_2 direct measurements were available for 12 out of 15 sites. For NO_x, the availability of direct measurements was much poorer. Ambient NH_3 concentrations were measured only at three sites in total over the CR. After rejection of one site which represented an area highly impacted by chemical industry, we used the mean calculated from the recorded values of the two remaining sites as a rough approximation for all sites in the CR. To summarize, total sulphur deposition was calculated as a sum of dry S–SO_2 and wet S–SO_4^{2-} deposition. Total nitrogen deposition was calculated as a sum of dry N–NO_x, N–NH_3, and wet N–NO_3^- and N–NH_4^+ deposition.

In the dry deposition calculation we did not account for particulate S–SO_4^{2-}, and N–NO_3^-, neither for N–HNO_3(g) due to the unavailability of measured data. Currently, particulate N–NO_3^- and gaseous HNO_3 is not measured in the CR. For particulate S–SO_4^{2-} the only data available are from three sites (two rural and one city-background). According to these records and using the $v_d = 0.25$ cm s^{-1} (Fiala et al., 2001) we estimated the annual mean particulate S–SO_4^{2-} deposition to be 0.18 g m^{-2} $year^{-1}$. When compared to the total deposition, this portion can reasonably be considered negligible.

We also did not account for occult deposition as the data is not available.

Table 1
Stations used for the analysis ranked according to increasing altitude.

Site	Characterization	Altitude [m a.s.l.]
Ostrava-Poruba	Suburban site	242
Hradec Králové	Suburban site	276
Praha-Libuš	Suburban site	301
Kuchařovice	Agricultural area, south Moravia	334
Ústí n.L.-Kočkov	Suburban site	367
Kocelovice	Agricultural area, Středočeská hilly area	519
Košetice	Agricultural area, Czech-Moravian Highlands	535
Svratouch	Agricultural area, Žďárské vrchy	735
Přimda	Český Les	740
Červená	Nízký Jeseník	749
Souš	Jizerské hory Mts.	771
Rudolice v Horách	Mountain site, Krušné hoty Mts.	840
Luisino údolí	Mountain site, Orlické hory Mts.	875
Bílý Kříž	Mountain site, Moravsko-Slezské Beskydy Mts.	890
Krkonoše-Rýchory	Mountain site, Krkonoše Mts.	1001

2.2. Temporal trends

Different methods are used for detecting trends in atmospheric deposition (Marchetto et al., 2013). We analysed the data for temporal trends using the Mann–Kendall non-parametric test, recommended by the WMO for this kind of data and used in similar studies (Salmi et al., 2002; Sicard et al., 2007; Bashir et al., 2008). This test is used to identify the monotonous trend in data-series which does not show any seasonal variation or cycles. Atmospheric chemistry data, however, usually have distinct seasonal variability and the Mann–Kendall test is thus applied to annual values to overcome the problem of seasonality and autocorrelation. Non-parametric methods generally have the advantage of being insensitive to outliers, missing values are allowed, and data do not need to be normally distributed (Gilbert, 1987; Salmi et al., 2002), which was appropriate for the analysed data set. For calculation we used software produced by the Finnish Meteorological Institute (Määttä et al., 2002). We analysed both nitrogen and sulphur deposition with respect to contribution of wet and dry deposition fluxes, and additionally the wet deposition of H^+. Apart from deposition fluxes, we also checked the ratios of major ions in precipitation – nitrate to sulphate, and ammonia to sulphate – on an equivalent basis. All trends were detected for the entire period of measurement which differed among individual sites (see Table 2).

2.3. Spatial patterns

A fine resolution maps were developed to study the spatial patterns of atmospheric deposition. For construction of maps of total deposition we used all sites