ORIGINAL PAPER



Atmospheric deposition of sulphur and nitrogen in forests of the Czech and Slovak Republic

Danica Krupová¹*, Věra Fadrhonsová², Hana Pavlendová¹, Pavel Pavlenda¹, Slávka Tóthová¹, Vít Šrámek²

¹National Forest Centre - Forest Research Institute Zvolen, T. G. Masaryka 2175/22, SK – 960 92 Zvolen, Slovak Republic ²Forestry and Game Management Research Institute, Strnady 136, CZ – 252 02 Jíloviště, Czech Republic

Abstract

Spatial and temporal variation of atmospheric depositions on the permanent monitoring plots of Level II of the ICP Forest programme in Czech and Slovak forest areas was described in this paper. The atmospheric bulk deposition of sulphur, ammonium and nitrate nitrogen from the years 2000–2016 were assessed and compared with the data obtained on European plots of this programme. The temporal developments of annual depositions in kg.ha⁻¹.year⁻¹ were determined by linear regression and displayed through maps. The most significant change during the reporting period was in the annual sulphur deposition, which was the statistically significant with rather sharp decrease on Slovak plots ($P \le 0.01$, $R^2 = 0.68$) and with milder decrease in the Czech Republic ($P \le 0.05$, $R^2 = 0.23$). There are small spatial and temporal differences in the deposition of the annual nitrate nitrogen depositions. The decrease was statistically more significant in Slovakia ($P \le 0.01$, $R^2 = 0.61$, $R^2 = 0.61$, $R^2 = 0.63$) and with milder decrease in the Czech Republic ($P \le 0.05$, $R^2 = 0.30$). The smallest changes were in annual depositions of ammonium nitrate, although the deposition was statistically significant in Slovakia ($P \le 0.01$, $R^2 = 0.42$). In the Czech Republic dropped only slightly over the whole territory (except in the Beskydy Mts.) during evaluated period. The temporal variations of polluting components revealed significant differences between developments of depositions on the different plots of both republics. The sulphur and nitrogen depositions in the Slovak and Czech Republic persistently ranking among the highest in Europe, what is caused by local and transboundary anthropogenic emission.

Key words: atmospheric deposition; sulphur; ammonia nitrogen; nitrate nitrogen; monitoring of forests

Editor: Milan Barna

1. Introduction

The atmosphere is a very complex system containing a wide range of natural compounds and anthropogenic pollutants that contribute to numerous air quality problems affecting the climate, human health and the environment (EEA 2012). The deposition of acidifying and eutrophying substances had been a major cause of disturbance to forest ecosystems in the northern hemisphere for several decades (de Vries et al. 2014). In the second half of the 20th century, acidic air pollution was a serious concern across most of the Europe (Stern 2005). The acidification and eutrophication of ecosystems have mainly been observed in the Europe and the North America in the past three decades (Pardo et al. 2011), and more recently in the Asia (Zhao et al. 2009). The increase in global emissions, mainly sulphur dioxide (SO₂) and nitrogen oxides (NOx), was unprecedented after World War II due to economic expansion. The SO₂ and NO₂ emissions in the Czech Republic increased sharply between 1950s and 1980s (Kolář et al. 2015).

The precipitation is the main factor affecting the quality of the environment (Thalman et al. 2002). Their composition is a result of many processes and chemical reactions associated with natural phenomena and emission of pollutants (Walna & Kurzyca 2007). If there are sufficient amounts of cations in the air, sulphide and nitrate anions are neutralized by the formation of salts. However, in the case of the excess of anions, the strong acids (sulphuric and nitric) are formed. Those acids, as precipitation fall down, may cause acidification of ecosystems. The anthropogenic pollution is formed mainly by sulphur emissions in the form of sulphur dioxide (SO₂) and nitrogen in the oxidized form (NOx) and reduced form (ammonia – NH₃).

Chemical content of atmospheric deposition has severe harmful effects on the environment by storage of

^{*}Corresponding author. Danica Krupová, e-mail: krupova@nlcsk.org

acidifying and eutrophying compounds, which generate nutrient imbalances and changes in biodiversity (deVries et al. 2014). The acidification of forest soils is caused by washing out of salts of strong acids (SO $_4^{2-}$ and NO $_3^{-}$) and basic elements from the soil (Ulrich et al. 1980). Consequently, the release of Al³⁺ ions from sorption complex into the soil solution causes toxic effects on the roots of plants and has a negative effect on mycorrhiza. On the other hand, the nitrogen is one of the main nutrients in the ecosystem. Its increased load causes the increase of the tree growth. It is also connected with the disturbance of the balance between other nutrients in soil, which have impact on consequent increased susceptibility to frost damage and biotic pests and also the changes in ground biomass composition and thus carbon sequestration (Bontemps et al. 2011; de Vries et al. 2014). The biogeochemical cycle of nitrogen has substantially changed due to the human activities mainly in recent decades (Elser 2011) with harmful impacts and negative consequences which are anticipated also in the future (Canfield et al. 2010).

The objective of this study was to assess nitrogen $(N-NO_3^{-} \text{ and } N-NH_4^{+})$ and sulphur $(S-SO_4^{2^-})$ in atmospheric bulk deposition in Czech and Slovak forest areas, to compare results with deposition in Europe and to evaluate the temporal variations of these compounds on the territory of both republics. The trends for all three parameters were tested by linear regression.

2. Material and methods

2.1. Study areas

The study was conducted on 14 Level II plots (Table 1) that belong to the forest monitoring network of permanent monitoring plots (PMP) of Level II of the Slovakia

(SK) and the Czech Republic (CZ). The national forest monitoring programmes are the part of the International Co-operative Programme on Assessment and Monitoring of Air Pollution effects on Forests (ICP-Forests). The plots were located in mature spruce, oak, pine, fir and beech stands in numbers reflecting the tree species composition of the forest stands in Slovakia and the Czech Republic.

2.2. Data collection and data analyses

Samples were collected and processed according to the Manual ICP Forests "Part XIV. Sampling and Analysis of Deposition, Manuals on methods and criteria for Harmonised sampling, assessment, monitoring and analysis of the effects of air pollution on forests", issued by the UN/ ECE ICP Forests Programme Co-ordinating Centre in Hamburg (Clarke et al. 2010). The data of analyses were collected from seven plots in both Slovakia and the Czech Republic during the period of 2000–2016. Samples were taken three times per month (10 days sampling interval) in the Czech Republic and in regular two-week intervals throughout the year in Slovakia.

According to the methodology of ICP Forests, the deposition samples are collected on Level II plots in the open field (bulk deposition) and under forest canopy (throughfall). Whereas bulk deposition is a basis for estimates of total atmospheric deposition rates in open fields, throughfall deposition differs from bulk deposition due to wash off of dry deposition from the forest canopy, "leaching" of the element from the tree crowns, and absorption of elements by the foliage, so-called "canopy uptake". Thus, throughfall deposition does not reflect total deposition but reflects the results of total deposition plus net canopy exchange (Fischer et al. 2010). Only bulk data has been evaluated in this study, because they are not influ-

Table 1. Description of permanent monitoring plots of Level II in the Czech Republic and Slovakia.

Code	Plot	Tree species	Age [years]	Altitude [m a.s.l.]	Soil type (FAO)	Precipitation [mm.year ⁻¹]	Mean temperature [°C]	Site
SK 201	Čifáre	Sessile oak	97	225	Haplic Luvisol	612	9.3	Polygonato latifolii Carpinetum
SK 203	Lomnistá dolina	Norway spruce, E. beech, S. maple	72	1250	Skeletic Umbrisol	1438	2.5	Vaccinio myrtilli Piceetum
SK 204	Poľana	E. beech, N. spruce, S. fir, E.ash, S. maple	90-120	850	Andic Cambisol	886	5.0	Asperulo odoratae Fagetum
SK 206	Turová	European beech, Oak	70	575	Eutric Cambisol	853	5.8	Dentario bulbiferae Fagetum
SK 207	Tatranská Lomnica	N. Spruce, E. larch, Scots pine, Silver fir,	60–140	1150	Dystric Hyperskeletic Leptosol	1207	2.8	Vaccinio myrtilli Piceetum
SK 208	Svetlice	E. beech, E. larch, Oak, S. Pine	53	570	Haplic Cambisol	994	6.1	Dentario glandulosae Fagetum
SK 209	Grónik	Norway spruce	94	875	Albic Podzol	1129	3.5	Avenello Piceetum excelsae
CZ 521	Lazy	Norway spruce	120	875	Dystric Cambisol	865	7.0	Luzulo Fagetum montanum
CZ 2061	Benešovice	Scots Pine	100	530	Haplic Cambisol	690	8.4	Vaccinio vitis-idaee Quercetum
CZ 2103	Všeteč	European beech	123	615	Epidystric Cambisol	680	8.3	Deschampsio flexuosae Abietinum
CZ 2161	Želivka	Norway spruce	115	440	Entri-Stagnic Cambisol	670	9.1	Luzulo Fagetum
CZ 2251	Luisino údolí	Norway spruce	104	940	Haplic Podzol	1250	4.9	Calamagrostio villosae Piceetum
CZ 2361	Buchlovice	European beech	117	350	Endoeutric Stagnic Cambisol	550	9.0	Carici-Pilosae Fagetum
CZ 2401	Klepačka	Norway spruce	94	650	Stagnic Dystric Cambisol	1300	4.5	Luzulo Fagetum

enced by the exchange processes within the tree canopies. Samples of precipitation water were taken from the samplers located on the open sites next to monitoring plots. The average sample of each sampling has been formed by proportional mixing of samples from 3 samplers on each plot. Monthly volume-weighted samples were pooled 3 sampling periods in the Czech Republic and respectively 2 sampling periods in Slovakia.

The chemical analyses were carried out in forestry laboratories. Namely, Central chemical laboratory of National Forest Centre Zvolen (SK) and Analytical laboratory of Forestry and Game Management Research Institute Prague (CZ). All analyses were conducted according to appropriate methods ISO, ČSN and STN for water quality obligatory used in programme ICP Forests. In separate samples after filtration through 0.45 µm membrane filters, the concentrations of sulphur and nitrate anions were determined by the method of ion chromatography with suppression (STN EN ISO 10304-2, ČSN EN ISO 10304-1). The content of N–NH $_4^+$ was determined by indophenol method in water (STN ISO 7150-1, ČSN ISO 7160-2). The concentrations (in mg.l⁻¹) were determined on the calibrated instruments, N-NH⁺ on PHARO Spectrometer 300, respectively Skalar Spectrometer. DIONEX IC CS-1000 was used for determination of $N-NO_3^-$ and $S-SO_4^{2-}$.

This data based on total rainfall in the measurement interval on the observed area were converted into the deposition values in kg.ha⁻¹ for one month according to the following equation:

Dx = (Cx. M) / 100

where: Dx – monthly deposition of ion in kg.ha⁻¹, Cx – concentration of the element in mg.l⁻¹, M – rainfall in mm.

The sum of monthly depositions (Dx) gives the deposition for corresponding year (yearly deposition) in kg.ha⁻¹.year⁻¹

The map outputs were modelled in ArcGis[©] by combination of two raster layers, regression of deposition to altitude and deviation from this value. DEM (digital elevation model) for the Czech and Slovak Republic was provided and processed by [©] GISAT (2007). Deviations were interpolated by using IDW technique (inverse distance weighted technique). After an overlay of these raster datasets, the resulting layer was cropped using a forest mask obtained from CORINE Land Cover (CLC2012) inventory.

Generally, the data obtained from the European territory within the ICP Forests programme, have been evaluated continuously in Technical Reports (TR) of ICP Forests. The spatial variations of depositions in TR have been determined as the arithmetic mean of the relevant period. Data in this work were compared with mean of bulk deposition from 219 European monitoring plots in period 2002–2004 (Lorenz et al. 2007). The values $4.5 \text{ kg.ha}^{-1}.\text{year}^{-1}$ for N–NO₃⁻, $5.1 \text{ kg.ha}^{-1}.\text{year}^{-1}$ for N–NH₄⁺ and $5.7 \text{ kg.ha}^{-1}.\text{year}^{-1}$ for S–SO₄²⁻ depositions had been used for the evaluation of spatial variation of bulk depositions in Europe in TR (2007–2013) as well as for evaluation of Czech and Slovak data (2000–2016).

3. Results

3.1. Annual depositions of sulphur and nitrogen

Atmospheric depositions, which are collected in open area, mostly reflect the properties of the atmosphere, including secondary dust. The statistical values of the annual bulk depositions of nitrogen and sulphur on PMPs of Level II are in Table 2 and 3.

Annual depositions of N–NO₃⁻ exceeded the reference value of 4.5 kg.ha⁻¹.year⁻¹ in 18% of data measured in Slovakia, respectively in 35% data measured in the Czech Republic in the period of years 2000–2016. High values had been repeatedly occurring on two plots Lazy and Luisino údolí (CZ), where the value 4.5 kg.ha⁻¹.year⁻¹ had been continuously exceeded (over 70% of all data). On the other hand, this value was not reached during this study never once on PMPs Čifáre, Poľana (SK), Všeteč, Buchlovice and Benešovice (CZ). The highest annual depositions were found on the PMP Grónik 8.3 kg.ha⁻¹.year⁻¹ (SK) in 2006 and on the PMP Luisino údolí 9.3 kg.ha⁻¹.year⁻¹ (CZ) in 2005.

Table 2. Statistical values of annual deposition of $N-NO_3^-$ and $N-NH_4^+$ on PMPs of Level II on the territory of the Slovak and the Czech Republic in the period 2000–2016.

Dista	N	-NO3 [kg.ha ⁻¹ .year	-1]	N	-NH4 [kg.ha ⁻¹ .year	-1	Period
Plots	mean	min	max	mean	min	max	[years]
Čifáre	2.7	2.1	4.1	6.6	3.4	14.6	17
Lomnistá	4.3	3.0	7.7	5.8	3.2	13.2	17
Poľana	3.1	2.0	3.9	4.7	2.8	8.4	17
Turová	3.3	1.8	4.7	6.1	2.9	10.7	17
Tatranská Lomnica	3.4	2.0	5.7	5.3	2.6	9.0	11
Svetlice	4.2	2.7	6.3	7.2	4.0	15.4	17
Grónik	4.5	2.4	8.4	6.3	3.6	10.1	13
Lazy	5.1	3.7	7.2	9.6	5.4	14.8	17
Benešovice	3.2	2.1	4.5	3.9	3.1	5.6	12
Všeteč	2.9	1.6	3.9	4.1	1.9	6.6	17
Želivka	4.5	2.6	7.3	5.3	0.8	8.0	17
Luisino údolí	5.8	4.2	9.3	8.5	5.9	12.8	13
Buchlovice	3.2	1.6	5.3	4.7	2.5	7.8	17
Klepačka	4.7		7.0	5.3		8.2	12

The average depositions of $N-NH_4^+$ on the most plots were nearly 1.5 times higher than $N-NO_3^-$ depositions. Despite of this fact, there are three plots which had similar load of both forms of nitrogen Želivka, Benešov, Klepačka (CZ). Value 5.1 kg.ha⁻¹.year⁻¹ was exceeded in 57% of all data from SK, respectively in 53% of data from CZ. PMP Lazy and Luisino údolí had all data higher than this reference value.

The highest depositions of $N-NH_4^+$ was 5.4 kg.ha⁻¹. year⁻¹ on PMP Svetlice (SK) in 2000 and 14.8 kg.ha⁻¹. year⁻¹ on PMP Lazy (CZ) in 2014.

Table 3. Statistical values of annual depositions of $S-SO_4^{2-}$ on PMPs of Level II in the Slovak and the Czech Republic in the period 2000–2016.

Diata	S-SC	04 ²⁻ [kg.ha ⁻¹ .ye	ear ⁻¹]	Period	
Plots	mean	min	max	[years]	
Čifáre	4.9	3.0	9.1	17	
Lomnistá	7.8	4.1	14.1	17	
Poľana	5.8	4.0	10.6	17	
Turová	6.3	3.9	13.8	17	
Tatranská Lomnica	7.3	4.7	13.9	11	
Svetlice	7.6	5.0	14.3	17	
Grónik	7.9	5.2	12.9	13	
Lazy	5.2	2.8	7.5	17	
Benešovice	3.7	1.8	5.3	12	
Všeteč	3.7	2.4	5.3	17	
Želivka	4.6	2.2	7.8	17	
Luisino údolí	6.8	4.1	15.7	13	
Buchlovice	4.9	2.8	7.8	17	
Klepačka	6.7	4.7	10.2	12	

57% of measured data (SK), respectively 27% (CZ) of annual S–SO₄^{2–} depositions were higher than the value 5.7 kg.ha⁻¹.year⁻¹. Till 2010, S–SO₄^{2–} depositions were higher in nearly 90 % of all data on PMPs Lomnistá, Grónik, Svetlice, Tatranská Lomnica in SK. The highest depositions of S–SO₄^{2–} were found on the PMP Luisino údolí (CZ) in 2005 (15.7 kg S.ha⁻¹.year⁻¹) and on the PMP Svetlice (SK) in 2002 (14.3 kg S.ha⁻¹.year⁻¹).

3.2. The development of annual deposition of sulphur and nitrogen

The results of linear regression are described in Table 4.

Table 4. The results of linear regression of two forms of nitrogen and $S-SO_4^{2-}$ deposition in the Czech and the Slovak Republic on PMPs of II. Level in the period 2000–2016.

Elements	Country	b_0	\mathbb{R}^2	P-Value	n
N NO -	CZ	-0.065	0.30	0.019	109
$N-NO_3$	SK	-0.077	0.53	0.000	105
N NUL+	CZ	-0.035	0.124	0.133	109
N-NH ₄	SK	-0.215	0.415	0.005	105
0.002-	CZ	-0.103	0.231	0.045	109
5-5042	SK	-0.365	0.681	0.000	105

Note: Statistically significant results are highlighted in bold (P \leq 0.05) and colour highlighted (P \leq 0.01).

The results of linear regression (Table 4) show higher significance of the temporal trends in sulphur and nitrogen deposition in bulk deposition of Slovakia.

Deposition of N-NO₃ in kg⁻¹year





Fig. 1. Trends of annual deposition of $(N-NO_3^{-} \text{ and } N-NH_4^{+})$ nitrogen and sulphur in the period 2000–2016 evaluated by linear regression.

All parameters (N–NO₃⁻, N–NH₄⁺, S–SO₄²⁻) show statistically significant decrease (P \leq 0.01) in SK. Milder decrease of N–NO₃⁻ and S–SO₄²⁻ annual depositions was determined in the Czech Republic, but was statistically significant (P \leq 0.05). Annual deposition of N–NH₄⁺ in the Czech Republic were dropped only slightly without statistically-significant. The most considerable decrease was observed in development of annual S–SO₄²⁻ deposition in the SK (b₀=-0.365, R²=0.684, P \leq 0.01).

It is clear from the maps created by the model that the most significant change during the reporting period was in the annual $S-SO_4^{2-}$ deposition. The decrease is more pronounced in Slovakia, resp. in the Carpathians. The relatively unfavourable situation is in the northern Bohemia (in Silesia) and in the majority of Slovakia (with the exception of southwestern SK). There are no significant spatial and temporal differences in the deposition of N-NO₃⁻. However, a pronounced decrease is also



Fig. 2. Mean annual sulphate sulphur $(S-SO_4^{2-})$ bulk deposition in years 2000–2005, 2006–2010 and 2011–2015.

D. Krupová et al. / Cent. Eur. For. J. 64 (2018) 249–256



Fig. 3. Mean annual nitrate nitrogen (N–NO $_3^-$) bulk deposition in years 2000–2005, 2006–2010 and 2011–2015.



Fig. 4. Mean annual ammonium nitrogen $(N-NH_4^+)$ bulk deposition in years 2000–2005, 2006–2010 and 2011–2015.

in Slovakia. $N-NH_4^+$ deposition in the Czech Republic dropped only slightly over the whole territory (except in the Beskydy Mts.), while in the whole Slovak territory the decrease was more significant, but again with a slight increase in the last evaluated period.

4. Discussion

4.1. Changes in emissions

The majority of European countries have reduced their emissions mainly thanks to the Convention on Long-Range Transboundary Air Pollution (CLRTAP) and the following protocols. Relatively significant decrease of NO_x emissions was recorded during 2000–2015 in the CZ from 291 305 to 164 406 t.year⁻¹ (CHMU 2015) and also in SK from 63 000 to 26 000 t.year⁻¹ during 2000–2017 (SHMU 2017). In the CZ; 33% of NO_x emissions were formed due to public electricity and heat production, 20% due to road transport, and 19% due to agriculture, forestry, fishing, off-road vehicles and other machinery (Dědina, 2013). Regarding the road transport the highest NOx emission are observed along the highways, major roads, and in the big cities.

On the other hand the decreasing trend in NH_3 emissions (from 84 937 to 69 716 t.year⁻¹) was much milder in CZ in this period (CHMU 2015). In SK, emissions of NH_3 declined from 8 500 to 4 000 t.year⁻¹ between 2000 -2017, mainly as the results of the reduction of the agriculture (SHMU 2017). The decrease of NH_3 emissions was recorded from livestock and poultry farming. However, the increase in NH_3 emissions was noticed from inorganic N-fertilizers. The increase in NH_3 emissions might be influenced also by the increase of the number of cars equipped with catalytic converters, and the introdu ction of denitrification technologies at stationary sources (Dědina 2013).

The greater decrease of NO_x emissions compared to NH₃ emissions is due to much better legislation concerning NO_x. Indeed, to reduce the emissions from industry and road transport seems to be easier than reduction of emissions from the agriculture (EC, 2013). Walaszek et al. (2013) pointed out, that the changes in emissions reduced nitrogen (NH_x) are smaller and further reductions will be difficult to implement. Because the main source of this matter is the agriculture (both animal husbandry and crop cultivation), therefore abatements of those would carry significant economic costs.

The most outstanding decrease in SOx emissions was recorded in the SK (from $110\,038$ to $24\,749$ t.year⁻¹) during the years 2000–2016 (SHMU 2017). The decline was found also in the CZ (from 215 000 to 125 000 t.year⁻¹) between 2007–2015 (CHMU 2015).

Lorenz et al. (2008) pointed that nitrogen emissions as well as the deposition of nitrogen compounds have decreased relatively little compared to sulphur since the 1990s. Emissions of SO_2 and NO_x have dropped significantly due to technological development in industry and reducing fuels from coal (Lawrence et al. 2007). The decline of SOx emissions was reached by modernization of industrial enterprises mainly by reducing sulphur content in fuels for power stations and their reconstruction with filters depicting sulphur (Thöni et al. 2008).

4.2. The development of deposition

In the past the changes of evaluated matter were higher. During the years 1980–2009 total S–SO₄^{2–} depositions dropped by 45% and N–NO₃[–] deposition was reduced 22%.on European PMP. On the contrary, depositions of N–NH₄⁺ showed only 4% decrease (Vestreng et al. 2007). This fact was depicted also in the significant decrease of sulphur on the most plots of ICP Forests, but only a certain decrease in nitrogen (N) deposition was noticed (EEA 2015).

In the years 2000–2010 total N deposition decreased by only 2% per year at ICP Forests European Level II plots, (Waldner et al. 2014), but critical load exceedances are still significant at the many sites in Europe (EEA 2015).

Hůnová et al. (2014) also described the milder decrease in the long term N emissions over the Czech forests as much as no trends in N deposition. The evaluation of data from all European ICP Forests plots Level II confirmed no significant changes in the development of N–NO₃⁻ deposition. The effect of the increasing traffic and also long-range transfer may cause permanently high deposition of this parameter (Lorenz et al. 2009).

Data averaging from 2005–2007 show bulk N–NO₃⁻ deposition in amount over 4.5 kg.ha⁻¹. year⁻¹ on 22.6% of the studied plots. Data from the CZ and the SK of this period confirmed higher value on the 50% of plots. The highest depositions occurred on the plots in Central Europe, in the south of France, north of Italy, and Spain (Fischer et al. 2010).

The bulk N–NH₄⁺ deposition in Europe are comparable with the data in our study. The deposition higher than value 5.1 kg.ha⁻¹.year⁻¹ was determined in the Europe on the 46.5% of plots and on 50% of plots in CZ and SK. Higher deposition occurred on the plots in the Central Europe and also in the south of the France, north of the Italy, and the Belgium. The highest N–NH₄⁺ deposition in Europe in the period 2005–2007 was 35.7 kg.ha⁻¹.year⁻¹ (Fischer et al. 2010). Significantly decreasing trends have been observed on smaller number of European plots, especially for nitrogen compounds. Actually, there were plots with significant increasing trends, especially in the period 2006–2011. However, the monitoring results reflect probably mostly regional patterns and trends, and regional industrial air pollution (Michel et al. 2014).

The values of bulk $S-SO_4^{2-}$ deposition in the Europe, which were calculated as the arithmetic mean of this deposition from the years 2005–2007, were higher than

5.7 kg.ha⁻¹.year⁻¹ on nearly one third (33.7%) of the European plots, but up to on 50% of SK and CZ plots. Generally most plots with high sulphur deposition were located in the central Europe (Poland, Germany, CZ, SK, Slovenia and Romania). The highest deposition of $S-SO_4^{2-}$ in Europe was 32.4 kg.ha⁻¹.year⁻¹ (Fischer et al. 2010).

High values of annual sulphur deposition were repeatedly measured on SK plots with higher altitudes. The deposition of 54.37 kg.ha⁻¹.year⁻¹ was measured in 2001 on the Predná Poľana (Blihárová & Škvarenina 2002). During the period of 2000-2002 Sitkova (2006) identified high annual bulk deposition of $S-SO_4^{2-}$ on the areas of TANAP within an altitude of 1 038 to 1 365 m (from 31.5 to 61.4 kg.ha⁻¹.year⁻¹).

The lowest depositions for all evaluated pollutant are mainly located in the Nordic countries and in the Alps. The lowest $S-SO_4^{2-}$ depositions ranges from 1 to 3.3 kg.ha⁻¹.year⁻¹; the lowest $N-NO_3^{-}$ depositions from 0.4 to 1.8 kg.ha⁻¹.year⁻¹; and the lowest $N-NH_4^+$ depositions from 0.3 to 1.6 kg.ha⁻¹.year⁻¹ (Fischer et al. 2010).

On the plots of this study, similarly to the other European plots, was identified markedly different development of evaluated depositions. This could be attributed mainly to influence of local and transboundary anthropogenic emissions.

Reducing of sulphur depositions has the beneficial effect on the growth of European silver fir (Elling et al. 2009; Bošeľa et al. 2014; Büntgen et al., 2014) as well as Norway spruce (Kroupová 2002; Kolář et al. 2015).

5. Conclusion

Though the major threat for forest ecosystems in Europe is considered climate change, forests in the Slovak and Czech Republic are constantly threatened by a considerable environmental impact from industry, agriculture, transport and transboundary emissions of nitrogen, sulphur and other specific pollutants. So in spite of regulations, the depositions of nitrogen and sulphur are still relatively high. Alarmingly, these depositions in the Slovak and Czech Republic belong persistently among the highest in Europe. The statistically significant decrease (P \leq 0.01) of both forms of nitrogen and sulphur deposition was determined in this study on Slovak plots. The decrease of N-NO₃⁻ and S-SO₄²⁻ depositions in the Czech Republic was less statistically outstanding $(P \le 0.05)$ and the changes of N–NH₄ depositions was not statistically significant. The development of annual depositions of all evaluated parameters has variations on individual plots. At any rate the most significant decrease was found in sulphur deposition on the majority of evaluated plots, which reflects significant sulphur reduction of the industrial emissions. The annual depositions are closely connected with the amount of emissions in both republics. However, in the future we can expect the slight increase in NH_3 emissions connected with increasing the number of bred animals, and of the number of cars equipped with catalytic converters. On the contrary further decrease in NOx emission is expected due to further tightening in emission limits in future, and the modernization of cars.

Qualitative and quantitative assessments of the atmospheric deposition are essential for understanding regional variations and demonstrating the effectiveness of policies to reduce emissions. Deposition monitoring can help clarify trends in atmospheric deposition through the years in order to assess whether emission patterns have an effect on deposition. As such, deposition monitoring is an important input to the creation and implementation of clean air policies at the European level.

Acknowledgement

We are grateful to GISAT© and CORINE for providing data. Data were obtained from the ICP Forest Monitoring Programme (icp-forest.net) which is funded by the Ministry of Agriculture in the Czech Republic. The evaluation was as well supported by the Ministry of Agriculture of the Czech Republic – Resolution RO 0117 (reference number 6779/2017-MZE-14151. This work was supported by the Ministry of Agriculture and Rural Development of the Slovak Republic under the assignment No. 5 "Monitoring of Forests – PMS Forests" of the Contract 406/2017-710/ MPRV SR.

References

- Blihárová, M., Škvarenina, J., 2002: The altitude influence on atmospheric deposition and chemism of he throughfall and stemflow in climax and monoculture spruce stands of biosphere reserve Polana. In: Proceedings of papers from XIV. Bioclimatological conference – Lednice in Moravia, p. 499–507.
- Bontemps, J. D., Hervé, J. C. J., Leban, M. J.-F., Dhôte, J. F., 2011: Nitrogen footprint in a long-term observation of forest growth over the twentieth century. Trees, 25:237–251.
- Bošeľa, M., Petráš, R., Sitková, Z., Priwitzer, T., Pajtík, J., Hlavatá, H. et al., 2014: Possible causes of the recent rapid increase in the radial increment of silver fir in the Western Carpathians. Environmental Pollution, 184:211–221.
- Büntgen, U., Tegel, W., Kaplan, J. O. et al., 2014: Placing unprecedented recent fir growth in a European-wide and Holocene-long context. Frontiers in Ecology and the Environment, 12:100–106.
- Canfield, D. E., Glazer, A. N., Falkowski, P. G., 2010: The evolution and future of Earth's nitrogen cycle. Science, 330:192–196.

- Clarke, N. et al., 2010: Sampling and Analysis of Deposition. Part XIV. In: Manual on methods and criteria for harmonized sampling, assessment, monitoring and analysis of the effects of air pollution on forests. UNECE, ICP Forests, Hamburg, 66 p. [http://www. icp-forests.org/Manual.htm] – Accessed on 16. 4. 2018.
- Dědina, M., 2013: Vývoj prachových částic ze zemědělství do roku 2020 a roku 2040. Výzkumná zpráva pro TAČR TA01020500 Podrobný emisně-imisní model ČR pro současný stav a výhled do roku 2030 a nástroje pro podporu rozhodování v oblasti ochrany ovzduší. VÚZT, Praha
- De Vries, W., Dobbertin, M. H., Solberg, S., van Dobben, H. F., M. Schaub, M., 2014: Impacts of acid deposition, ozone exposure and weather conditions on forest ecosystems in Europe: an overview. Plant and Soil, 380:1–45.
- Elling, W., Dittmar, C., Pfaffelmoser, K., Rötzer, T., 2009: Dendroecological assessment of the complex causes of decline and recovery of the growth of silver fir (*Abies alba* Mill.) in Southern Germany. Forest Ecology and Management, 257:1175–1187.
- Elser, J. J., 2011: A world awash with nitrogen. Science, 334:1504–1505.
- Fischer, R., Lorenz, M., Granke, O., Mues, V., Iost, S., Van Dobben, H. et al., 2010: Forest Condition in Europe, 2010 Technical Report of ICP Forests. Work Report of the Institute for World Forestry 2010/1. ICP Forests, Hamburg, 2010, 175 p.
- Hunová, I., Maznová, J., Kurfürst, P., 2014: Trends in atmospheric deposition fluxes of sulphur and nitrogen in Czech forests. Environmental Pollution, 184:668–675.
- Kolář, T., Čermák, P., Oulehle, F., Trnka, M., Štepánek, P., Cudlín, P. et al., 2015: Pollution control enhanced spruce growth in the "Black Triangle" near the Czech-Polish border. Science of the Total Environment, 538:703–711.
- Kroupova, M., 2002: Dendroecological study of spruce growth in regions under long-term air pollution load. Journal Forest Science, 48:536–548.
- Lawrence, G., Sutherland, J., Boylen, C., Nierzwicki-Bauer, S. A., Momen, B., Baldigo, B. P., 2007: Acid rain effects on aluminum mobilization clarified by inclusion of strong organic acids. Environmental Science & Technology, 41:93–98.
- Lorenz, M., Fischer, R., Becher, G., Granke, O., Roskams, P., Nagel, H.-D. et al., 2007: Forest Condition in Europe 2007. Technical Report of ICP Forests. Work report of Thunen Institute for Word Forestry. Hamburg, ICP Forests, 98 p.
- Lorenz, M., Nagel, H.-D., Granke, O., Kraft, P., 2008: Critical loads and their exceedances at intensive forest monitoring sites in Europe. Environmental Pollution, 155:426–435.

- Lorenz, M., Fischer, R., Becher, G., Mues, V., Granke, O., Braslavskaya, T. et al., 2009: Forest Condition in Europe 2009. Technical Report of ICP Forests. Work report of Thunen Institute for Word Forestry. Hamburg, ICP Forests, 86 p.
- Michel, A., Seidling, W., Lorenz, M., Becher, G., 2013: Forest Condition in Europe, 2013. Technical Report of ICP Forests. Report under the UNECE convention CLRTP, Thunen working paper 19, Hamburg, 134 p.
- Pardo, L. H., Fenn, M. E., Goodale, C. L., Geiser, L. H., Driscoll, C. T., Allen, E. B. et al., 2011: Effects of nitrogen deposition and empirical nitrogen critical loads for ecoregions of the United States. Ecological Applications, 21:3049–3082.
- Sitková, Z., 2006: Influence of selected ecological factors on forest ecosystems in relation to the dying off mountain spruce forests in parts of TANAP. Dissertation, Zvolen, TU in Zvolen, 146 p.
- Stern, D. I., 2005: Global sulphur emissions from 1850 to 2000. Chemosphere, 58:163–175.
- Thalmann, E., Burkard, R., Wrzesinsky, T., Eugster, W., Klemm, O., 2002: Ion fluxes from fog and rain to an agricultural and a forest ecosystem in Europe. Atmospheric Research, 64:147–158.
- Thöni, L., Seitler, S., Meier, M., Zürcher, F., Hertz, J., 2008: The development of loads of cations, anions, Cd and Pb in precipitation and of atmospheric concentrations of N-components, in Switzerland from 1988 to 2003. Environmental Monitoring Assessment, 138:207–218.
- Ulrich, B., Mayer, R., Khanna, P. K., 1980: Chemical changes due to acid precipitation in a loess-derived soil in central Europe. Soil Science, 130:193–199.
- Vestreng, V., Myhre, G., Fagerli, H., Reis, S., Tarrasón, L., 2007: Twenty-five years of continuous sulphur dioxide emission reduction in Europe. Atmospheric Chemistry and Physics, 7:3663–3681.
- Walaszek, K., Kryza, M., Dore, A. J., 2013: The impact of precipitation on wet deposition of sulphur and nitrogen compounds. Ecological Chemistry and Engineering S, 20:733–745.
- Waldner, P., Marchetto, A., Thimonier, A., Schmitt, M., Rogora, M., Granke, O. et al., 2014: Detection of temporal trends in atmospheric deposition of inorganic nitrogen and sulphate to forest in Europe. Atmospheric Environment, 95:363–374.
- Walna, B., Kurzyca, I., 2007: Evaluation of Bulk Deposition in Protected Woodland Area in Western Poland. Environmental Monitoring Assessment, 131:13–26.
- Zhao, Y., Duan, L., Xing, J., Larssen, T., Nielsen, C. P., Hao, J., 2009: Soil acidification in China: is controlling SO₂ emissions enough Environmental Science & Technology, 43:8021–8026.