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EVALUATION OF SULPHUR AND NITRATE-NITROGEN DEPOSITION TO FOREST ECOSYSTEMS

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Abstract

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The paper evaluates the load of sulphur and nitrate-nitrogen deposition to beech ecosystems in the Kremnické vrchy Mts, over the period 1987–2001. The values of deposition in precipitation water sampled on two plots: on an open area (OA) and in a stand (S) at the site have been compared with several other localities. In the evaluated 15-year study period ranged the sulphate concentration from 6.38 to 17.26 mg l^{-1} on OA and from 8.47 to 22.05 mg l^{-1} in the stand. The corresponding values obtained for nitrate are 1.54–12.36 mg l^{-1} (OA) and 2.34–12.73 mg l^{-1} (S). The mean values of nitrate concentration on both plots: 3.99 mg l^{-1} (OA) and 4.54 mg l^{-1} (S) were lower compared to the sulphate: 11.29 mg l^{-1} (OA) and 13.58 mg l^{-1} (S). The mean deposition of sulphur to the open area was more than four times higher than that of nitrate-nitrogen, in the stand it was more than five times. The sulphur deposition to OA and stand was practically the same: 25 and 24.9 kg S ha^{-1} $year^{-1}$, respectively, the nitrate-nitrogen deposition to OA (5.7 kg) was higher compared to the stand (4.7 kg N-NO_3^- ha^{-1} $year^{-1}$). The total amounts of substances passed through the tree crowns and accumulated in the forest soil over the 15-year study period were: 372 kg of sulphur and 57 kg of nitrate-nitrogen, together 181.5 kg of nitrate- and ammonium-nitrogen (summary values) per one hectare. Comparing the obtained values of sulphur and nitrate-nitrogen deposition to the corresponding values from other localities in Slovakia and abroad we can see that the examined beech ecosystem in the Kremnické vrchy Mts has been exposed to a much lower loading with the two elements.

Key words: sulphur and nitrate-nitrogen deposition, forest ecosystems

Introduction

Forest ecosystems are loaded with considerable amounts of atmospheric deposition, both wet and dry, consisting of a large number of various soluble and non-soluble materials and particles. The most important are sulphur and nitrogen compounds and hydrogen ions having acidic character (Binkley, Richter, 1987). These materials have a considerable influence on forest ecosystems – on their ecology related to atmospheric pollutants, their health status and stability (Johnson, Lindberg, 1992). Situated at the same centre of the European continent, the territory of Slovakia is loaded considerably with atmospheric pollution from long-range transport (Poland, Czech Republic, Germany and especially Hungary). Domestic industrial sources also take their share in the load to the ecosystems. The chemistry of precipitation is well reflecting the input of both acid and base components. The first comprise primarily sulphate, nitrate, fluoride and hydrogen ions. From the base cation is important calcium entering forest ecosystems in dust formed from the parent rock material of the forest soils themselves. Precipitation and throughfall sampled in forest ecosystems are powerful tool for obtaining data on amounts, time dynamics and fluxes of nutrients and pollutants in the examined ecosystems. The aim of this work is to document the trends in sulphur and nitrate-nitrogen deposition in the examined beech ecosystem over the study period of 15 years and to point out and interpret extremes in the obtained time series.

Material and methods

The research on sulphur and nitrate-nitrogen deposition to beech ecosystems was carried out in a beech stand growing at the Beech Ecological Experimental Site (BEES) in the south-eastern part of the Kremnické vrchy Mts, upper catchment of the Kováčovský potok water stream. The site is situated at 460–510 m a.s.l., on a western-oriented slope with an inclination of 15°. The stand is aged 100 years and consists of beech trees with a small admixture of other species only. All the species in the stand, primarily the trees, are dynamically responding to the permanent changes in processes running at the site after a cutting intervention in 1989. The state of art was explored with profit by Barna (2000a, b) who obtained more precise data about the trends in density development of the beech stand on all the research plots at the experimental site.

The annual precipitation totals on the BEES plots recorded from 1987 up to the evaluation year (2001) ranged from 508 to 1036 mm, the total amounts in vegetation periods were from 160 to 530 mm (Dubová, 2001). The mean annual temperature is 6.8°C (Střelec, 1988). More details about the site, including description of the geological substrate, soil and climate conditions are in Bublinec, Dubová (1989).

Before the evaluation of the bulk deposition itself, we perform physical and chemical analysis of precipitation and throughfall. The material is sampled at the BEES, on five plots with differentiated stocking intensity. The samples are analysed in the laboratories of the Institute of Forest Ecology SAS in Zvolen. The list of the used physical and physical-chemical methods is in Table 1. The analyses give results about electrical conductivity

T a b l e 1. Physical and chemical methods used in analyses of precipitation and throughfall-sampled in forest ecosystems.

Characteristics	Method
electrical conductivity	conductometry
pH	potentiometry
NO_3^-	ISE – potentiometry
NH_4^+, F^-	spectrophotometry
SO_4^{2-}	titrimetry
Ca^{2+}, Mg^{2+}	atomic absorption spectrophotometry
K^+, Na^+	atomic emission spectrophotometry

(µS cm^{-1}), pH value, and concentration of inorganic ions (mg l^{-1}). The measured values provide a background for deriving time dynamics of the variables, their trends over the period of study, amounts of deposition of the individual components and the fluxes in the examined forest ecosystems in kg ha^{-1} year^{-1} (Bublinec, Dubová, 1989, 1993a; Bublinec et al., 2002, 2003; Kellerová, 1999; Kellerová, Dubová, 2002). Our former research of forest ecosystems has been focused on acid deposition of sulphur, nitrogen, fluorine and hydrogen ions (Bublinec et al., 1995; Bublinec, Dubová, 1997, 1998; Dubová, 1995, 1998; Dubová et al., 1995) and alkaline deposition of calcium, magnesium and potassium (Bublinec, Dubová, 1993b; Dubová, Bublinec, 2002a, 2004). In this contribution we evaluate the trends in sulphur and nitrogen deposition in the examined beech ecosystem over the study period of 15 years and point out and interpret extremes in the obtained time series.

Results and discussion

In this paper we present determined sulphate and nitrate concentration in precipitation and throughfall and the derived values of sulphur and nitrate-nitrogen deposition on two plots at the BEES – the open area (OA) and the stand (S) at the experimental site – control plot with stocking density of 0.9, omitted from the management. Sulphur and nitrate-nitrogen deposition (kg ha^{-1} year^{-1}) entering the open area and entering the forest soil after having passed through the tree crowns were calculated from the determined concentrations of sulphate and nitrate (mg l^{-1}) and from precipitation amounts (mm) collected on the open area and in the stand. Table 2 gives a survey of extreme and mean precipitation totals, concentrations of sulphate and nitrate, deposition of sulphur and nitrate-nitrogen to the plots (OA and S) over 1987–2001. In addition, trends and time dynamics of sulphur and nitrate-nitrogen deposition have been evaluated for the two plots.

T a b l e 2. Extreme and average values of precipitation totals, sulphate and nitrate concentration, sulphur and nitrate-nitrogen deposition to open area (OA) and stand (S) at the BEES Kremnické vrchy Mts in 1987–2001.

Characteristic		Precipitation		SO_4^{2-}		NO_3^-		Sulphur		Nitrate-Nitrogen	
		mm	%	mg l^{-1}	%	mg l^{-1}	%	kg ha^{-1} year^{-1}	%	kg ha^{-1} year^{-1}	%
Minimum	OA	508	78	6.38	57	1.54	39	13.5	54	2.7	47
	S	353	76	8.47	54	2.34	52	10.8	43	2.3	49
Average	OA	654	100	11.29	100	3.99	100	25.0	100	5.7	100
	S	466	100	15.68	100	4.54	100	24.9	100	4.7	100
Maximum	OA	1036	158	17.26	153	12.36	310	43.4	174	16.2	284
	S	720	155	22.05	141	12.73	280	43.6	175	13.3	283

The one-year precipitation totals over the evaluated 15-year period (1987–2001) on the open area reach a mean value of 654 mm with a range of 528 mm. The corresponding values in the stand are 466 mm and 367 mm (Dubová, 2001). The range is considerable (more than 100%), the maximum values are more than two times higher than the minimum values. Variation coefficients on the two plots are very similar (22–23%).

The sulphate concentration on the open area range from 6.38 to 17.26 mg l^{-1} and in the stand from 8.47 to 22.05 mg l^{-1}. The maximum values are 2.6 to 2.7-times higher than the minimum values. The range is 10.88 mg l^{-1} (OA), and 13.58 mg l^{-1} (S). The mean value over the 15-year period in the open area is 11.29 mg l^{-1} (variation coefficient 32%) and in the stand 15.68 mg l^{-1} (22%). The maximum sulphate concentration on the open area is by 50% higher compared to the mean. In the stand is the maximum sulphate concentration higher by about 40% compared to the mean.

The nitrate concentration on the open area range from 1.54 to 12.36 mg l^{-1} – we can see that the maximum value is 8-times higher than the minimum, the range is 10.82 mg l^{-1}. The corresponding values in the stand are from 2.34 to 12.73 mg l^{-1} (10.39 mg l^{-1}). The mean values over the 15-year study period are 3.99 mg l^{-1} in the OA and 4.54 mg l^{-1} in the stand. These values result in higher variation coefficient compared to the sulphate (58–68%). The maximum concentration values are higher compared to the mean by about 210% on the open area and by about 180% in the stand.

The sulphur deposition (kg S ha^{-1} $year^{-1}$) – mean annual value is 25 kg to the open area and 24.9 kg to the stand, the corresponding maximum values are 43.4 kg and 43.6 kg. The values for the two plots are nearly the same. The range on the OA is 29.9 kg, in the stand 32.8 kg (Table 2). The maximum values are by about 75% higher than the mean values. Evaluating the 15-year time series of sulphur deposition we can see conspicuous extremes and disagreements with the remarkable overall decrease in emissions of sulphur dioxide in the Slovak Republic (Fig. 1) in 1987–2001 (Anonymous, 2001, 2003).

The nitrate-nitrogen deposition (kg N-NO_3^- ha^{-1} $year^{-1}$) – the mean annual value to the open area (5.7 kg) is higher than to the stand (4.7 kg). The range on the open area is 13.5 kg (2.7 to 16.2 kg) and in the stand 11 kg, from 2.3 to 13.3 kg (Table 2). We can see that the maximum values are about 6-times higher than the minimum deposition (5.8 to 6-times).

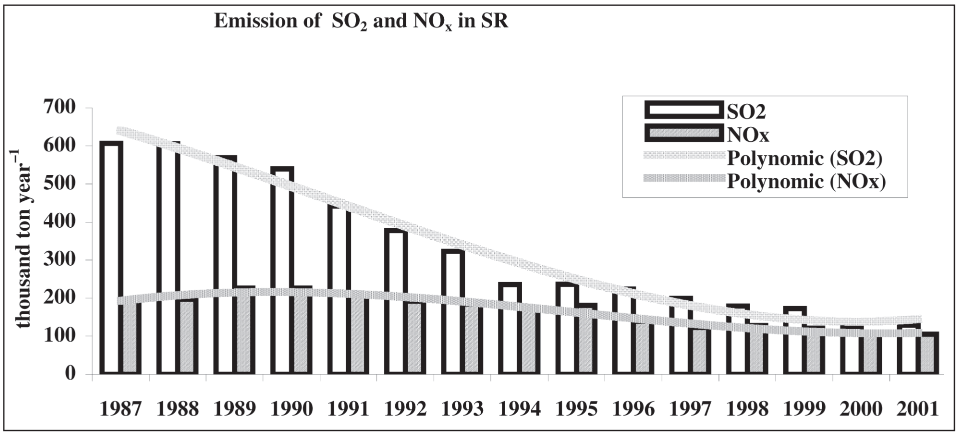

Fig. 1. Development trends in emission of basic pollutants (SO_2 and NO_x) in the Slovak Republic.

Examining the sulphur and nitrate-nitrogen deposition over the 15-year period of study we obtain much higher mean values of sulphur deposition compared to the nitrate-nitrogen deposition – by more than 4-times in the case of the open area and more than 5-times in the case of the forest stand. The throughfall has transported to the forest ground considerable amounts of both nutrients and pollutants. Summary values of the deposition in the stand over the 15-year period to one-hectare are 372 kg of sulphur and 57 kg of nitrate-nitrogen form. Together with the ammonium-nitrogen form, the nitrogen deposition has increased to 181.5 kg N (Bublinec, Dubová, 2003).

The crown impact coefficient expressing the enrichment or impoverishment of the precipitation water passing through the crowns is calculated as the ratio between the mean values of the deposition to the stand and the deposition to the open area. For the period of study we have obtained the following values: 1.0 for sulphur, 0.8 for nitrate nitrogen and 0.9 for ammonium nitrogen (Bublinec, Dubová, 2003).

The highest annual sulphur deposition was found in 1994 and 1995 (Fig. 2). In the year 1994 were high deposition values found on both plots, in 1995 primarily on the open area. This fact is also documented with the results reported in the Report for the Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe. The high values of sulphur deposition found in 1995 are higher than the mean value for the study period (1985–1996). The high sulphur deposition (> 20 kg S ha^{-1} $year^{-1}$) had been transported from emission sources in the south-eastern part of the Great Britain and in Central Europe (Germany, Poland, the Czech Republic and the Slovak Republic). The sulphur deposition in this year was probably increased also due to SO_2 contribution from the Sicily Island – heavy volcanic activities of the Etna Mt. in 1995. In the following year 1996, there was a decrease in sulphur deposition over the whole Europe. In 1995, the

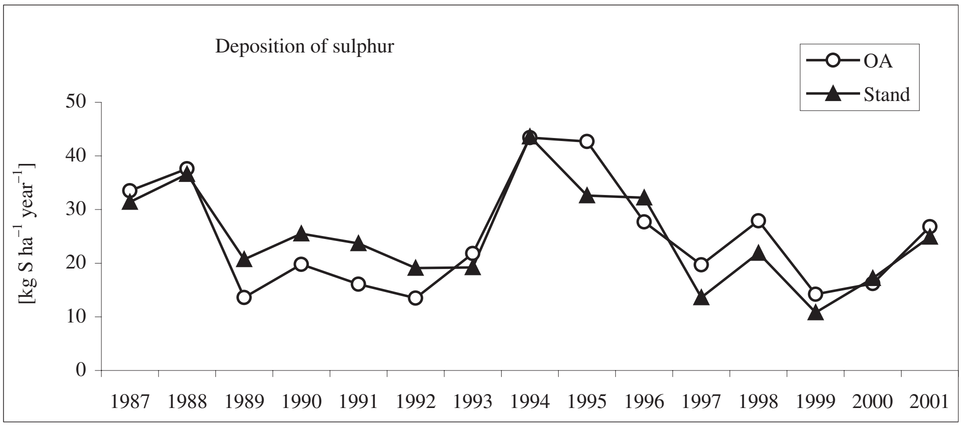

Fig. 2. Deposition of sulphur (kg S ha^{-1} $year^{-1}$) to open area (OA) and beech stand (Stand) at the BEES Kremnické vrchy Mts.

sulphur deposition in Slovakia was kept at levels of 20 to 40 kg S ha^{-1} year^{-1}, except of the western and eastern borders with values < 20 kg S ha^{-1} year^{-1} (Tsyro, 1998). Our data from the BEES in the Kremnické vrchy Mts were also compared with these values. Also Peters et al. (1999), monitoring in 1977–1997 the deposition in bulk precipitation and stream water chemistry in the Krušné hory Mts (Ore Mountains), recorded in 1995 increased amounts of sulphur both in precipitation and in surface water.

The situation was different for nitrate-nitrogen deposition (Fig. 3), also in this case, however, some extremes can be identified (e.g. the last documented year 2001). The highest values of the annual nitrate-nitrogen deposition to the two plots were found in the first and the second year of the precipitation sampling (1987–1988). According to the data reported by Tsyro (1998) for year 1996, the nitrogen deposition (nitrogen oxidised) at boundaries of Europe was 2 to 4 kg N ha^{-1} year^{-1}. In Central Europe, Great Britain and in southern Scandinavia were reached amounts higher than 4 kg, sometimes up to 10 kg N ha^{-1} year^{-1}. In this range is also situated the Slovak Republic. The values obtained at the BEES in the Kremnické vrchy Mts are consistent with these data.

On each plot, a direct linear dependence exists between the precipitation amount and the sulphur deposition, which means that the higher is the precipitation, the higher is also the sulphur deposition. The values of correlation coefficient (r) are 0.560 and 0.756 on the open area and in the stand, respectively. On the other hand, it is interesting that no direct dependence exists between the precipitation amount and the nitrate-nitrogen deposition either to the open area or to the forest stand. In this case correlation coefficients are close to zero.

On each plot was found a direct linear dependence between the sulphate concentration and the sulphur deposition. The correlation coefficients are 0.754 and 0.737 in the open area and in the stand, respectively. The coefficients of correlation between the nitrate concentra-

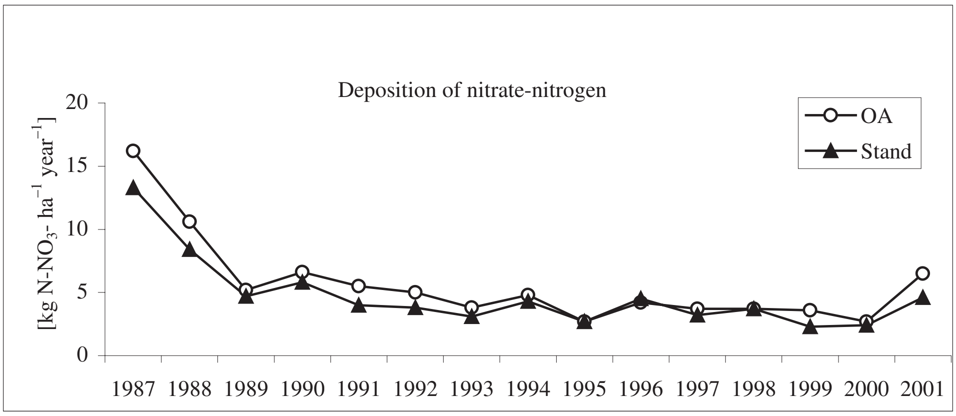

Fig. 3. Deposition of nitrate-nitrogen (kg $N\text{-}NO_3^-$ ha^{-1} rok^{-1}) to open area (OA) and beech stand (Stand) at the BEES Kremnické vrchy Mts.

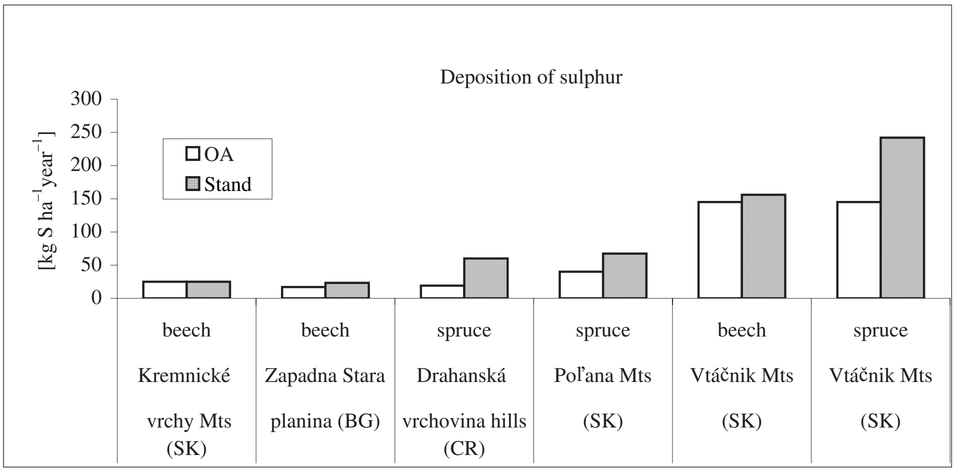

Fig. 4. Comparison between the values of sulphur deposition to several localities in Bulgaria (BG), the Czech Republic (CZ) and the Slovak Republic (SK) (OA – open area, Stand – throughfall in beech and spruce stands).

tion and the nitrate-nitrogen deposition are high and very similar: 0.887 for the open area and 0.920 for the stand. So, the nitrate-nitrogen deposition is significantly dependent on the nitrate concentration; on the other hand, it is fully independent of the precipitation amount. This is also confirmed by the fact that the highest nitrate concentration is connected with the highest nitrate-nitrogen deposition which does not hold for sulphur.

The coefficients of skewness and kurtosis, testing the significance of differences between the actual and normal distribution have revealed that there is no significant difference between the examined distributions of the values of sulphate concentration and sulphur deposition and the normal distribution. In the case of nitrate concentration and nitrate-nitrogen deposition, the differences between the actual and normal distribution were significant.

It has also been found that the variability of sulphate concentration is lower than the variability of the values of sulphur deposition (36% in the stand, 42% on open area). On the other hand, the variation coefficient in nitrate concentration is nearly the same (58–68%) as in the case of nitrate-nitrogen deposition (60–61%).

The values of the sulphur and nitrate-nitrogen deposition to the two plots at the BEES were compared with the data from other sites in Slovakia and abroad (Fig. 4 and 5). Ignatova and Fikova (1998) published the data about deposition of sulphur and nitrate-nitrogen to beech ecosystems in the Zapadna Stara planina Mts in Bulgaria. The authors found an annual sulphur deposition per one hectare in open area ranging from 9.5 to 25.6 kg (with an average of 17.1 kg) and in beech stand from 15.2 to 30.8 kg (an average of 23.4 kg). The corresponding values for nitrate-nitrogen deposition were 1.9 to 6 kg (an average of 3.4 kg) and 3.0 to 5.6 kg (an average of 4.0 kg). The mean values are similar to the values

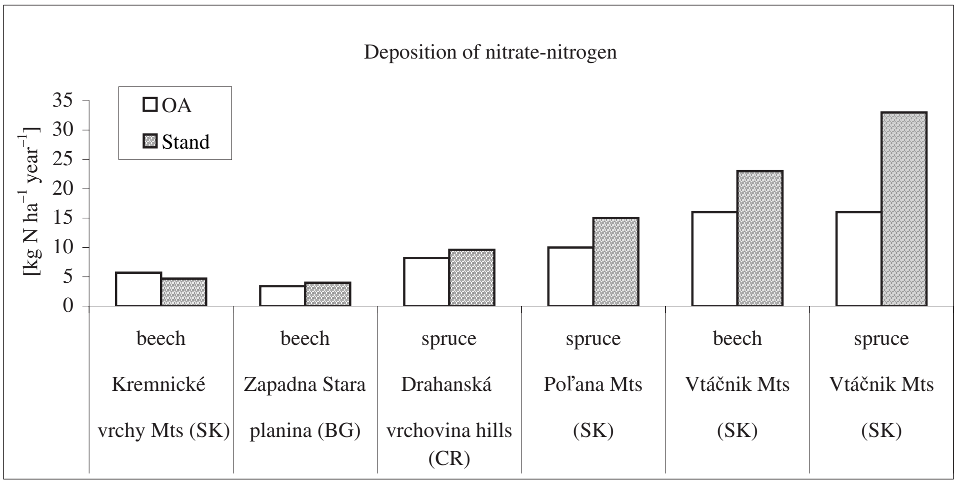

Fig. 5. Comparison between the values of nitrate-nitrogen deposition to several localities in Bulgaria (BG), the Czech Republic (CZ) and the Slovak Republic (SK) (for the abbreviations see Fig. 4).

obtained at the BEES in the Kremnické vrchy Mts. On monitoring plots situated in a spruce ecosystem in the Poľana Mts (1430 m a.s.l.), the mean annual deposition of sulphur per one hectare (10-year average) is 40.1 kg to open area and 67.3 kg in stand. The range limits of sulphur deposition to open area are 16.2 and 87.8 kg (minimum 5.4-times lower than maximum) and to stand 23 and 197.8 kg, what means the minimum 8.6-times lower than the maximum. The corresponding limit values for nitrate-nitrogen are 2.8 and 14.1 kg (10-year average of 8.2 kg) for open area 4.7 and 13.8 kg (10-year average of 9.6 kg) for forest stand (Dubová, Bublinec, 2002b). The mean deposition values are more than two times higher than the values obtained at the BEES. Kulhavý (2001), providing with the data by Klimo (1992) reports for a spruce ecosystem in the Drahanská vrchovina hills (625 m a.s.l.) in the Czech Republic the following mean annual values for deposition per one hectare: 19 kg of sulphur and 10 kg of nitrate-nitrogen to open area, 60 kg of sulphur and 15 kg of nitrate-nitrogen to spruce stand – so these values are two and more than three times higher than the corresponding values found at the BEES. They are more similar to the values found for the spruce ecosystem in the Poľana Mts. Ďurečková (2003) evaluated the sulphur and nitrate-nitrogen deposition to the top area of the Vtáčnik Mts (1340 m), heavy loaded with atmospheric pollution. The author reports the following mean annual values found in this locality for sulphur (nitrate-nitrogen) deposition over a 7-year study period (1990–1996): 145 kg S (16 kg N-NO_3^-) to open area, 156 kg S (23 kg N-NO_3^-) to beech stand and 242 kg S (33 kg N-NO_3^-) to spruce stand per one hectare. The figures are 3 to 6-times higher compared to the BEES.

Conclusion

Concentration of sulphates determined in the precipitation and throughfall sampled at the Beech Ecological Experimental Site in the Kremnické vrchy Mts range from 6.38 to 17.26 mg l^{-1} (OA) and from 8.47 to 22.05 mg l^{-1} (S) with the mean values of 11.29 mg l^{-1} (OA), and 13.58 mg l^{-1} (S), evaluated over the 15-year study period. The mean concentration of nitrates has lower values: 3.99 mg l^{-1} for OA and 4.54 mg l^{-1} for the stand. The determined values of nitrate concentration in OA range from 1.54 to 12.36 mg l^{-1} and in the stand from 2.34 to 12.73 mg l^{-1}. The mean values of sulphur deposition to OA and to the stand are very similar: 25 kg, and 24.9 kg S ha^{-1} $year^{-1}$, respectively. The mean annual deposition of nitrate-nitrogen to OA (5.7 kg) is higher than to the stand (4.7 kg $N\text{-}NO_3^-$ ha^{-1} $year^{-1}$). The sulphur deposition at the BEES is higher than the nitrate-nitrogen deposition: to open area by four times, to the stand by more than five times. During the 15-year period of study, the throughfall imported to one-hectare area of the soil surface represented 372 kg of sulphur and 57 kg of nitrate-nitrogen. Including also the ammonium nitrogen, we obtain 181.5 kg of nitrogen (summary value for both nitrogen forms). Comparing our values of sulphur and nitrate-nitrogen deposition with the values from other localities in Slovakia and abroad we can conclude that the beech ecosystems in the Kremnické vrchy Mts are exposed to lower load by the two evaluated elements.

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V príspevku je hodnotenie záťaže bukového ekosystému depozíciou síry a nitrátového dusíka v Kremnických vrchoch v období rokov 1987 až 2001. Hodnoty záťaže získané zo zrážkových vôd odobratých na voľnom priestranstve (VP) a v poraste (P) stacionára sa porovnávajú s inými lokalitami. V hodnotenom 15-ročnom období sa koncentrácia síranov pohybuje od 6,38 do 17,26 mg l^{-1} na VP a od 8,47 do 22,05 mg l^{-1} v poraste, dusičnanov od 1,54 do 12,36 mg l^{-1} (VP), resp. od 2,34 do 12,73 mg l^{-1} (P). Priemerná koncentrácia dusičnanov 3,99 mg l^{-1} (VP) a 4,54 mg l^{-1} (P) je nižšia ako síranov (11,29 mg l^{-1} na VP, resp. 13,58 mg l^{-1} v poraste). Priemerná depozícia síry prevyšuje depozíciu nitrátového dusíka na VP viac ako 4-krát a v poraste viac ako 5-krát. Depozícia síry na VP a v poraste je takmer rovnaká, a to 25 kg, resp. 24,9 kg S ha^{-1} rok^{-1}, depozícia nitrátového dusíka je na VP vyššia ako v poraste (5,7 kg, resp. 4,7 kg N-NO_3^- ha^{-1} rok^{-1}). Na obidvoch plochách je medzi množstvom zrážok a depozíciou síry priama lineárna závislosť, teda čím sú väčšie zrážky, tým je väčšia depozícia síry. Hodnoty koeficientov korelácie (r) sú 0,560 na VP a 0,756 v poraste. Zaujímavé však je, že v prípade množstva zrážok a depozície nitrátového dusíka na VP i v poraste nie je priama závislosť. Korelačné koeficienty majú hodnoty blízke nule. Za 15-ročné obdobie sa podkorunovými zrážkami na povrch lesnej pôdy (ha^{-1}) bukového ekosystému dostalo 372 kg síry a 57 kg nitrátového dusíka, resp. 181,5 kg N spolu s amoniakálnou formou (kumulatívne hodnoty). Porovnaním depozície síry a nitrátového dusíka s inými lokalitami na Slovensku, či v zahraničí možno konštatovať, že bukový ekosystém nachádzajúci sa v Kremnických vrchoch predstavuje lokalitu s nižšou záťažou síry a nitrátového dusíka.