

## The concentration of $\text{SO}_4^{2-}$ and amount of $\text{S-SO}_4^{2-}$ in soil water and throughfall in beech forest of Štiavnické vrchy Mts, Slovakia

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### Abstract

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The paper deals with the results of concentration  $\text{SO}_4^{2-}$  and amount of  $\text{S-SO}_4^{2-}$  in soil water from the beech forest situated in the Štiavnické vrchy Mts. The mean concentration of  $\text{SO}_4^{2-}$  increased from 19.06 mg l<sup>-1</sup> in the depth of surface humus ( $F_{00}$ ) to 29.32 mg l<sup>-1</sup> in the depth 0.25 m ( $F_{25}$ ). To the soil during the study period input 415 kg  $\text{S-SO}_4^{2-}$  ha<sup>-1</sup> –  $F_{00}$  and 587 kg  $\text{S-SO}_4^{2-}$  ha<sup>-1</sup> in the layer  $F_{25}$ . The highest  $\text{SO}_4^{2-}$  concentration were observed during the year 1988 in the all depths, but the lowest values were found in the year 2010 in the  $F_{00}$  layer with 8.02 mg l<sup>-1</sup>. The  $\text{SO}_4^{2-}$  concentration in soil water with the depth increased. The regression analysis found out a statistically significant influence of the sulphur content in the atmospheric deposition on the sulphur content in soil water. A significant correlation between the precipitation amount and the content of sulphur in precipitation and in soil water was observed. Student's t-test for dependent variables confirmed statistically strong significant differences of the sulphur content in soil water in the 0.1 m and 0.25 m depth between the study areas. No significance of differences between the years at the same plot was found.

### Key words

$\text{SO}_4^{2-}$  concentration, sulphate, soil water, Štiavnické vrchy Mts, throughfall

### Introduction

Sulphur dioxide inputs to soil via atmospheric precipitation (QUILCHANO et al., 2002; STACHURSKI and ZIMKA, 2000). In soil it causes displacement of base ions and consequent acidification. Acidification is a long-term cumulative and dynamic process (HRUŠKA et al., 2001; COSBY et al., 1986). Its effect is expressed after long period and has mainly a negative sense.

The alarmed situation of environmental and forest soil degradation led to the introducing of the deposition limits for forest ecosystems (MINĐAŠ et al., 2001). The determination of deposition limits has become an important tool in the development of the national strategies for decreasing emissions of sulphur and nitrogen in Europe and North America (MATZNER and MEIWES,

1994). These efforts, industrial slowdown (predominant the mining in Poland, Northern Bohemia and the formed GDR) and conversion of economy in the whole of Eastern Europe had a considerable impact on decreasing of the emission amount of sulphur and nitrogen (DUBOVÁ and BUBLINEC, 2006; LINDROOS et al., 2006; ZAPLETAL, 2006; PRECHTEL et al., 2001). The region of Slovakia is moderately environmentally sensitive according to the assessment of the selected sustainable development indicators. The critical values of sulphur deposition for Slovakia is 10–30 kg ha<sup>-1</sup> yr<sup>-1</sup> but it has been exceeded approximately at 25% of forest soil.

In Slovakia during 1989–1999 period was observed the diminution of the emitted primary pollutants by 57.9%, it presented average annual decreasing by about 6%. In 1998 to Slovakia were imported approxi-

mately 75,000 tons of sulphur (151,400 tons of SO<sub>2</sub>) and exported 74,600 tons of sulphur (149,200 tons of SO<sub>2</sub>), it means less by 2,200 tons.

In 2000 the emissions of particle matter released to the atmosphere were by 83% (58,408 t yr<sup>-1</sup>) lower than in the 1985–1987 period. The decreasing of emission amount was observed also in the SO<sub>2</sub> emissions (by 77% to the value 134,376 t yr<sup>-1</sup>) and the NO<sub>x</sub> emissions (by 42% to the value 113,877 t yr<sup>-1</sup>) (KALÚZ, 2004). According to The Slovak Forest Health Monitoring Report 2003 (www.forestportal.sk) was the atmospheric deposition of sulphur at all monitored sites lower than the atmospheric deposition of nitrogen. In 2006 was observed decrease of the sulphur atmospheric deposition by 50% (3.8–16.4 kg ha<sup>-1</sup>yr<sup>-1</sup>) compared to 2001 levels.

In 2000 in the Czech Republic the SO<sub>2</sub> emissions decreased by 86% and the total ammonia emissions by 53% compared to 1990 levels. The dry deposition of SO<sub>x</sub> (SO<sub>2</sub> + SO<sub>4</sub><sup>2-</sup> in aerosol) declined by 81% and the wet deposition of SO<sub>4</sub><sup>2-</sup> by 32% (ZAPLETAL and CHROUST, 2005). FIALA et al. (2001) showed the data about the decreasing of SO<sub>4</sub><sup>2-</sup> concentration from 7.5 mg l<sup>-1</sup> in 1987 to average 2.5 mg l<sup>-1</sup> in 1997.

Sulphur dioxide emissions in the west Europe (Netherlands, Germany, and Sweden) have decreased since the beginning of the 1980 (PRECHTEL et al., 2001).

In spite of these optimistic results the danger of acidification is still actual problem. It follows from two facts: acidification is a long-term process and the cumulative ability of sulphur in the bottom soil layers. The aim of this paper was to show the trends of SO<sub>4</sub><sup>2-</sup> concentration and in deposition of sulphate sulphur in the area of the Štiavnické vrchy Mts which was strong affected by pollutants.

### Sites description

The research plots (RP) is situated in the south part of the Štiavnické vrchy Mts (48°35' N, 18°51' E), middle Slovakia. The stands plots are formed by beech stands (*Fagus sylvatica* L.) of the age 110 years with the stocking 0.9. The elevation 470 m a.s.l. and climatologically belongs to the warm district. The mean annual precipitation is in the range 700–750 mm. The average air temperature ranges from 8.0 to 8.5 °C and in the growing period and during the year is 14.5–5.5 °C (KELLEROVÁ, 2005).

The Štiavnické vrchy Mts was strong affected by pollutants from the regional sources (production of aluminium, power industry, transport, waste dump). The main contaminants were oxides of fluorine, sulphur and nitrogen, heavy metals as arsenic and cadmium, ozone and particulate matter. The long-time increased input of these pollutants from air to the ecosystems of the Štiavnické vrchy Mts caused the changes in the ecological conditions. This situation led to the negative quality of the beech stands in the study region. The worst effect

for the beech forests had sulphur dioxide (SO<sub>2</sub>) and fluorine (MIHÁLIK and BUBLINEC, 1995). Many years the region of the Štiavnické vrchy Mts around the aluminium plant was regarded as an area with the most polluted environment in Slovakia. For this reason in the 1990's this area was classified to the II.–III. degree of exhalation. Since the end of the 1990's the amount of the pollutants has been decreasing rapidly because of the modernization production processes and more strictly legislation. The concentration of fluorine was cutting down on the tolerable level 1 µg m<sup>-3</sup> (URMINSKÁ et al., 2000).

### Material and methods

Soil solution was sampled by plate plastic lysimeters (1,000 cm<sup>2</sup> each). The first set of the lysimeters were installed in the organic layer in the depth 0.0 m. The second and third set were located at 0.10 m (upper mineral layer) and 0.25 m depths (lower mineral layer) at both study areas (KUKLA, 2002). The samples had been collected monthly since 1988. After sampling the samples were chemical treated and evaluated (JANIČ et al., 2011).

The samplers for throughfall consisted of a bottle equipped with a funnel (660 cm<sup>2</sup> each) inserted into the cap of the bottle. Ten sampling devices were installed on each site (both open field and stand). The samples were collected monthly, eventually after a strong precipitation events. Samples of the open field and stand were individually pooled after each sampling period. These representative samples were analyzed.

Sulphate ions were determined by direct titration with lead nitrate with dithizone as indicator. The results were converted to the sulphate sulphur content.

The data were processed using the statistical program Statistica 7. Student's t-test for dependent variables was used to assess the statistical significance of differences between the study areas. Two-sample test by using the program Statgraphics was used for confirming these results. The impact of precipitation amount to the sulphate sulphur amount in precipitation and in soil water was estimated by simple regression analysis.

### Results

The average SO<sub>4</sub><sup>2-</sup> concentration in soil water with the depth increased from 17.13 mg l<sup>-1</sup> (surface humus), 23.72 mg l<sup>-1</sup> in depth 0.10 m to 29.32 mg l<sup>-1</sup> in the 0.25 m depth (Fig. 1a). This same phenomenon was observed at the plots in the Kremnické vrchy Mts. The mean SO<sub>4</sub><sup>2-</sup> concentration at the open field was lower 14.92 mg l<sup>-1</sup> than in beech throughfall (17.13 mg l<sup>-1</sup>). It can be explained by the washing-off effect. The forest canopy enriched the passing precipitation by captured sulphur compounds. The maximum values of

SO<sub>4</sub><sup>2-</sup> concentration in soil water was measured in 1988 in each soil depth and ranged from 30.184 mg l<sup>-1</sup> (surface humus) to 42.031 mg l<sup>-1</sup> (0.25 m soil layer). The lowest values of SO<sub>4</sub><sup>2-</sup> concentration in soil water were

monitored in 2010 and varied from 8.02 mg l<sup>-1</sup> – humus layer, 13.43 mg l<sup>-1</sup> year 1989 in 0.10 m soil depth and 15.16 mg l<sup>-1</sup> SO<sub>4</sub> 0.25 m soil depth, respectively in the year 2005.

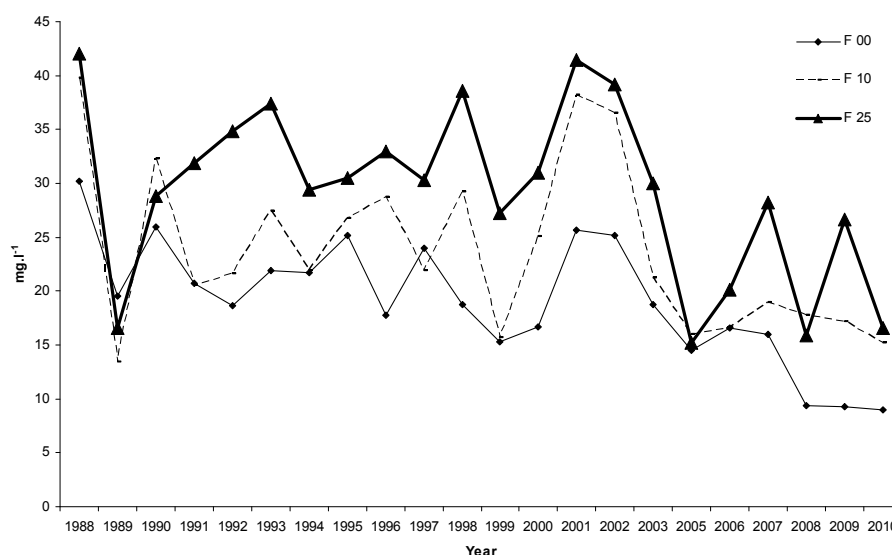


Fig. 1a. The concentration of SO<sub>4</sub><sup>2-</sup> in soil water in the Štiavnické vrchy Mts in years 1988–2010.

Table 1. Descriptive statistics of SO<sub>4</sub><sup>2-</sup> concentrations and S-SO<sub>4</sub> in the Štiavnické vrchy Mts in years 1988–2010

Plot	F00	F10	F25	Open plot	Forest plot
mg l <sup>-1</sup>					
Valid	22	22	22	16	16
Mean	19.10	23.72	29.32	14.92	17.13
Minimum	8.01	13.43	14.16	3.25	9.64
Maximum	30.18	39.73	42.03	25.33	24.68
Standard deviation	5.77	7.76	8.22	5.48	4.07
Standard Error	1.23	1.65	1.75	1.37	1.01
Coefficient of variation	30.40	32.60	27.90	36.70	23.80
kg ha <sup>-1</sup>					
Valid	22	22	22	16	16
Mean	18.9	20.5	26.7	24.3	19.1
Minimum	8.9	9.1	11.8	12.4	7.2
Maximum	35.1	37.9	52.6	45.2	33.3
Standard deviation	7.2	9.0	11.7	9.6	6.2
Standard Error	1.5	1.9	2.5	2.4	1.5
Coefficient of variation	38.3	43.9	43.8	39.5	32.5

F<sub>00</sub> – Lysimeter in the soil depth 0.0 m. F<sub>10</sub> – Lysimeter in the soil depth 0.1 m. F<sub>25</sub> – Lysimeter in the soil depth 0.25 m.

Table 2. Amount of sulphate [kg ha<sup>-1</sup>] in the Štiavnické vrchy Mts in years 1988–2010

Lysimeter	F <sub>00</sub>	F <sub>10</sub>	F <sub>25</sub>	Forest through fall	Open plot wet deposition
1988	22.2	29.52	52.61		
1989	10.89	13.5	20.1		
1990	22.2	31.7	33.3		
1991	11.1	17.5	27.3		
1992	11.1	17.1	33.1		
1993	11.7	14.1	15.6		
1994	21.9	32.2	47.4	7.2	12.4
1995	21.4	24.4	33.8	23.9	24.9
1996	26.8	34.5	37.2	19.8	32.3
1997	21.8	15.1	21.9	19.5	28.4
1998	17.3	26.6	29.4	15.8	23.6
1999	21.8	15.9	26.1	18.4	26.6
2000	16.1	16.8	18.2	12.9	20.8
2001	35.1	37.9	37.6	27.9	45.2
2002	33.8	32.2	41.1	19.5	27.8
2003	18.8	17.5	22.8	22.3	38.9
2005	20.4	14.2	11.8	16.2	18.1
2006	22.8	10.8	12.2	14.9	30.4
2007	16.5	9.7	17.2	13.7	13.6
2008	8.9	9.1	14.1	18.1	20.7
2009	9.5	9.9	12.5	22.3	12.5
2010	13.3	22.2	21.9	33.3	12.8
Mean	18.9	20.5	26.7	19.1	24.3
Total	414.8	451.9	587.1	305.8	389.1

F<sub>00</sub> – Lysimeter in the soil depth 0.0 m.; F<sub>10</sub> – Lysimeter in the soil depth 0.1 m.; F<sub>25</sub> – Lysimeter in the soil depth 0.25 m.

The yearly mean S-SO<sub>4</sub><sup>2-</sup> deposition to the surface humus was 18.9 kg ha<sup>-1</sup> yr<sup>-1</sup>. The highest values of S-SO<sub>4</sub><sup>2-</sup> were measured in 2001 with the yearly deposition 35.1 kg ha<sup>-1</sup> yr<sup>-1</sup> (Table 2). In this year the maximum sulphur contents were observed also in precipitation (45.2 kg ha<sup>-1</sup> yr<sup>-1</sup>), it corresponded to the SO<sub>4</sub><sup>2-</sup> concentration 21.84 mg l<sup>-1</sup>. The lowest annual flux of S-SO<sub>4</sub><sup>2-</sup> to the soil (8.9 kg ha<sup>-1</sup> yr<sup>-1</sup>) was obtained in 2008. The variability of results is cca 30% (Table 1).

In the stand in the 0.10 m soil depth was observed yearly 20.5 kg ha<sup>-1</sup> yr<sup>-1</sup> S-SO<sub>4</sub><sup>2-</sup>. It was caused by the accumulation ability of sulphur in the bottom soil horizons. This phenomenon was actual during the 1988–1999 and 2008–2010 period.

The variability of the results is approximately 43.9%. The highest concentration SO<sub>4</sub><sup>2-</sup> and sulphur content was measured in the 0.25 m soil depth. The mean concentration SO<sub>4</sub><sup>2-</sup> is 29.32 mg l<sup>-1</sup> and content in this soil horizon was 26.7 kg ha<sup>-1</sup> yr<sup>-1</sup>, it was higher

in about 29.3% than in the surface humus layer. The highest values of S-SO<sub>4</sub><sup>2-</sup> were observed at the beginning of the research in 1988 with average values 52.6 kg ha<sup>-1</sup> yr<sup>-1</sup> resp. 42.03 mg l<sup>-1</sup> SO<sub>4</sub><sup>2-</sup> concentration. The lowest values of sulphate sulphur content 11.8 kg ha<sup>-1</sup> yr<sup>-1</sup> in this soil depth were measured in 2005 (15.16 mg l<sup>-1</sup> SO<sub>4</sub><sup>2-</sup>). During the research period the content of sulphate sulphur in each soil depth declined, though the highest contents were observed in 2001 and 2002. The cause can be still in active lignite incineration plant in Nováky (Slovakia).

During the research the highest fluxes of sulphur in the soil were measured in the autumn months. The values of throughfall deposition and open field deposition during the study period decreased, the exception was year 2001. In this year were found the highest SO<sub>4</sub><sup>2-</sup> contents and concentrations in both depositions. At the open field the S-SO<sub>4</sub><sup>2-</sup> flux to the humus layer was 45.3 kg ha<sup>-1</sup> yr<sup>-1</sup> – 21.84 mg l<sup>-1</sup> SO<sub>4</sub><sup>2-</sup>. In year 2003 were found

total highest  $\text{SO}_4^{2-}$  concentration in the throughfall also at the open plot ( $25.33 \text{ mg l}^{-1} \text{SO}_4^{2-}$ ), also in the forest stand ( $24.68 \text{ mg l}^{-1} \text{SO}_4^{2-}$ ). The lowest concentration of  $\text{SO}_4^{2-}$  were found in the year 2010 with  $3.3 \text{ mg l}^{-1} \text{SO}_4^{2-}$  at the open plot and  $9.6 \text{ mg l}^{-1} \text{SO}_4^{2-}$  in the forest stand (Fig. 1b).

The results of testing of differences did not confirm the differences among the soil horizons. Evidently was observed the effect of the sulphur content in precipitation on the sulphur content in soil water. However were proved very significant differences ( $p < 0.001$ ) in the sulphur content in the 0.10 m and 0.25 m soil depth.

It implies that the present emissions in atmospheric deposition do not have effect on the sulphur content in the soil. On the contrary, the accumulation ability of sulphur in the bottom soil horizons was occurred and there sulphur persistently impacts on pedochemical processes.

## Discussion

The most important factors are the composition of parent material (COSBY et al., 1986) and the soil depth (MANDERSCHIED et al., 2000). KATUTIS et al. (2008) pointed at the composition of humus layer. Important factor is also the forest stand composition. To these important factors can we integrate physical and chemical soil properties. A negative strong correlation between the sulphur content in soil water and the precipitation amount was found (LINDROOS et al., 2006). Furthermore, the sulphur concentration in soil water is influ-

enced by sulphur content in atmospheric deposition (NOVOTNÝ et al., 2008). To these „natural“ determining factors belongs also the influence of the altitude (KOPEČ and GONDEK, 2002).

On the present the anthropogenic factors are predominant. These factors influenced the variability of the results from sulphur and its compounds research mainly in the form of deposition to all parts of forest and aquatic ecosystems.

DUBOVÁ AND BUBLINEC (2006) measured at the same stands in the Kremnické vrchy Mts  $25 \text{ kg S ha}^{-1} \text{ yr}^{-1}$  in bulk atmospheric deposition and  $24.9 \text{ kg S ha}^{-1} \text{ yr}^{-1}$  in throughfall deposition. They confirmed the highest sulphur content in precipitation during the 1994–1995 period. They presented that the maximum sulphur concentrations in this period were observed in atmospheric deposition and soil water in the whole Europe. According to the results from the emission monitoring in Europe in this period was observed the increasing of the emissions amount ( $>20.0 \text{ kg S ha}^{-1} \text{ yr}^{-1}$ ) in the southeast of the Great Britain and in the industry areas of Central Europe.

In the low polluted area the Kremnické vrchy Mts the highest values of sulphate sulphur were measured in the  $\text{H}_{00}$  horizon. Similar data presented KÁNA and KOPÁČEK (2005) whose research was done in forest soils of the Czech Republic. Interesting fact is the highly sulphur content in soil water than in throughfall after some years (4, 5, 6 years) after the cutting intervention aimed at the reduction of stocking. It can be explained by high sulphur concentrations and sulphur content in horizontal precipitation (mist, dew) (ŠKVARENINA,

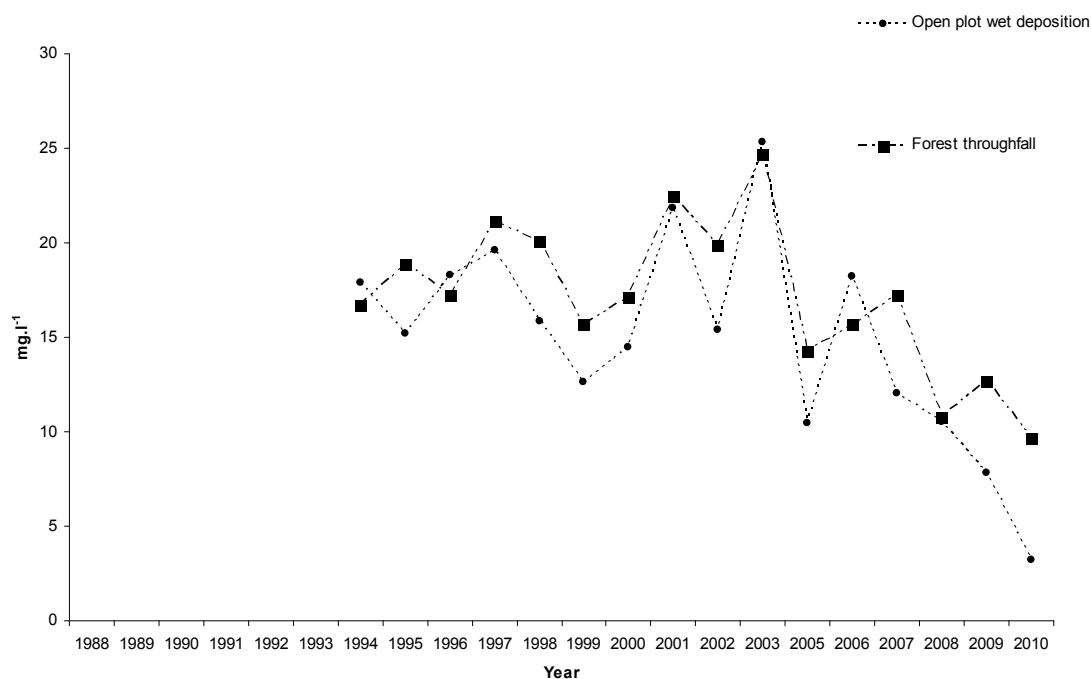


Fig. 1b. The concentration of  $\text{SO}_4^{2-}$  in the precipitation and throughfall in the Štiavnické vrchy Mts in years 1988–2010.

1998). YAMADA et al. (2001) found out the sulphur concentration in throughfall and in stemflow in the south-west Japan is higher than in open bulk precipitation.

At the research plots in the Štiavnické vrchy Mts (Slovakia) in a 180–190 years old oak stand (altitude 680 m a.s.l.) the soil received via throughfall 23 kg S ha<sup>-1</sup> (KUNCA, 2007).

Similar occurrences of the highest sulphate sulfur contents in precipitation and soil water in the autumn and the winter months found out BUBLINEC and DUBOVÁ (1995).

The significance of differences between the years in the sulphur content in soil water and in precipitation was statistically insignificant.

## Conclusions

The situation in the polluted area the Štiavnické vrchy Mts is principally different in two facts: the sulphur content and its accumulation in the soil horizons. The lowest mean content of S-SO<sub>4</sub><sup>2-</sup> (18.9 kg ha<sup>-1</sup> yr<sup>-1</sup>) was observed in the surface humus and the highest mean content (26.7 kg ha<sup>-1</sup> yr<sup>-1</sup>) in the 0.25 m depth. During the research the total S-SO<sub>4</sub><sup>2-</sup> flux to this soil depth was 587.1 kg ha<sup>-1</sup>. Interesting data were observed in throughfall deposition, the sulphur content in it was lower than in soil water. It can be caused by the filtration effect of the forest canopy or the accumulated sulphur content in soil.

The testing of differences showed no significant difference between the sulphur content in precipitation between the experimental sites. In contrast, the differences between the sulphur content in 0.10 m and 0.25 m soil depth between the study areas were statistically significant. It can be explained by accumulation of sulphur in soil in these areas till 1990. We proved that the precipitation amount and the sulphur content in atmospheric deposition significantly influenced the sulphur content and concentration in each soil horizon.

Temporal variability in sulphur content pointed out the occurrence of the maximum S-SO<sub>4</sub><sup>2-</sup> values in the autumn months. The lowest S-SO<sub>4</sub><sup>2-</sup> values were observed in the summer and in the spring. To this fact are also linked the SO<sub>4</sub><sup>2-</sup> concentrations.

On the basis of the mean sulphur fluxes to each soil horizon and the sulphur content in precipitation during the research the forests of the Štiavnické vrchy Mts are presented as an area with lower sulphur load not only on the national level, but on the European level. The exception is the sulphur content in precipitation at the open field during years 1996, 2001, 2003 and 2006. In these years was the sulphur critical limit exceeded.

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## Koncentrácia $\text{SO}_4^{2-}$ a množstvo $\text{S-SO}_4^{2-}$ v pôdnych vodách a zrážkach v bučinách Štiavnických vrchov, Slovensko

### Súhrn

V práci vyhodnocujeme výsledky koncentrácie síranových iónov a množstva síranovej síry v pôdnej vode a zrážkach v podmienkach bukoveho porastu Štiavnických vrchov. Priemerná koncentrácia  $\text{SO}_4^{2-}$  kolíše v rozmedzí  $19,06 \text{ mg l}^{-1}$  vo vrstve povrchového horizontu v lese do  $29,32 \text{ mg l}^{-1}$  vo vrstve  $0,25 \text{ m}$  pod povrchom zeme. Do pôdy sa počas doby výskumu deponovalo  $415 \text{ kg S-SO}_4^{2-} \text{ ha}^{-1}$  – do vrstvy povrchového humusu a  $587 \text{ kg S-SO}_4^{2-} \text{ ha}^{-1}$  – do hĺbky  $0,25 \text{ m}$ . Najvyššia koncentrácia  $\text{SO}_4^{2-}$  bola zaznamenaná v roku 1988 vo všetkých pôdnych hĺbkach, naopak najnižšia v roku 2010 vo vrstve povrchového humusu s  $8,02 \text{ mg l}^{-1}$ . Koncentrácia  $\text{SO}_4^{2-}$  smerom do hĺbky stúpala.

Regresnou analýzou bol potvrdený vplyv úhrnu zrážok a množstva síry v zrážkach na obsah a koncentráciu síry v pôdnej vode. Studentovým t-testom bola potvrdená rozdielnosť množstva síry a koncentrácie  $\text{SO}_4^{2-}$  v pôdnych hĺbkach  $0,1 \text{ m}$  a  $0,25 \text{ m}$ . Významnosť medzi jednotlivými rokmi sa nepreukázala.

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