

Sulphate Contamination, pH and Conductivity of Forest Soils in Two Neighbouring Mountains with Different Pollution in Slovakia from 1989 to 2013

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Abstract

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The Štiavnické vrchy Mts. were strongly affected by pollution mostly from an aluminium plant in 1953–1989. This paper compares contamination of soils between Štiavnické vrchy Mts. and the neighbouring little polluted Kremnické vrchy Mts. from results of a 25-year study. After a decrease of emissions in Slovakia at the beginning of the 1990s the sulphate sulphur content, acidity and conductivity of soil water have decreased only on the surface and at a depth of 0.10 m at the study site in the Štiavnické vrchy Mts. At the depth 0.25 m the increase of sulphurization (23.68 kg/ha S-SO₄²⁻ per year) and acidity (pH 4.92) was observed. During the research, the total sulphate sulphur influx to this soil depth was 568.3 kg/ha. The average sulphur input in the study areas of the Kremnické vrchy Mts. decreased with depth: from 18.48 kg/ha/year in the surface humus to 6.85 kg/ha/year at a depth of 0.25 m. The maximum sulphur influx at the open plot was 24.06 kg/ha/year and in total 553.34 kg S-SO₄²⁻. A small increase of acidity at soil depths of 0.25 m at some sites was observed also in the Kremnické vrchy Mts. Regression analysis revealed a statistically significant influence of sulphate sulphur content in the atmospheric precipitation on the sulphur amount in the soil water. A significant correlation was also observed between the precipitation amount and the sulphur content in soil water. Data from monitoring revealed significant differences between the sulphur amounts at depths of 0.10 m and 0.25 m in these study areas.

Keywords: aluminium plant; conductivity; soil acidification; soil water; sulphur; Western Carpathians

Research on sulphate contamination in precipitation water and in throughfall is very important. This pollution belongs to the main factors involved in the process of soil acidification. Sulphur, nitrogen and solid particles account for the major part of acidification in the atmosphere (PAVLŮ *et al.* 2015) and in the forest soil development.

Acidification is a long-term and cumulative process (HRUŠKA *et al.* 2001) exhibiting dynamic development. The alarming degradation of environment, especially forest soils, unveiled urgent needs to set deposition limits for forest ecosystems – as valid tools for governments in designing mitigation strategies for sulphur and

nitrogen emissions in Europe and in North America (MATZNER & MEIWES 1994; JANDL *et al.* 2012).

Reduction of industry, reduction of coal mining in Poland and in the former East Germany and conversion of industry across the whole “Eastern Block” in the 1990s contributed to a decrease in sulphur and nitrogen emissions (ZAPLETAL 2006).

RIEK *et al.* (2012) reported a decrease in sulphur influx into the soil from 63 to 5.3 kg/ha in southern Brandenburg during the 1986–1989 period.

Nevertheless, the danger of acidification is a still persisting ecological problem of extraordinary importance (ALEWEL *et al.* 2000); mainly due to its

long-term character and due to the fact that sulphur accumulates in deeper soil layers.

We have studied pollution in two neighbouring mountains which are close to three towns Žiar nad Hronom, Banská Bystrica and Zvolen. Near Žiar nad Hronom there is an aluminium plant. This plant produced great amount of emissions from 1953 to 1998 so that this area was and remains one of the most polluted in Slovakia, with the main contaminants being oxides of sulphur, nitrogen, fluorine, arsenic, cadmium and ozone. SO₂ and fluorine had the worst effect on beech forests in the Štiavnické vrchy Mts. Since the end of the 1990s, the pollutant level has been decreasing rapidly due to the modernization of production processes in the plant, so that the fluorine concentration was reduced to the tolerable level of 1 µg/m³ (URMINSKÁ *et al.* 2000).

The aim of this paper is to assess sulphurization and acidification trends in the studied areas after a decrease in emissions.

MATERIAL AND METHODS

Study areas. In the Kremnické vrchy Mts., the research was conducted on 2 study sites, each 40–50 × 75 m in size. H site: Forest stand by 1988. Clear cutting in 1989. Natural reforestation in the following years. Now after 24 years, there is a young beech forest of 12 m height. K site: Forest without cutting. The prevailing soil-forming substrate consists of andesite tuff agglomerates from which a saturated variant of andosolic Cambisol with the skeleton content increasing with depth has been formed. The soil body is layered, composed of the main and the basal layer system (PICHLEK *et al.* 2006, 2009a, b). The soils were formed in the Pleistocene period under the active presence of solifluction processes and have these basic macromorphological characteristics: soil at the stand plot is eutrophic andosolic Cambisol, humus form acid mull. Oo: 3 cm, layer of beech leaves and twigs. Oof: 0.5 cm, layer of partially decomposed beech leaves. Aoq: 0–8 cm, black-brown, freshly moist, with moderate amounts of roots, very fine andesitic gravel 10–15%. Bv1: 8–45 cm, dark brown, clay loam, lumped, coarse andesitic gravel 20%. Bv2: 45–75 cm, brown, clay loam to clayey, compact, moist, coarse tuffaceous gravel and stones 30–40%. C: 75–90 cm, parent rock with small layers of brown clay, moist, without roots. R: > 90 cm, solid tuff. The Fieldes-Perrot test for B horizons was slightly positive. Open plot: soil is eutrophic andosolic Cambisol, humus form acid

mull. Oo1: 1–2 cm, layer of beech leaves and twigs. Oof: 0.5 cm, layer of partially decomposed leaves of beech and grasses (*Carex pilosa*). Aoq: 0–8 cm, black-brown, loamy, with fine moderate scrums, freshly moist, with moderate amounts of roots, very fine to coarse andesitic gravel 5%. Bv1: 8–40 cm, dark brown, clay loam, lumped, coarse andesitic gravel 20%. Bv2: 40–60 cm, dark brown, around the tuffaceous skeleton with grey and yellow stains of sandy clay, very fine to coarse gravel 40%. B/C: 60–85 cm, brown with fine yellow shade, clay-loam with admixture of sand, lumped, strongly compact, moist, coarse gravel and stones 60%. R: 85–110 cm, soft tuff, rare with red colour. The Fieldes-Perrot test for B and B/C horizons was slightly positive (KUKLA 2002).

In the Štiavnické vrchy Mts., there are two study sites: K site: Forest without cutting. The atmospheric deposition was also measured at the open site. The research plot is facing north or north-west. The second study area is a meadow (open plot). The plots in the Štiavnické vrchy Mts. are in close proximity (1750 m) of a huge industrial pollution source (aluminium plant), whereas the plots in the Kremnické vrchy Mts. are 18 km away from this plant (Figure 1).

Substances contaminating the Štiavnické vrchy Mts. originated from local sources (aluminium plant, energy plants, local waste dumping site, highway system built recently), and they represented fluorine, sulphur and nitrogen oxides, arsenic, cadmium, ozone, solid particles, and several others. At the end of the last century, the emission load considerably decreased, primarily due to modernising the production processes and due to new environmental legislation in force (Table 1).

Lysimeter waters. Lysimeter waters were sampled by plastic collectors (each 1000 cm² in area) in three soil horizons both in the Kremnické vrchy Mts. and in the Štiavnické vrchy Mts. The first series of collectors was placed on the surface (organic layer – K₀₀),

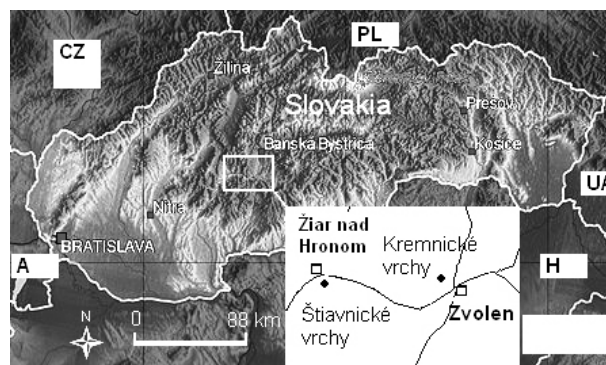


Figure 1. The location of research plots

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the second was installed at a depth of 0.10 m (upper mineral layer – K_{10}), the third at 0.25 m (lower mineral layer – K_{25}) (KUKLA 2002). The collectors on the surface were covered with old leaves and twigs throughout the year. Also, at two research sites in the Kremnické vrchy Mts. the soil solution was sampled by the collectors at horizons 0.6 and 0.8 m. Samples were analysed and evaluated (JANÍK *et al.* 2012a, b) monthly since 1988.

Throughfall and bulk precipitation. The waters were collected into bottles with the sampling area of 660 cm² with a funnel in their caps. Ten sampling devices were installed both in the open field and at stand sites. The samples were taken once a month and after strong precipitation. The representative samples were defined as a mixture from all the collectors in the stand and at the open plot (TUŽINSKÝ 2002). The amounts were measured to the nearest 10⁻² l.

Sulphates. Sulphate ions were determined by titration with lead nitrate in the presence of dithizone as an indicator and results were converted to sulphate sulphur content.

Acidity. The values of pH were measured potentiometrically in the laboratory from the samples of water with the glass high ohmic electrode.

Conductivity. The values of conductivity were determined electrometrically at 20°C in the laboratory.

Statistical methods. Data were processed by the Statistica 7 programme. Normality was tested by

the Shapiro-Wilk W test. Student's *t*-test for dependent variables was used for testing significance of differences between the horizons of both study localities. The Statgraphics programme (Version 2.0) two-sample test was conducted to confirm results. Relationships between precipitation amount and sulphate amount in precipitation and soil water were described by the simple regression analysis.

RESULTS

Sulphates

Kremnické vrchy Mts. We studied the concentration of sulphates at K site. The highest annual sulphate sulphur influx of 33.9 kg/ha/year was recorded in the K_{00} layer in 1995 (Figure 2). An annual minimum of 1.48 kg/ha/year at K_{00} was observed in 2011 (Figure 2). The mean content at a depth of 0.1 m is 31% lower than that on the surface at both sites. The lowest mean sulphur values of 0.88 kg/ha/year at K_{25} were measured in 2011 and also in 1989. The highest values were recorded in autumn, the lowest in spring and summer.

The maximum sulphur amount in precipitation was recorded in the rainy year 2002. We determined 67.2 kg/ha at H site with the total annual precipitation amount of 1532 mm (mean 653 mm) and 67.24 kg/ha at K site with the annual throughfall of 925 mm.

Štiavnické vrchy Mts. The maximum annual sulphur influx of 35.1 kg/ha/year was registered in 2001

Table 1. Basic characteristics of the EES Kremnické vrchy Mts. and Štiavnické vrchy Mts.

| Orographic unit | Kremnické vrchy Mts. | Štiavnické vrchy Mts. |
|-----------------------------|---|---|
| Partial research plot | ecological-experimental stationary Kováčová | research monitoring plot Žiar nad Hronom – Horné Opatovce |
| Localisation | 48°38'10"N, 19°04'08"E | 48°35'08"N, 8°51'10"E |
| Altitude (m a.s.l.) | 470–490 | 470 |
| Exposition | SW | NW |
| Slope (°) | 20 | 15 |
| Geological substrate | andesite, tuffaceous agglomerates | ryolites tufits |
| Soil type | Cambisol (Andosol) saturated | Cambisol, Luvisol |
| Humus form | mull | moder |
| Throughfall (mm) | 653 | 750 |
| Temperature (°C) | 8.3 | 8.0–8.5 |
| Forest type groups | Fagetum pauper inferiora | Fagetum pauper |
| Tree composition (%) | beech 95, fir 2, hornbeam 2, oak 1 | beech 100 |
| Age of mature stand (years) | 105–110 | 100–105 |
| Area (ha) | 1.2 | 0.15 |

SCHIEBER *et al.* (2013), KUKLOVÁ *et al.* (2015)

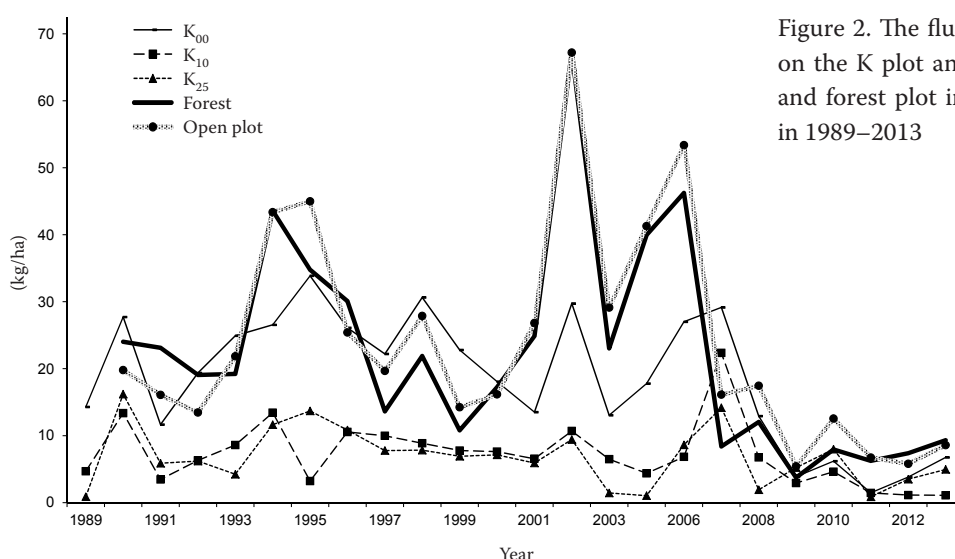


Figure 2. The flux of S-SO_4^{2-} to soil horizons on the K plot and precipitation on the open and forest plot in the Kremnické vrchy Mts. in 1989–2013

(Figure 3). The minimum value of 8.87 kg/ha/year was measured in 2000. The coefficient of variation is 39.5%. This coefficient is lower than that in the Kremnické vrchy Mts. The amount of sulphur in the layer of 0.10 m was 6.75% higher than that in the surface horizon. This trend was distinct mainly from 1988 to 1999. During the study period, the highest amount of sulphur was at the depth of 0.25 m. It was on average 28.8% more than in K_{00} . The highest annual sulphate sulphur value of 52.6 kg/ha/year was registered at the beginning of the study in 1988, the lowest in 2005 (11.8 kg). Soil water manifested decreasing trends in all horizons, except the years

2001 and 2002. The probable cause of pollution is a still active power station in Nováky. The highest values of sulphur input were recorded in autumn.

The maximum sulphur content of 45.19 kg/ha in rainfall at the open site was recorded in 2001. The maximum content of 33.3 kg/ha in throughfall in the stand was determined in 2010. It is interesting that these maximum values were lower than those recorded in the low polluted Kremnické vrchy Mts.

Very significant differences were recorded in soil sulphur at depths of 0.10 and 0.25 m between the plots in the Kremnické vrchy Mts. and in the Štiavnické vrchy Mts. (Table 2). We can say that today the emitted

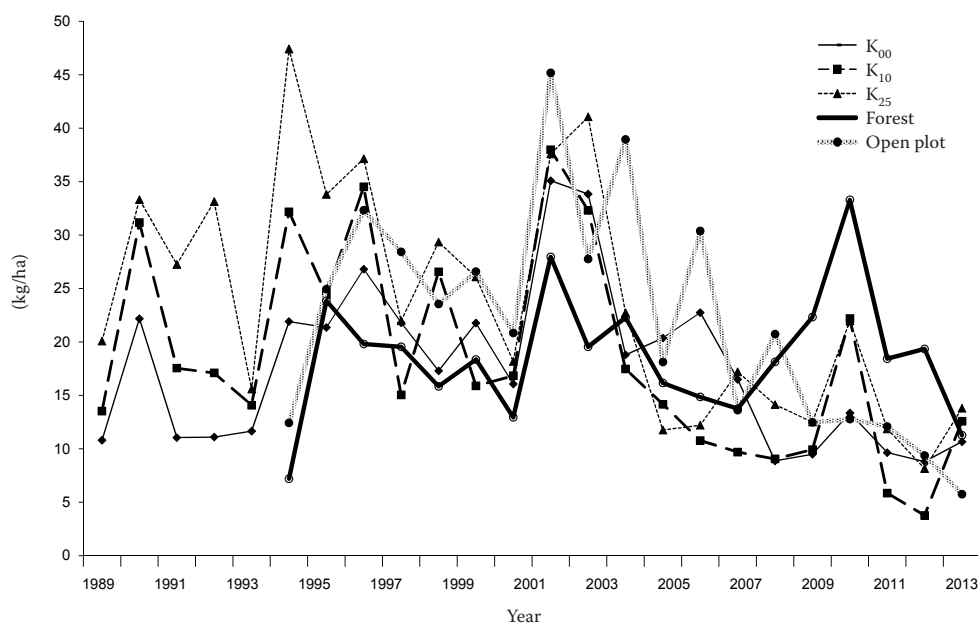


Figure 3. The flux of S-SO_4^{2-} to soil horizons on the K plot and precipitation on the open and forest plot in the Štiavnické vrchy Mts. in 1989–2013

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pollutants in atmospheric deposition do not exert a marked influence on soil sulphur content. In contrast, there was an evident long-term accumulation of sulphur in lower soil horizons in the Štiavnické vrchy Mts. Inter-annual differences in sulphur content in soil water and in precipitation at the same plot have not been found statistically significant.

Acidity

Kremnické vrchy Mts. We observed a small increase of soil water acidity at all sites and horizons. The highest increase of acidity was registered at the deepest horizons K₂₅. Although the monthly

amplitude at the horizons was 1.5 with a minimum of 4.03 pH in August 2010, these soil waters remain weakly acid.

Acidity of precipitation diminished at all sites. The most acid years were 1988, 1989, 1990 and 1997 (Figure 4). The lowest annual pH ranged from 4.54 to 5.44. The acidity of throughfall was smaller than that of precipitation at the clear-cut site.

Štiavnické vrchy Mts. Acidity of soil water decreased only on the surface. At the horizons K₁₀ and K₂₅, a decrease of pH was observed. Soil water at a depth of 0.25 m was always the most acid with a minimum of 3.7 pH in April 2006 (Figure 5). The acidity diminished in periods of rains or thaws.

Table 2. Testing of S-SO₄²⁻, pH values in the Kremnické vrchy Mts. and Štiavnické vrchy Mts. between partial plots and soil horizons in 1989–2013

| Soil horizons | Kremnické vrchy Mts. | | | | | Štiavnické vrchy Mts. | | | | |
|--|----------------------|-----------------|-----------------|---------------|--------------------|-----------------------|-----------------|-----------------|---------------|--------------------|
| | K ₀₀ | K ₁₀ | K ₂₅ | K throughfall | Bulk precipitation | K ₀₀ | K ₁₀ | K ₂₅ | K throughfall | Bulk precipitation |
| S-SO₄²⁻ (kg/ha) | | | | | | | | | | |
| Kremnické vrchy Mts. | | | | | | | | | | |
| K ₀₀ | – | 5.1** | 5.4** | –1.0 | –1.5 | 0.4 | –0.0 | –1.8 | – | – |
| K ₁₀ | –5.1** | – | 0.3 | –4.5** | –4.9** | –5.7** | –5.2** | –6.8** | – | – |
| K ₂₅ | –5.4** | –0.3 | – | –4.7** | –5.0** | –6.1** | –5.5** | –7.1** | – | – |
| K throughfall | 1.0 | 4.5** | 4.7** | – | –0.4 | – | – | – | 1.0 | – |
| Bulk precipitation | 1.5 | 4.9** | 5.1** | 0.4 | – | – | – | – | – | 0.5 |
| Štiavnické vrchy Mts. | | | | | | | | | | |
| K ₀₀ | 0.4 | 5.8** | 6.1** | – | 1.5 | – | –0.4 | –2.3** | –0.5 | 1.6 |
| K ₁₀ | 0.0 | 5.2** | 5.5** | 4.5** | 1.5 | 0.4 | – | –1.8 | –0.0 | –1.1 |
| K ₂₅ | 1.8 | 6.8** | 7.1** | 4.7** | 5.1** | 2.2** | 1.8 | – | 1.8 | 0.5 |
| K throughfall | – | – | – | 0.9 | – | 0.5 | 0.0 | –1.8 | – | 1.2 |
| Bulk precipitation | – | – | – | – | 0.5 | 1.6 | 1.1 | –0.5 | 1.2 | – |
| pH | | | | | | | | | | |
| Kremnické vrchy Mts. | | | | | | | | | | |
| K ₀₀ | – | –2.2** | 1.6 | 6.3** | 4.7** | 7.9** | 13.7** | 17.7** | – | – |
| K ₁₀ | 2.2** | – | 3.4** | 8.7** | 6.4** | 10.5** | 18.4** | 23.6** | – | – |
| K ₂₅ | –1.6 | –3.4** | – | 3.7** | 2.9** | 4.9** | 8.5** | 10.9** | – | – |
| K throughfall | –6.3** | –8.7** | –3.7** | – | –0.3 | – | – | – | –1.7 | – |
| Bulk precipitation | –4.7** | –6.3** | –2.9** | 0.3 | – | – | – | – | –1.5 | – |
| Štiavnické vrchy Mts. | | | | | | | | | | |
| K ₀₀ | –7.9** | 10.5** | –4.9** | – | – | – | 3.4** | 6.2** | –2.8** | –3.6** |
| K ₁₀ | –13.7** | 18.4** | –8.5** | – | – | –3.4** | – | 3.6** | –6.4** | –7.7** |
| K ₂₅ | –17.7** | 23.6** | –10.9** | – | – | –6.2** | –3.6** | – | 8.9** | 10.7** |
| K throughfall | – | – | – | 0.3 | – | 2.8** | 6.4** | –8.9** | – | –0.6 |
| Bulk precipitation | – | – | – | 1.7 | – | 3.6** | 7.7** | –10.7** | 0.6 | – |

t-test; **statistically significant at *P* < 0.01

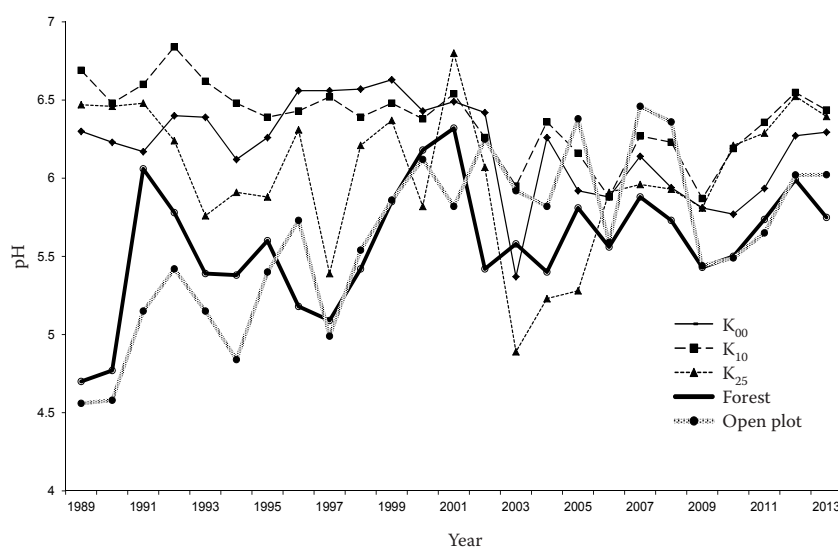


Figure 4. The pH values in soil horizons on the K plot and precipitation on the open and forest plot in the Kremnické vrchy Mts. in 1989–2013

Table 3. Basic data on beech stands on the plots after experimental cutting in February 1989 at the Ecological Experimental Site Kremnické vrchy Mts. (Western Carpathians Mts.)

| Plots | Year | Stem density (No./ha) | Height (m) | DBH (cm) | Stocking density | Basal area (m ² /ha) | Area (m ²) | Height of beech seedling (cm) |
|-------------|------|--------------------------|---------------|-------------|---------------------|------------------------------------|---------------------------|----------------------------------|
| K-control | 1990 | 700 | 23.6 | 23.9 | 0.90 | 40.90 | 1500 | 6 |
| | 1996 | 633 | | | 0.87 | 41.20 | | 20 |
| | 2002 | | 26.3 | 27.6 | 1.00 | | | 50 |
| H-clear cut | 1990 | 0 | | | 0.00 | | 4000 | 15 |
| | 1996 | | | | 0.00 | | | 210 |
| | 2002 | | | | 0.00 | | | 480 |

DBH – mean diameter at breast height

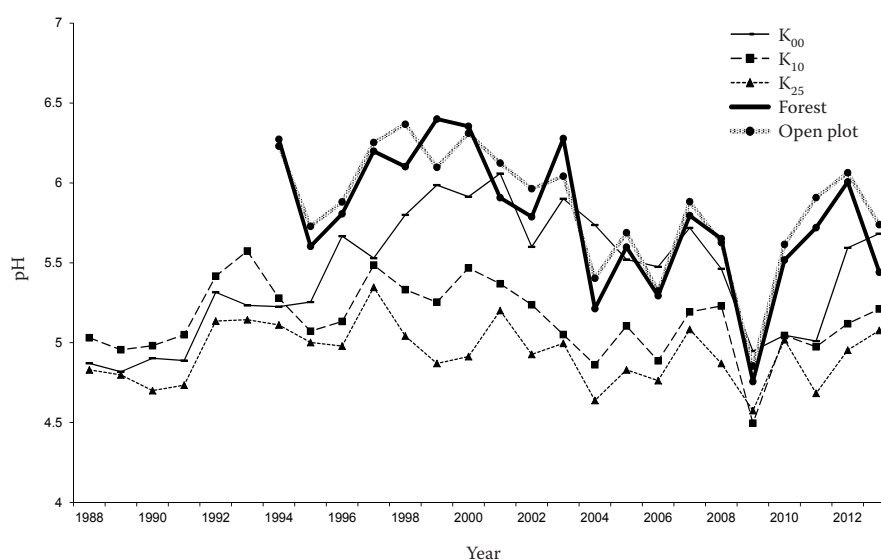


Figure 5. The pH values in soil horizons on the K plot and precipitation on the open and forest plot in the Štiavnické vrchy Mts. in 1989–2013

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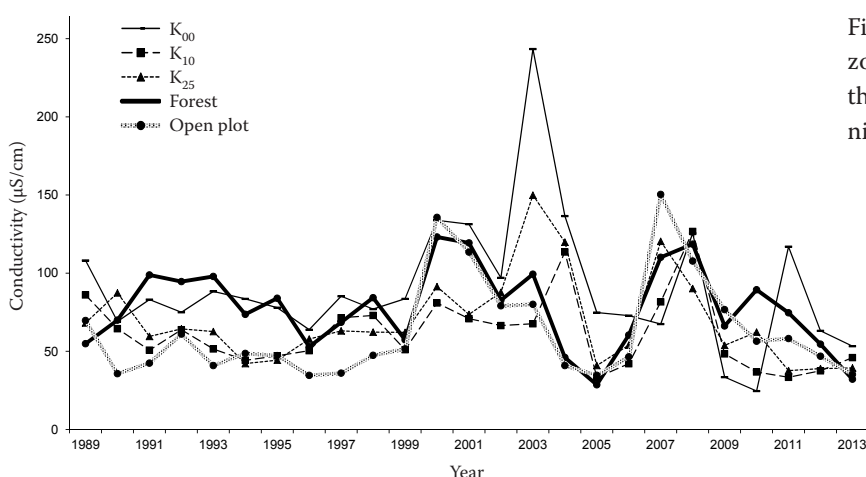


Figure 6. The conductivity in soil horizons on the K plot and precipitation on the open and forest plot in the Kremnické vrchy Mts. in 1989–2013

Acidity of precipitation at the open plot and in the forest during the 1994–2013 period, described by linear regression, increased because of three acid years 2004, 2006 and 2009. In 2009 we registered summer rains with acidity from 4.3 to 3.99 pH. The performed tests revealed significant differences between the individual soil horizons and also between localities (Table 2).

Conductivity

Kremnické vrchy Mts. Conductivity of soil water by linear regression slowly decreased at sites. A small increase of values with the annual maximum of 243.37 μS was measured only at the horizons K_{00} in 2003 (Table 3). Throughfall conductivity decreased at all sites from 2002. Inter-annual values ranged from 34.58 to 150.34 μS at the open plot. We measured low values also in 1996. In 2007 we observed annual maxima (Figure 6).

Štiavnické vrchy Mts. Conductivity of soil water tends to fluctuate from year to year. On the surface, annual means decreased. At the horizons of 0.1 m and 0.25 m, values slowly increased. The highest annual values were often observed at K_{25} . The maximum of 198.66 μS was measured in 2003 (Figure 7).

Conductivity of precipitation at the open plot decreased. In the last nine years, we registered lower annual maxima ranging from 28.76 to 90.41 μS . Conductivity of throughfall was higher and ranged from 64.07 to 191.75 μS . The annual maximum of 246.82 μS was recorded in the stand in 2003.

The amount of SO_4^{2-} , pH values and conductivity of the H plot in the Kremnické vrchy Mts. are given in Figures 8–10.

DISCUSSION

The highest amounts of sulphate sulphur in the Kremnické vrchy Mts. were measured in the H_{00} hori-

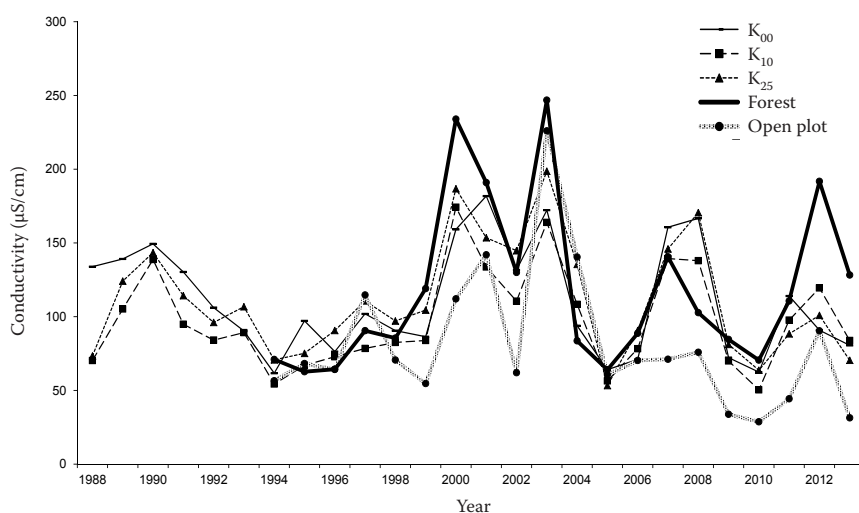


Figure 7. The conductivity in soil horizons on the K plot and precipitation on the open and forest plot in the Štiavnické vrchy Mts. in 1989–2013

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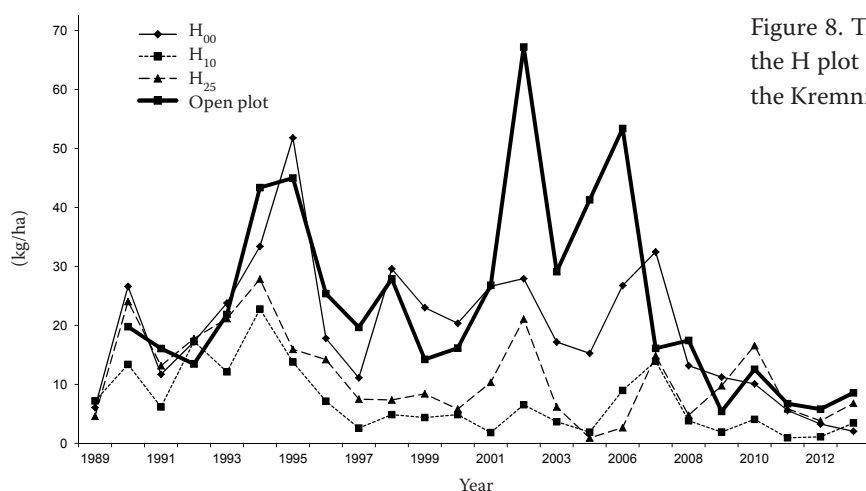


Figure 8. The flux of S-SO_4^{2-} to soil horizons on the H plot and precipitation on the open plot in the Kremnické vrchy Mts. in 1989–2013

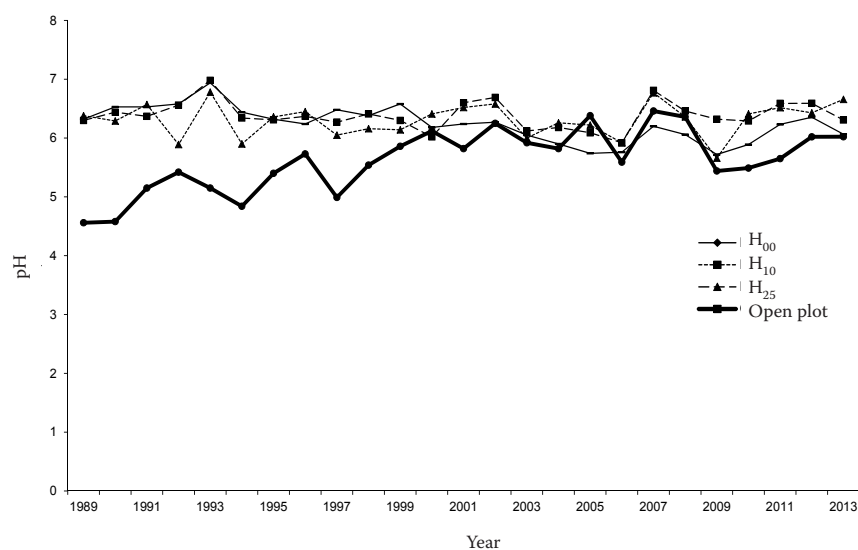


Figure 9. The pH values in soil horizons on the H plot and precipitation on the open plot in the Kremnické vrchy Mts. in 1989–2013

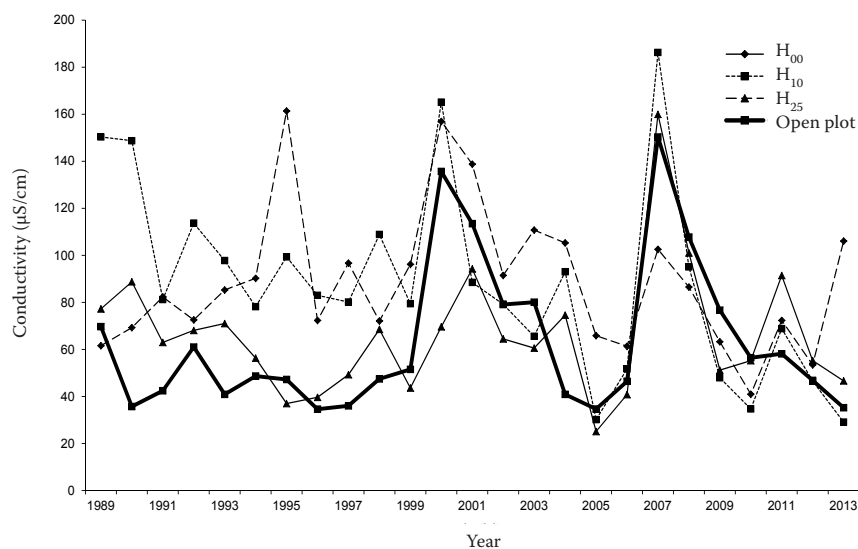


Figure 10. The conductivity in soil horizons on the H plot and precipitation on the open plot in the Kremnické vrchy Mts. in 1989–2013

zon, which is in accordance with the values obtained by KAŇA and KOPÁČEK (2005) and TEJNECKÝ *et al.* (2013) as well in soils of selected areas in the

Czech Republic. It is interesting that the amount of sulphur in soil water in H_{00} horizon 4–6 years after the clear-cut was higher than that in the throughfall

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before cutting. MINĎÁŠ *et al.* (2001) explained this phenomenon by an increase of sulphate concentration from horizontal precipitation (mist, dew), not involved in our total precipitation amounts. TESAŘ *et al.* (2004) measured from 0.31 to 77.6 mg/l in horizontal precipitation in forests of the Šumava Mts. The annual sulphur deposition in mist can reach 14.37 kg/ha/year and it can account for 79.6% of total annual deposition.

SZAREK-ŁUKASZEWSKA (2003) reported the annual influx of S-SO_4^{2-} to the soil amounting to 7.3–8.9 kg/ha in oak-pine forests in southern Poland (Niepolomice). It confirms a decrease of sulphur pollution during the last twenty years.

The regression analysis revealed an important influence of precipitation amount on the amount of sulphate sulphur in soil water. The value of correlation coefficient in the stand was 0.93. The sulphur content in the total precipitation amount had a very substantial influence on a sulphur amount in the surface horizon in the stand in the Kremnické vrchy Mts.

Very significant differences were recorded in soil sulphur at depths of 0.10 and 0.25 m between the plots in the Kremnické vrchy Mts. and in the Štiavnické vrchy Mts. We can say that today the emitted pollutants in atmospheric deposition do not exert a marked influence on soil sulphur content. In contrast there was an evident long-term accumulation of sulphur in lower soil horizons in the Štiavnické vrchy Mts. This may have been due to the bulk precipitation and the chemical composition of soil.

Concentrations of S-SO_4^{2-} in southern Poland were higher in throughfall than in bulk precipitation (SZAREK-ŁUKASZEWSKA 1999). Also in our research, the throughfall in beech stands contained a larger amount of sulphates than precipitation at the open sites in both mountains.

The acidification and sulphates reached the deepest horizons of 0.6 and 0.8 m, which were observed in the low polluted Kremnické vrchy Mts. We registered the fluctuation of pH and conductivity with a small increase of values. MEIWES *et al.* (1998) confirmed that the sulphate pollution can reach only horizons from the surface to a layer of 0.4 m.

The pH value of water percolated through the surface humus may be lower (reported from the Želivka and Rájec localities in the Czech Republic, LOCHMAN 1997) or somewhat higher (documented in southern England, GOWER *et al.* 1995) than the original pH value on the surface. As far as we know,

the highest pH increase in the water percolated under the surface humus was observed in the Hukavský grúň locality, Poľana Mts., Slovakia (MINĎÁŠ 2005).

The pH values decreased with increasing depth. The opposite has been reported for lysimeter water sampled on a clear-cut plot at the Biely Váh locality (High Tatras Mts.) for which KUKLA (2002) obtained a pH value of 6.67 in the surface humus and 7.97 at 0.1 m. MIHÁLIK *et al.* (1993) compared pH values between bulk precipitation and beech throughfall in the Mláčik locality in the Štiavnické vrchy Mts., and they obtained (in 1990) values from 4.92 on the open plot to 5.29 in beech throughfall in case of 130 years old stands and to 5.26 in young forest.

MINĎÁŠ (2006) speaks about a distinct change after percolation through the surface humus where the original precipitation pH values were mostly shifted from the acid or strongly acid range (throughfall) to the moderately acid range (statistically significant at $\alpha = 0.01$).

R-squared values of the linear regression of acidity and conductivity are mainly low. These approximations refer to slow-moving natural changes with various fluctuations and local differences.

It is important to mention the values of stemflow, at least by means of discussion, which are closely connected with the balance of S-SO_4^{2-} input into the soil horizon as well as pH values and conductivity. KANTOR (1985), AUGUSTO and RANGER (2001), ANDRE *et al.* (2007) explained this fact by different precipitation amounts in the forests, with a significant effect of the plant species. According to JÓŹWIAK *et al.* (2013) pH values of the beech stand litter horizon in southeastern Poland vary with the distance from the stem. The lowest pH values (3.36) were measured at a distance of 0.15 m from the stem. The pH values reach 3.61 at a distance of 1 m. Some differences were also confirmed in the annual dynamics. KRUSZYK *et al.* (2015) came to similar findings about S-SO_4^{2-} , the highest S-SO_4^{2-} values were measured very close to the stem of beech stands in northeastern Poland.

CONCLUSION

The average sulphur input in study areas of the Kremnické vrchy Mts. decreased with depth: from 19.7 kg/ha/year on the surface to 7.1 kg/ha/year at a depth of 0.25 m, but it increased in the Štiavnické vrchy Mts.: from 18.9 kg/ha/year on the surface to 26 kg/ha/year at 0.25 m. The average SO_4^{2-} concentration increased: from 14 mg/l on the surface to

16.17 mg/l at 0.25 m in the Kremnické vrchy Mts. and from 18.73 mg/l on the surface to 28.8 mg/l at 0.25 m in the Štiavnické vrchy Mts. During the research, the total sulphur influx into this soil depth in the Štiavnické vrchy was 647.04 kg/ha (159.35 kg/ha to the K_{25} layer in the Kremnické Vrchy Mts.).

In the last 25 years the emissions of sulphate have decreased both in the Štiavnické vrchy Mts. and the Kremnické vrchy Mts. The stands in the Kremnické vrchy Mts. represent one of the areas with the lowest sulphur load in Central Europe. The critical values of sulphur amounts on study sites were exceeded in 2002, 2005 and 2006 only in precipitation but not at soil depths of 0.1 and 0.25 m. In contrast, the limits for soil sulphur content were distinctly exceeded in the neighbouring Štiavnické vrchy Mts. in 1988, 1990, 1992, 1994, 1996, 2001 and 2002.

The observed soil in the Štiavnické vrchy Mts. remains under the influence of long-term acidification, which moves to the deepest horizons. Meanwhile, we determined a small increase of acidity at the depths of 0.25 m in the Kremnické vrchy Mts.

Despite the decreasing conductivity of precipitation we observed a small increase of values in soil water in some horizons in both mountains. During the research period, the highest values were at K_{25} , which was influenced by precipitation amounts.

We can compare the differences in annual pollution between the mountains during the last 20 years. In the Štiavnické vrchy Mts., we registered the maximum sulphate amount in precipitation in 2001, the most acid rains in 2004, 2006, 2009 and the highest conductivity of rains in 2003. In the Kremnické vrchy Mts., we observed the maximum sulphate pollution in 1995, 2002, 2006, the most acid rains came in 1997, 2009, 2010 and the highest values of conductivity were measured in 2000 and 2001.

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