Throughfall chemistry and atmospheric deposition in a Norway spruce – subalpine climax forest in the Poľana Biosphere reserve, Slovakia

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Abstract

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This work deals with chemistry of precipitation and atmospheric deposition in a climax spruce stand in the Poľana Mts. Vertical precipitation and throughfall was sampled on the research plot Predná Poľana in years 2004–2006. Precipitation was collected from two plots − a spruce forest stand and open area. The purpose of our work was to interpret the chemical-physical characteristics of vertical precipitation (pH, electric conductivity), to determine concentrations of selected chemical substances (SO_4^{2-}, NO_3^-, K^+, Na^+, Mg^{2+}, Ca^{2+}, NH_4^+) and to evaluate the wet atmospheric depositions of main elements (H^+, S, N and alkaline elements). We observed increasing precipitation acidity and significant pollution more enriching stand precipitation than the open area. The pH values in throughfall ranged from 3.43 to 6.09, in lysimetric waters from 3.29 to 5.86 and in open area from 4.9 to 6.42. The results show an increase in pH values, and decrease in all the followed elements and substances in both concentration and deposition, both on the open plot and in the stand.

Key words

vertical precipitation, throughfall, chemistry of precipitation, atmospheric deposition, acidification

Introduction

The human activities at the millennium change are influencing, intentionally or unintentionally, the entire living environment on the Earth. The vigorous development in areas of industry, agriculture, car and air transport and changes to landscape have entailed progressive disturbances to equilibrium of the natural environment. Very negative impact is owing to air pollution that does not take in consideration any borders. The attributes of airborne pollutants are: wide range of chemical transformations, dispersion, distribution and transboundary transport. Apart from the precipitation water, the forests and other ecosystems are also entered by many other in-the-air-present chemical elements and substances that cause and accelerate acidification of the natural environment (Fišák et al., 2006). Over the past years, atmospheric emissions in Central Europe decreased considerably (Matschullat et al., 2000), however, the impact of anthropogenic pollutants on forest ecosystems is still a hot topical problem. The Slovak Republic is situated in Central Europe, on the boundary of the territory with the heaviest regional air pollution on the continent. The level of regional pollution influences the load to most forest plots in Slovakia, and it requires, consequently, a special attention. The transboundary transport of pollutants represents, under the prevailing zonal airflow, about 60%. The harmful impact has not avoided our biggest volcanic mountain massive Poľana, too (Kunca, 2003). The natural environment of the PLA – BR has been, already since the 60-s, a spot for natural environment study, especially for workers from the University in Zvolen (SLÁVIKOVÁ, 1993). A special attention should be paid to the work MIHÁLIK and SLÁVIK (1988), focussing on polluted precipitation in the area of the Poľana massive, already in the 80-s of the last century. Since 1991, the load with airborne pollutants has been being studied by workers from the Forest Research Institute and from the Faculty of Forestry of the Technical University in Zvolen who established the comprehensive research-demonstration subject Poľana – Hukavský Grúň. In 2002, there was started monitoring of wet atmospheric deposition at the research locality Predná Poľana, situated in a mountain ridge spruce forest at 1347 m asl. The data obtained for this locality are subjected to closer examination in this work.

Methods

The experiment was realised on a research site Predná Poľana which is localised in the southwestern part of Biosphere Reserve Poľana. The main characteristics of the research plot are shown in the Table 1.

The material from the research plot was sampled in vegetation periods 2004–2006, once or two times a month. The sampling periods were the following: in year 2004 from 24.6. to 10.11., in year 2005 from 16.6. to 15.11., and in year 2006 from 30.5 to 30.11. The vertical precipitation and precipitation in the stand (throughfall) was collected into polyethylene collectors each with a catching area of 200 cm^2, made from material chemically inert to precipitation water. The lysimetric water was collected into plate lysimeters made from stainless steel, with catching area of 500 cm^2. The type of precipitation sampler and lysimeter was the same.

The precipitation was sampled from five collectors and four lysimeters. The open plot was provided with one collector situated in the young growth, one in the age-differentiated group, one in the stand gap, and one under an old spruce tree, the last one together with the lysimeter. The lysimeters were installed under the humus horizon, at a depth of 30 cm. In our work we use the following abbreviations for the sampling spots (Table 2).

In the text, the abbreviations are appended with symbols 04, 05, 06, expressing the sampling year. Chemical laboratory analysis was carried out in a common way, following the methods listed in Table 3, (MIHÁLIK et al., 1992).

From the data sets of results of chemical analysis we have calculated the values of the following statistic variables (ŠMELKO, 1998): arithmetic mean, weighed arithmetic mean, median, modus, standard deviation, variation coefficient, percentiles (10%, 25%, 75%, 90%). Apart from these variables, we also examined the maximum and minimum value of the sampling set and precipitation totals. To calculate the atmospheric deposition, the precipitation total over the vegetation period was converted to the annual total. Statistical significance of differences between arithmetic means obtained for individual plots was tested using variance analysis, Duncan test.

Table 1. Characteristic of the research plot Predná Poľana

Altitude [m]	Exposition	Tree composition [%]	Mean annual temperature	Mean precipitation total	Soil type
1347	SW	spruce 93 beech 4 mountain ash 3	3.5–4.0 °C	900–1000 mm	andosols cambisols

Table 2. Abbreviations of the individual sampling spots

Sampling spot	Abbreviation
Open plot	PV
Precipitation gauge in the young growth	PZ1
Precipitation gauge in the age-differentiated group of trees	PZ2
Precipitation gauge in the stand gap	PZ3
Precipitation gauge under the old spruce tree	PZ4
Lysimeter in the young growth	PL1
Lysimeter in the age-differentiated tree group	PL2
Lysimeter in the stand gap	PL3
Lysimeter under the old spruce tree	PL4

Table 3. Methods used in chemical analysis for determining concentrations of individual components in the samples

Parameter	Method	Unit
pH	Potentiometer. with a high-resistance electrode	
Conductivity	Conductometry	µS cm^{-1}
SO_4^{2-}	Nitration with lead nitrate on indicator ditizon in acetone environment	mg l^{-1}
NO_3^-	Calorimetry. with sodium salycil. in sulphuric acid environment	mg l^{-1}
K^+	Atomic absorption spectrophotometry	mg l^{-1}
Na^+	Atomic absorption spectrophotometry	mg l^{-1}
Mg^{2+}	Atomic absorption spectrophotometry	mg l^{-1}
Ca^{2+}	Atomic absorption spectrophotometry	mg l^{-1}
NH_4^+	Colometry. using Nessler agent	mg l^{-1}

Results and discussion

Values of pH, electric conductivity

Graphical interpretation of the statistic characteristics of pH values is in Fig 1. The range of weighted means is from 3.45 (PL1 04) to 5.57 (PV 05). The highest acidity was found for throughfall (4.27) and lysimetric water (3.86) in the young growth (PZ1). The lowest precipitation acidity was on the open plot (5.57). Progressing over the study period 2004–2006, there were observed increasing pH values, then followed a drop in 2006 almost on the all plots. Lysimetric waters were more acid than the corresponding throughfall. Comparison between pH values of precipitation on open plot and of throughfall, using the variance analysis and Duncan test, revealed statistically very significant differences (significance level $\alpha < 0.01$) between PV and all the other plots, with exception of PZ4, where the difference was only significant (significance level $\alpha < 0.05$).

Electric conductivity (EC) of a solution is a measure of the total amount of ions in the solution. On the research plot Predná Poľana, the highest values were observed for the lysimeters, followed by the throughfall and open plot. The weighed means of EC for the period 2004–2006 were: on the open plot (PV) 18.48 µS cm^{-1}, in the throughfall 16.46–37.79 µS cm^{-1}, in lysimetric waters 41.7–68.44 µS cm^{-1}.

Concentration of sulphates, nitrates, nitrogen – ammonium cation and basic cations

The lowest mean SO_4^{2-}, NO_3^- and NH_4^+ concentrations were measured on open plot. Soil solution was more concentrated than the corresponding throughfall. The young growth (PZ1) characterizes the highest concentration of observed chemical substances. For the entire study period 2004–2005 we can see decrease in concentrations.

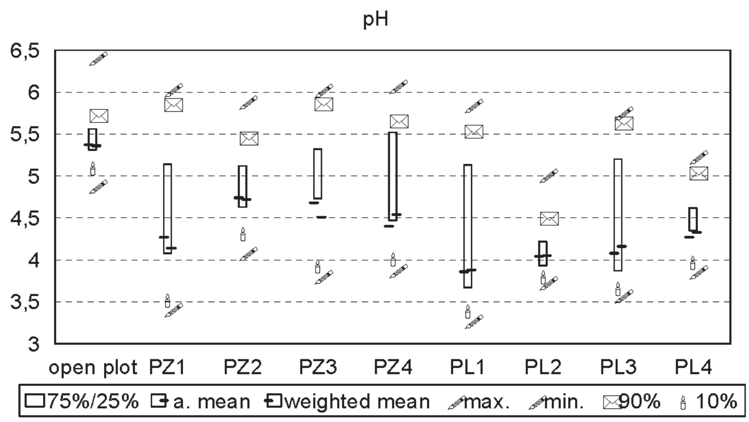

Fig 1. Statistical interpretation of pH values in the period 2004–2006

The mean values in the throughfall ranged from 4.41(PZ2) to 8.32 mg l^{-1} (PZ1), in lysimetric water from 6.15 (PL2) to 13.43 mg l^{-1} (PL1). The lowest mean SO_4^{2-} concentration 4.68 mg l^{-1} was found on open plot. The absolute maximum, 75.2 mg l^{-1}, was measured on plot PL4 in 2006, the minimum 0.8 mg l^{-1} on plot PZ3 in 2004. In Fig 2 is evident that the lysimeters at the individual sampling sites copy the course of throughfall, their values, however, are shifted upwards. The results of Duncan test confirmed significant differences in arithmetic means only in case of open plot and the lysimeter situated in the young growth (PL1).

Graphical evaluation of NO_3^- values is in Fig 3, NH_4^+ concentrations are in Fig 4. Absolute measured maximum, 63.25 mg l^{-1}, was recorded on plot PL4 06, minimum 0.2 mg l^{-1} on plot PZ2 05. The mean NO_3^- concentration on open plot was 2.52 mg l^{-1}. The mean value in the throughfall ranged between 1.55 (PZ3)–4.57 (PZ4) mg l^{-1}, in the lysimetric water from 8.05 (PL3) to 13.81 mg l^{-1} (PL1), what is 3.7–6-times more than in the throughfall. The trend of concentration values in individual years is very unbalanced, we can however agree on a decrease up to 2006 on most plots. The most conspicuous was this decrease in lysimetric water.

The highest concentrations of the second form of nitrogen – ammonium cation NH_4 were found in lysimeters ranging from 2.42 to 3.77 mg l^{-1} with the maximum on plot PL2. The weighed means of the throughfall moved from 1.2 to 2.38 mg l^{-1}, with the biggest weighed mean reached on plot PZ1. We observed the lowest mean NH_4^+ concentration 1.08 mg l^{-1} on the open plot. The same hierarchy: open plot – stand – lysimeters can also be observed in the separate years, the trends, however, are specific for different sampling sites.

The results of variance analysis point out, however, a difference, statistically very significant (significance level $\alpha < 0.01$), in NO_3^- concentrations between

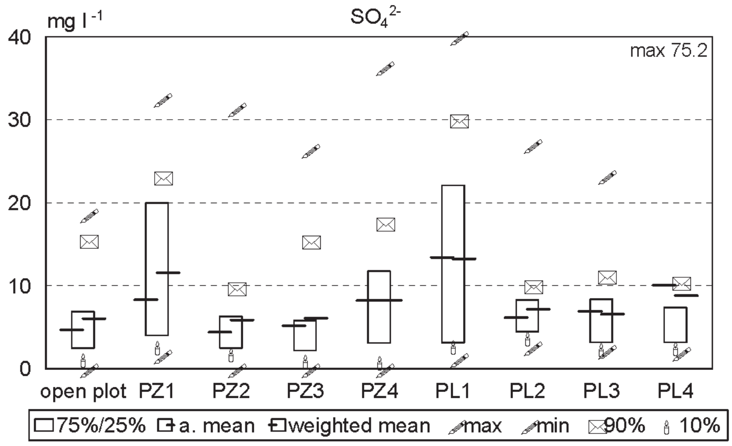

Fig 2. Graphical interpretation of SO_4^{2-} concentrations in 2004–2006

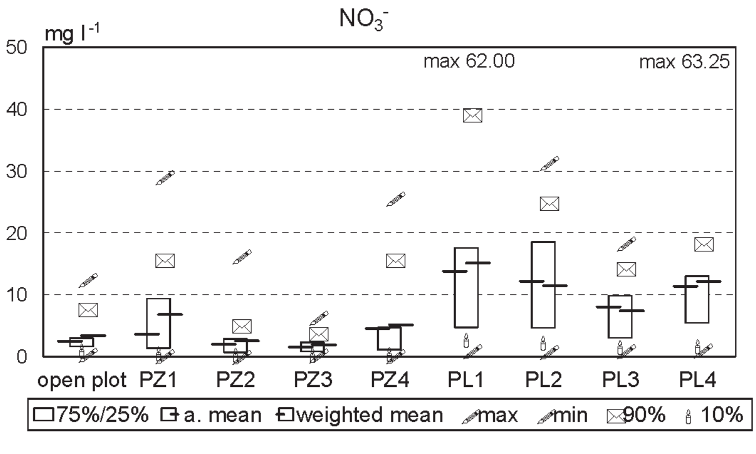

Fig 3. Graphical interpretation of statistic values of NO_3^- in 2004–2006

open plot and plots PL2 , PL3; in case of NH_4^+, there were significant differences between plots PV and PZ1, PZ2, PZ3.

Comparing our results with the results other authors (Table 4), we can see they are comparable. It is evident decrease in pH values and reduction in SO_4^{2-}, NO_3^-, NH_4^+ concentrations in the recent years.

For all the studied alkaline elements is in general true that their concentrations increased both in the lysimetric water and in throughfall, unlike the precipitation on the open plot. The alkaline elements content was most enhanced in the soil solutions. Examining the values of these cations on the research plot Predná Poľana, we can see that the most abundant elements on the open plot were Ca^{2+} (0.77 mg l^{-1}) and Na^+ (0.51 mg l^{-1}), in the throughfall it was K^+ and Ca^{2+}. The conspicuous increase in K^+ and Ca^{2+} concentrations in the throughfall suggests that the origin of these elements is in the biomass leaching K^+ and Ca^{2+} cations intensively after having had contact with acid precipitation. The increase in concentrations of K^+ and Ca^{2+} is evident, beginning with the open plot towards the plots with developing canopy. An increase in K^+ and Ca^{2+} concentrations in throughfall opposite to open area has also been documented by Minďáš and Kunca (1997), who observed calcium concentrations 1.1–1.9-times higher after having passed spruce crows, in case of magnesium the original value increased by 6.5–10.9-times. Pavlenda (2007) reports for Poľana in year 2005 values of calcium on open plot 0.85 mg l^{-1}, in the stand 1.03 mg l^{-1}, concentration potassium in the stand 1.93 mg l^{-1}, on open plot 0.2 mg l^{-1}.

The mean magnesium concentrations were lower than concentrations of other alkaline elements. The lowest increase was observed on the open plot 0.51 mg l^{-1}. Also in this case is well observable increase in mean concentration with increasing stand canopy.

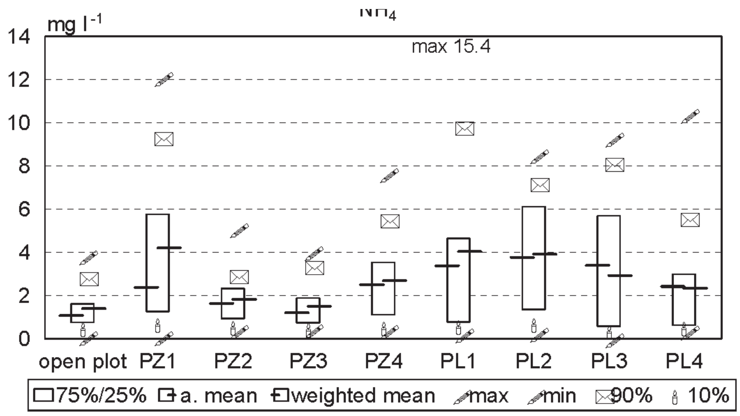

Fig 4. Graphical interpretation of statistic values NH_4^+ in 2004–2006

Table 4. Comparison of selected characteristics according to various authors

Locality		Altitude [m]	pH	SO_4^{2-} [mg l^{-1}]	NO_3^- [mg l^{-1}]	NH_4^+ [mg l^{-1}]	Authors Measured period
Predná Poľana	op	1347	5.37	4.68	2.52	1.08	This work (2007)
	ss		4.27–4.73	4.34–8.32	1.55–4.57	1.20–2.50	2004–2006
Poľana – Hukavský Grúň	op	850	4.58	1.92	2.26	0.44	Pavlenda (2007)
	ss		4.67	3.05	3.89	0.92	2005
Zadná Poľana	op	1430	4.88–5.66	5.33–17.31	1.56–3.59	1.10–3.29	Bublinec and Dubová (2000)
	ss		3.75–5.24	5.24–31.18	1.56–4.59	1.74–4.53	1991–1997
Poľana – Hukavský Grúň	op	850	4.64	3.90	2.01	1.32	Minďáš and Kunca (1997)
	ss		–	13.81	4.99	3.32	1996
Predná Poľana	op	1320	–	7.36	2.07	1.36	Mihálik and Slávik (1988)
	ss		–	53.29	8.31	7.13	1986

op – open plot, ss – spruce forest

The mean sodium concentrations in the throughfall ranged from 0.64 (PZ3) to 0.95 mg l^{-1} (PZ4), in lysimetric water from 0.73 (PL2) to 1.0 mg l^{-1} (PL1), the weighed mean on the open plot was 0.51 mg l^{-1}.

Duncan test confirmed a difference, statistically high significant, in Ca^{2+} and Mg^{2+} concentrations for plots PZ1, PL1, PL4 compared with the open one. The difference PV – PL2 in concentrations of both elements was significant at a level $\alpha < 0.05$. In case of potassium, the difference between PV and the other plots was found very significant, with plot PZ3 significant. Very significant differences were identified in concentrations Na^{+} between the open plot and plots PZ1, PZ4, PL1 and PL4.

Deposition of sulphur, total nitrogen, H^{+} and basic cations

On the research plot were found the considerable values sulphur, total nitrogen and the hydrogen ion deposition. The lowest deposition input was recorded on the open plot. The deposition in lysimeters in all the followed elements was lower than in the stand, in spite of the fact that the concentrations were showing an opposite trend.

Graphical representation of sulphur deposition at the individual sampling sites is in Fig 5. For the entire study period 2004–2006 is possible to observe increase in sulphur deposition. In the given time sulphur deposition ranged between 6.66–33.12 kg ha^{-1} $year^{-1}$ (except for maximum), in the stand, from 2.04 to 17.11 kg ha^{-1} $year^{-1}$ in the lysimeters and reached 12.03–23.11 kg ha^{-1} $year^{-1}$ on the open plot. Conspicuous increase was on sampling spot under the old spruce (PZ4), on which deposition reached its maximum (51.74 kg ha^{-1} $year^{-1}$). This is the result of the highest precipitation total and, at the same time, concentration on plot PZ4 compared to the preceding years.

The final values of total nitrogen deposition in 2004–2006 are illustrated in Fig 6. The trend of total nitrogen deposition is very unbalanced for different sampling sites. Both in open plot and in the stand it doesn't cross value 30 kg ha^{-1} $year^{-1}$, except for plot PZ4, when the deposition input in year 2006 reached 60.55 kg ha^{-1} $year^{-1}$. Similar to other elements, the nitrogen depositions in lysimeters were lower than in the forest stand.

We compare our measured values of sulphur and nitrogen deposition with results other authors, which deal with this theme over the Poľana orographic area. Table 5 refers about individual differences found on open plot and in spruce stand.

The mean annual values of H^{+} deposition in 2004 ranged from 0.07 to 1.23 kg ha^{-1} $year^{-1}$. In 2005 was observed a notable, more than 50% reduction in H^{+} deposition on all the plots except for PZ4 and PL4, where the deposition input was higher. In 2006 there followed a moderate increase again, from 0.02 to 0.12 kg ha^{-1} $year^{-1}$ on most plots. Pavlenda (2007) reports for open plot in Poľana H^{+} deposition 0.08 kg ha^{-1} $year^{-1}$ in 2004, 0.02 kg ha^{-1} $year^{-1}$ in 2005. Our results are higher, but the deposition in 2005 is comparable (0.03 kg ha^{-1} $year^{-1}$).

The input of base cations in throughfall deposition results from two different sources, total deposition (wet + dry + interception), and internal leaching. The latter is the dominating mechanism for K^{+}. The leaching efficiency depends on ambient SO^{2-} concentrations (Slovik et al., 1996) and on precipitation, because K^{+} is highly mobile and present as an electrolyte within the plant. The SO^{2-} decrease consequently led to a reduction of K^{+}

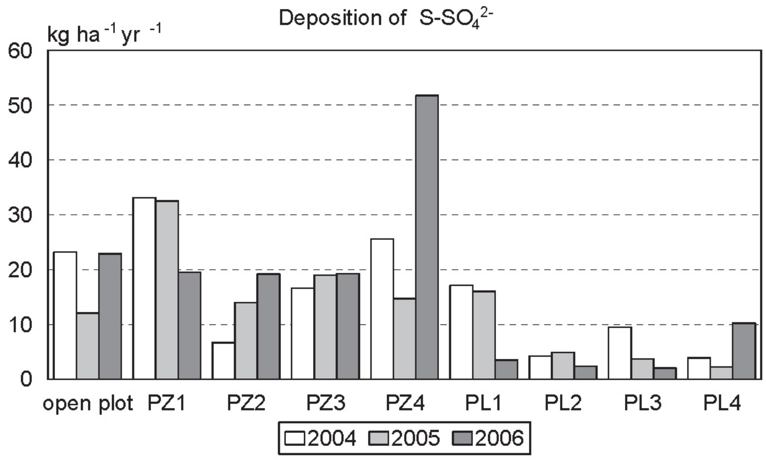

Fig 5. Graphical interpretation of elementary sulphur deposition

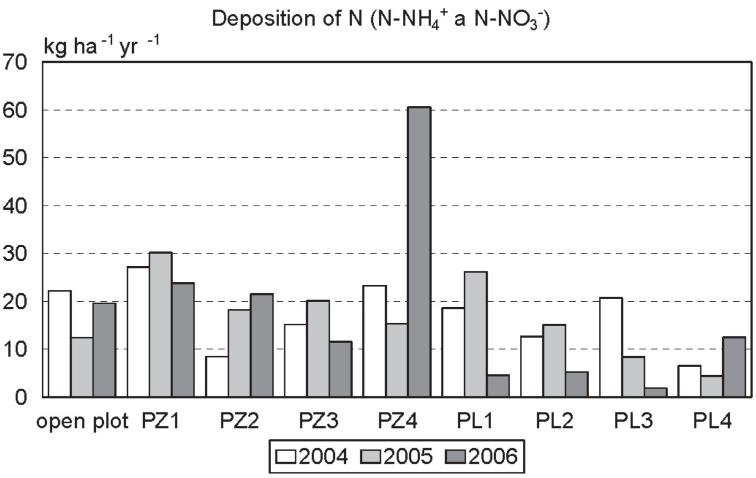

Fig 6. Graphical interpretation of total nitrogen deposition

Table 5. Comparison of S and N deposition according to various authors in area Poľana

Locality		Altitude [m]	S	N	Authors Measured period
Predná Poľana	op ss	1347	19.38 13.27–30.66	18.07 15.61–33.04	This work (2007) 2004–2006
Poľana – Hukavský Grúň	op ss	850	5.70 7.52	7.46 10.99	Pavlenda et al. (2007) 2005
Zadná Poľana	op ss	1430	39.00 78.00	15.20 33.40	Bublinec and Dubová (2000) 1996–1997
Poľana – Hukavský Grúň	op ss	850	15.00 26.80	14.30 23.10	Minďáš (1999) (1993–1997)
Predná Poľana	op ss	1320	20.20 99.00	11.70 40.70	Mihálik and Slávik (19918) (1986–1987)

op – open plot, ss – spruce forest

input. Observed variations were simply related to precipitation variability. Different from K^+, both Ca^{2+} and Mg^{2+} are bound to plant tissue and are being exchanged via ion exchange processes against protons. Lower pH-values thus lead to higher leaching rates. Thus, the reduction of Ca^{2+} and Mg^{2+} deposition is not only related to a reduction in dust emissions but to the increase of pH-values in precipitation (Zimmerman et al., 2003).

Graphical interpretation of the values of wet deposition of basic cations in the period 2004–2006 is in Fig 7 (a–c), highest deposition inputs over the study period were observed, in accord with the concentration values, also for potassium and calcium. Their depositions are highest in forest stand. The deposition input of calcium on open plot represented 7.25–13.51 kg ha^{-1} year^{-1}, in forest stand it ranged from 4.19 (PZ2) to 42.63 kg ha^{-1} year^{-1} (PZ1), in lysimeters from 1.55 (PL2) to 8.27 kg ha^{-1} year^{-1} (PL4).

Considerably conspicuous differences in potassium deposition could be observed between the open plot and forest stand. The deposition on open plot was moderately increasing, starting with a value of 1.48 in year 2004 up to 3.84 kg ha^{-1} year^{-1} in year 2006. The potassium input on open plot was the lowest among all the studied basic cations. The trend of potassium deposition in lysimeters was decreasing, except for plot PL4. Blihárová and Škvarenina (1999) for locality Predná Poľana in year 1999 give deposition of calcium 15.1 ha^{-1} year^{-1} in stand, for K^+ 15.1 ha^{-1} year^{-1}.

Deposition of magnesium is most conspicuous below the stand. On the open plot, the values in study period ranged from 1.43–2.89 kg ha^{-1} year^{-1}, the values in lysimeters were showing a slightly decreasing trend (except for PL4 – a steep increase in 2006). The values of sodium deposition on open plot and in forest stand are more equalized than in case of the other alkaline elements. The deposition input of sodium on the open plot was the highest one among all the basic cations. For all the studied elements, lysimeters were showing lower deposition values compared to forest stand.

MINĎÁŠ and TÓTHOVÁ (2004) suggest that assessment and interpretation of basic elements deposition should be carried out with a bit of care, especially in the stand, because the values measured in stand precipitation

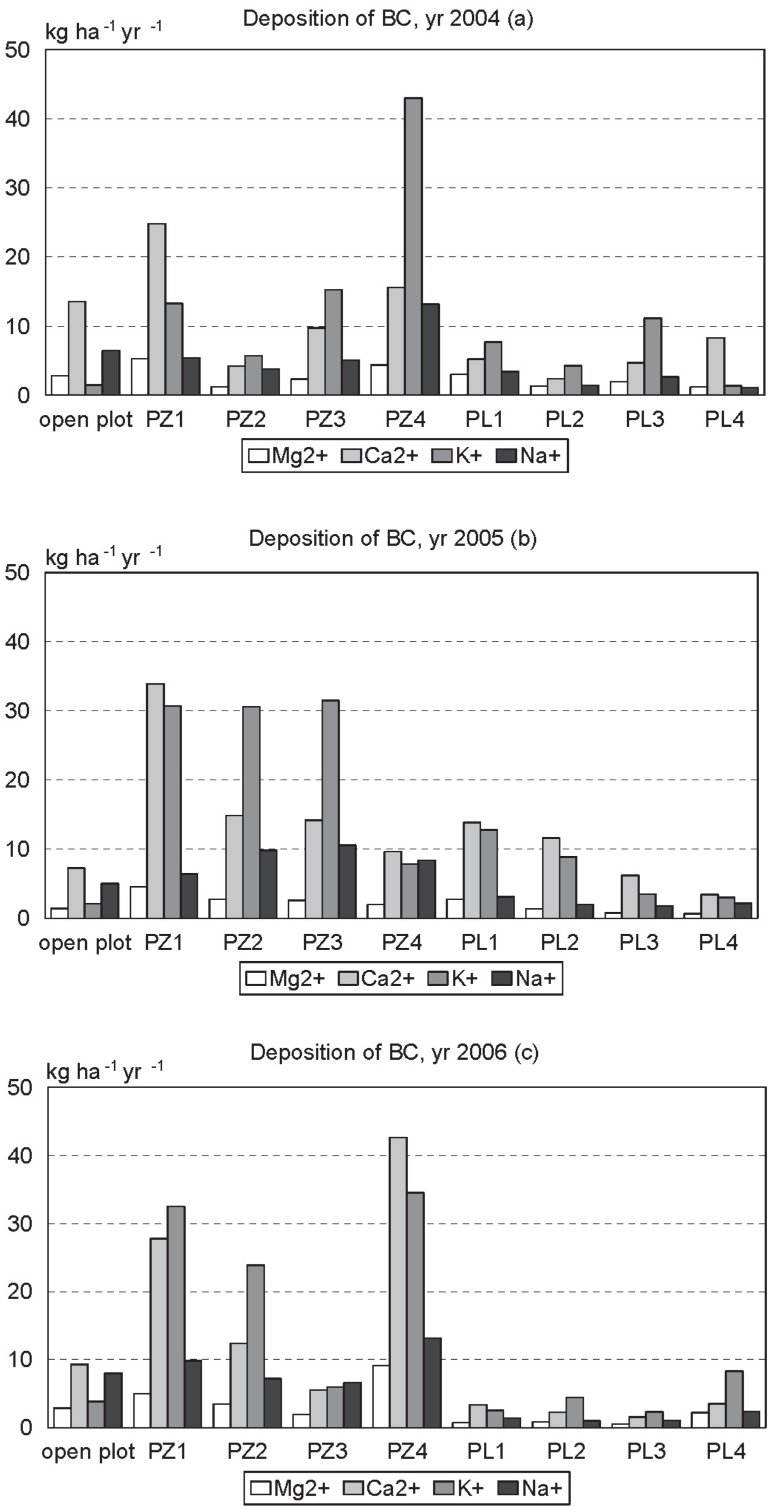

Fig 7. Deposition of basic cations (BC) in 2004(a), 2005(b) and 2006(c)

are influenced by interaction with the stand biomass and they need not reflect the true deposition of basic ions into the stand.

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Chemizmus podkorunových zrážok a atmosférická depozícia v klimaxovom subalpínskom smrekovom lese Biosférickej rezervácie Poľana, Slovensko

Súhrn

Predložená práca je zameraná na zisťovanie fyzikálno-chemických vlastností zrážkových vôd (pH, elektrická vodivosť), na analýzy koncentrácií vybraných chemických komponentov (H^+, SO_4^{2-}, NO_3^-, K^+, Na^+, Mg^{2+}, Ca^{2+}, NH_4^+) a na výpočet imisných atmosférických depozícií hlavných elementov (H, S, N a bázických prvkov). Objektom práce bola horská smrečina 7. smrekového vegetačného stupňa, skupiny lesných typov Sorbeto-Piceetum, Acereto-Piceetum na lokalite Predná Poľana. Z práce možno vyvodiť nasledovné závery:

V priebehu sledovaného obdobia 2004–2006 možno pozorovať rast pH hodnôt a ich mierny pokles v roku 2006, takmer na všetkých plochách. Lyzimetrické vody v porovnaní s podkorunovými zrážkami sú kyslejšie. Najnižšia kyslosť bola preukázaná na voľnej ploche (5,57).

Koncentrácie H^+, SO_4^{2-}, NO_3^- a NH_4^+ sú najnižšie na voľnej ploche. Lyzimetrické vody sú viac koncentrované ako podkorunové zrážky. V rámci podkorunových zrážok sa najväčšími koncentráciami vyznačuje mladina. Za sledované obdobie 2004–2006 možno pozorovať pokles koncentrácií sledovaných komponentov.

Na sledovaných plochách sme zaregistrovali značné obohatenie koncentrácií bázických katiónov v porastových zrážkach (najväčšie obohatenie vykazuje pôdny roztok) v porovnaní s voľnou plochou. Táto skutočnosť indikuje nebezpečný jav vylúhovania bázických živín z organického materiálu. Pri sledovaní hodnôt bázických katiónov na výskumnej ploche Predná Poľana možno vidieť, že na voľnej ploche má najväčšie zastúpenie Ca^{2+} (0,77 mg l^{-1}) a Na^+ (0,51 mg l^{-1}), v podkorunových zrážkach sú to K^+ a Ca^{2+}.

Na výskumnej ploche Predná Poľana boli zistené značné hodnoty depozície síry, celkového dusíka a vodíka. Najnižší depozičný vstup vykazuje voľná plocha. Depozícia v lyzimetroch všetkých zisťovaných prvkoch je nižšia ako v poraste, hoci koncentrácie mali opačnú tendenciu.

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