

Sulphur and nitrogen concentrations and fluxes in bulk precipitation and throughfall in the mountain and highland spruce stands in the Czech Republic

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ABSTRACT: Concentrations and fluxes of sulphur and nitrogen compounds in bulk precipitation and in throughfall were evaluated and compared for two experimental sites in the Czech Republic: one situated at Rájec (Drahanská upland, 610 m a.s.l.) and the second one at Bílý Kříž (Moravian-Silesian Beskids, 908 m a.s.l.) both with similar stands of young Norway spruce. The three-year study performed during 2006–2008, revealed statistically significant differences in nitrate nitrogen concentrations in bulk precipitations and in ammonium nitrogen concentrations both in bulk precipitation and in throughfall between the two sites. Higher nitrogen compounds concentrations in bulk precipitation were found at Rájec. Differences between the two sites in sulphur concentrations were not found out neither in bulk precipitation nor in throughfall waters. Total sulphur deposition amounted to 8.1, 8.3 and 6.7 kg S·ha⁻¹ at Rájec and to 14.8, 16.9 and 15.4 kg S·ha⁻¹ at Beskids for the three years studied, respectively. Total inorganic nitrogen throughfall flux amounted to 12.1, 11.6 and 11.6 at Rájec and 13.8, 18.9 and 15.0 kg·ha⁻¹ at Bílý Kříž for the years 2006, 2007 and 2008, respectively.

Keywords: Bílý Kříž (Moravian-Silesian Beskids Mts.); Czech Republic; nitrogen deposition; rainfall; Rájec (Drahanská Highland); sulphur deposition; throughfall

From the second half of the 20th century large-scale forest decay has been observed in many places of central Europe. Acid atmospheric precipitation destabilized the ecological site valence mainly of allochthonous spruce monocultures. The acidification shows a number of negative consequences for forest ecosystems – qualitative and quantitative changes in water (SCHULZE et al. 1989; JOHNSON et al. 1991; KREUTZER 1994) and in soils (BROWN 1985; POSTEL 1986; EVERS, HÜTTL 1990; FRANZ-LUEBBERS et al. 1995; UCHMANSKI et al. 1995) and decline of forest stands (ANDERSSON, PERSSON 1988; GORHAM 1989; ULRICH 1991; BUSSOTTI, FERRETTI 1998; PERCY, FERRETTI 2004).

Sulphur and nitrogen deposition in forest ecosystems have been intensively studied mainly due to its influence on nutrient imbalances and leaching of nitrogen compounds from soils and because sul-

phates and nitrates are the dominant anions in bulk deposition (NILSSON, WIKLUND 1995; CALANNI et al. 1999; LADANAI et al. 2007). Nitrogen is an important component of vital organic compounds, amino-acids, proteins and nucleic acids, and its deficiency limits plant growth. Plants are able to utilize nitrogen in the form of ammonium and nitrate ions. Sulphur is an essential macronutrient required for plant growth. It is, like nitrogen, a component of amino acids, proteins and enzymes. The natural balance of nitrogen and sulphur cycling in forest ecosystems has been disturbed by increased inputs of those elements from atmosphere polluted by industrial and other anthropogenic activities (agriculture, transportation). Forest soils receive these elements also through wet and dry deposition. Wet deposition is the flux of dissolved components from the atmosphere with rain, snow and hail, dry deposition

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is flux of gases and particles from the atmosphere during dry periods, due to gravity (sedimentation), impaction, diffusion and other processes. Dry deposition fluxes are strongly influenced by the type of surfaces (leaves, needles, rocks, and water), humidity of surfaces, macro- and micrometeorology (stomata closure) (UN-ECE 2006).

Sulphur and nitrogen are markedly captured by the canopy and coniferous forests are particularly important in this respect because they have a high aerodynamic roughness and a dense canopy surface over the whole year resulting in high rates of precipitation interception and the turbulent exchange of aerosols, particles and gases from the atmosphere (AUGUSTO et al. 2002; MATSON et al. 2002; CRISTIENSEN et al. 2006; GUNDERSON et al. 2006). In forest ecosystems dry deposition may contribute to the input of these elements to the soil considerably because the precipitation is being enriched with dry deposition (DRAAIJERS et al. 1997; SOLBERG, TØRSETH 1997; DISE et al. 1998; WHELAN et al. 1998; NEAL 2002; BÉLANGER et al. 2004; CHIWA et al. 2004; SOLBERG et al. 2004; BARBIER et al. 2008; BERGER et al. 2008; SHACHNOVICH et al. 2008). Throughfall waters collected under canopy thus contain larger amounts of ions than rainfall water collected in the nearby open areas (YOSHIDA, ICHIKUNI 1989; SAXENA, LIN 1990; BEIER, HANSEN 1993; BALESTRINI, TAGLIAFFERI 2001).

Although Europe has been successful in reducing the emissions of nitrogen (ERISMAN et al. 2003) and sulphur compounds (KVAALLEN et al. 2002; UKONMAANAHO, STARR 2002) over the past two decades the problem of forest soil acidification still persists, especially at localities with high emission load. One of the most polluted areas in the Czech Republic is situated at the borders with Poland and Slovakia is known as "Black Triangle II" (MARKERT et al. 1996). The Dražanská Upland, on the other hand, belongs among the cleaner areas (BOHÁČOVÁ et al. 2009).

Various methods can be used to measure or estimate deposition to forests. One group of methods is based on micrometeorology (SEINFELD, PANDIS 1998; WESELY, HICKS 2000) the other group is based on forest stand throughfall and open plot precipitation sampling (DE VRIES et al. 2001; STAELENS et al. 2008). The second approach is used for the deposition monitoring within the framework of International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (UN-ECE 2006) and is used also in this study. The main drawback of the throughfall method is the interaction between the canopy and the throughfall water for nitrogen, protons and base cations. How-

ever, throughfall deposition can give information on the lower limit of the true deposition of nitrogen and the upper limit of true deposition of base cations other than sodium. For sodium and sulphur the canopy uptake and leaching is considered to be negligible and consequently the throughfall flux is used to estimate the total deposition (UN-ECE 2006).

The aims of this study were:

- to compare concentrations of sulphur and nitrogen compounds in bulk precipitation and throughfall samples at two experimental sites with young spruce stands in the Czech Republic, one situated in Moravian-Silesian Beskids at an altitude of 908 m and the second one in Dražanská Upland at an altitude of 632 m;
- to assess the total sulphur deposition within young spruce stands for these two localities with various levels of atmospheric pollution and compare it with the sulphur deposition fluxes at other sites in the Czech Republic.

MATERIAL AND METHODS

Description of the study sites and characteristics of the two spruce stands studied is given in Table 1.

Bulk precipitation and throughfall sampling were carried out at both sites during 2006–2008. For sampling throughfall in the spruce stand and atmospheric precipitation on the open area permanently open polyethylene sampling vessels of an area of 335.33 cm² were used (BLOCK, BARTELS 1985; NIEHUS, BRUGGEMANN 1995). The vessels were inserted into the thick-walled plastic pipes in order to shield the samples from solar radiation and to hold the funnels approximately 1 m above the ground. There were 7 collectors randomly distributed at the mountain spruce stand and 5 at the highland spruce stand. During the winter the number of collectors at the mountain spruce stand was reduced to 5. Bulk atmospheric precipitation was sampled at both sites with one collector installed in the nearby open area. Samples were taken once a month in the winter season and in 14-day intervals in other seasons. The samples were transferred to the laboratory and prepared for the analyses usually the next day after sampling. One proportional sample was prepared from water collected in all samplers at a particular plot. In winter it was sometimes necessary to wait one day because the samples were frozen. Determination of ammonia was done immediately in freshly prepared samples; the aliquots for sulphate and nitrate determination

Table 1. Description of study sites and spruce stands

		Mountain spruce stand	Highland spruce stand
Study site	locality	Bílý Kříž – Moravian-Silesian Beskids Mts.	Rájec – Dražanská Highland
	geographic coordinates	49°30'N, 18°32'E	49°29'N, 16°43'E
	altitude	908 m a.s.l.	610 m a.s.l.
	geological subsoil	flysh layer with dominant sandstone	acid granodiorite
	soil characteristics (soil type)	Typical humo-ferric Podzol (PZhz') ¹ Podzols (PZ) ² with mor-moder form of surface humus	Modal oligotrophic Cambisol (KAmd') ¹ Cambisols (CM) ² with moder form of surface humus ³
Climatic characteristics	summer days	10 to 30 ⁴	30 to 40 ⁴
	days with mean temperature 10°C and more	120 to 140 ⁴	140 to 160 ⁴
	days with frost	140 to 160 ⁴	110 to 130 ⁴
	ice days	50 to 60 ⁴	40 to 50 ⁴
	mean temperature in January	–4 to –5°C ⁴	–2 to –3°C ⁴
	mean temperature in April	2 to 4°C ⁴	6 to 7°C ⁴
	mean temperature in July	14 to 15°C ⁴	16 to 17°C ⁴
	mean temperature in October	5 to 6°C ⁴	7 to 8°C ⁴
	mean number of days with precipitation equal 1 mm and more	120 to 130 ⁴	100 to 120 ⁴
	sum of precipitation during the vegetation period	600 to 700 mm ⁴	400 to 450 mm ⁴
	sum of precipitation during the winter period	400 to 500 mm ⁴	250 to 300 mm ⁴
	days with snow cover	120 to 140 ⁴	60–100 ⁴
	cloudy days	150 to 160 ⁴	120 to 150 ⁴
	cloudless days	40 to 50 ⁴	40 to 50 ⁴
	mean annual precipitation total	1,115.3 (1961–1990) ⁷	643.4 (1957–2000) ⁷
	mean annual temperature	4.91 (1961–1990) ⁷	6.95 (1961–2000) ⁷
Spruce stand	tree species	<i>Picea abies</i> [L.] Karst.	<i>Picea abies</i> [L.] Karst.
	stand age (years)	2008	34
	mean stand height (m)	2006	11.6
		2007	12.2
		2008	12.5
	stand density (trees·ha ^{–1})	2006	1,456
		2007	1,456
		2008	1,428
	forest typology	5S1– <i>Abieto-Fagetum</i> mesotrophicum ⁷	5S1– <i>Abieto-Fagetum</i> mesotrophicum with <i>Oxalis acetosella</i> ⁷

¹NĚMEČEK et al. (2001); ²IUSS Working Group WRB (2006); ³MENŠÍK et al. (2009); ⁴QUITT (1971); ⁵HADAŠ (personal communication); ⁶HADAŠ (2002); ⁷PLÍVA (1987); NA – not available

were frozen and kept in freezer until analyzed (usually within a month after the sample preparation). The methods used for determination of SO₄^{2–}, NO₃[–] and NH₄⁺ are listed in Table 2.

Mean amount of throughfall water was calculated for the individual sampling term as the arithmetic average of the water captured in the parallel collectors at the given plot. Mean annual concentra-

Table 2. List of methods and instruments used for the analysis of rainfall and throughfall waters

Analyte	Analytical method	Instrumentation
NH ₄	spectrophotometric at the wave-length of 650 nm or flow injection analysis with diffusion chamber	UV/VIS spectrophotometer or flow injection analyzer FIALab 2500
NO ₃ , SO ₄	high performance ion exchange liquid chromatography with gradient elution	chromatograph DX-600 DIONEX with gradient pump GP50

tions for individual ions and plots were calculated as weighted means (weights were precipitation amounts) of ions concentrations measured during the year at the particular plot. The fluxes of SO₄²⁻, NO₃⁻ and NH₄⁺ (mg·m⁻²) in bulk precipitation and in throughfall for each sampling event were calculated as a product of the amount of water (l·m⁻²) and the relevant element concentration (mg·l⁻¹) and summed for every single year and plot to give annual flux for the particular plot.

Statistical software STATISTICA 9.0 (StatSoft 2008) was used for results evaluation.

RESULTS

Water fluxes

Bulk precipitation totals on the open area and amounts of throughfall within spruce stands at both study sites in 2006–2008 are shown in Fig. 1. Annual precipitation totals were 1,171 mm, 1,282 mm, 1,033 mm and 661, 551, 458 mm for the three years, 2006–2008, at Bílý Kříž and Rájec, respectively. The driest year was 2008 when precipitation total at Bílý Kříž was only 1,033 mm and at Rájec 458 mm. The values measured at Bílý Kříž were above the long term average (which was 1,115 mm for the period of 1961–1990 according to Hadaš – personal communication) in 2006 and 2007 and lower than the long term average in 2008. In Rájec the precipitation total in the first year studied was above the long term average (i.e. 643 mm for the period of 1957–2000, HADAŠ 2002), the next two years were under the long term average from the point of view of precipitation

totals. At Bílý Kříž the amounts of throughfall were higher by 1–4% than precipitation totals. Negative interception was recorded also in the spruce stand at Rájec in 2006 and 2007 (increase of throughfall amounts by 3% and 13%, respectively), whereas in 2008 the throughfall amount was by 3% lower than the bulk precipitation total on the open area.

Ion concentrations in bulk precipitation and throughfall

Mean annual concentrations of sulphate, nitrate and ammonium ions are summarized in Table 3. Mean annual sulphate concentrations in bulk open area (BOA) precipitation were 1.81–2.01 mg·l⁻¹ and 1.68–2.30 mg·l⁻¹, for throughfall (THR) waters the values were 3.77–4.30 mg·l⁻¹ and 4.53–4.50 mg·l⁻¹, at Bílý Kříž and Rájec, respectively, and as can be seen, the values for throughfall precipitation were not substantially different for both sites in individual years. Mean annual nitrate concentrations in BOA precipitation were 1.59–1.72 mg·l⁻¹, and 2.08–3.49 mg·l⁻¹ at Bílý Kříž and Rájec, respectively. In throughfall the values were within 2.66–3.29 mg·l⁻¹ at Bílý Kříž and within 3.43–5.45 mg·l⁻¹ at Rájec (Table 3). Mean annual ammonium concentrations in BOA precipitation were 0.63–0.71 mg·l⁻¹ and 0.90–1.63 mg·l⁻¹, for throughfall the values were 0.73–0.93 mg·l⁻¹ and 1.23–1.84 mg·l⁻¹, at Bílý Kříž and Rájec, respectively. A substantial difference between the two sites in concentrations of nitrogen compounds both in bulk precipitation and throughfall is apparent with higher concentrations found always for waters from Rájec.

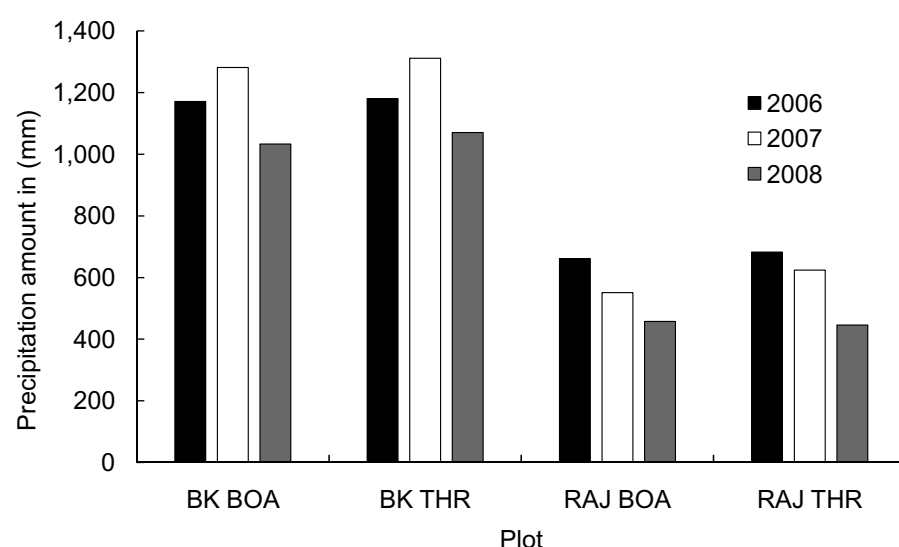


Fig. 1. Bulk open area precipitation (BOA) totals and throughfall (THR) amounts within spruce stands at Bílý Kříž (BK) and Rájec (RAJ) experimental sites in 2006–2008

Table 3. Mean annual concentrations of sulphate, nitrate and ammonium ions in bulk open area (BOA) precipitation and in throughfall (THR) within the spruce stands at the study sites in Bílý Kříž and Rájec

	Sampler/ plot	SO ₄ ²⁻ (mg·l ⁻¹)			NO ₃ ⁻ (mg·l ⁻¹)			NH ₄ ⁺ (mg·l ⁻¹)		
		2006	2007	2008	2006	2007	2008	2006	2007	2008
Bílý Kříž	BOA	2.01	1.97	1.81	1.72	1.59	1.61	0.63	0.71	0.65
	THR	3.77	3.87	4.30	2.66	3.20	3.29	0.73	0.93	0.85
Rájec	BOA	1.68	2.30	2.14	2.08	3.49	3.09	1.31	0.90	1.64
	THR	3.53	3.97	4.50	3.43	3.99	5.45	1.28	1.23	1.84
The relative increase of annual element fluxes in throughfall when compared with bulk precipitation in (%)										
Bílý Kříž		88	97	138	55	101	105	16	31	31
Rájec		110	73	110	65	14	76	-2	36	12

A 2-way ANOVA was performed to test the significance of plot (Bílý Kříž open area, Bílý Kříž throughfall, Rájec open area and Rájec throughfall) and year (2006, 2007, 2008) influence on sulphate, nitrate and ammonium concentrations in precipitation. The results obtained are shown in the Fig. 2. The concentrations of sulphates and nitrates did not differ significantly between 2006 and 2007. Significantly higher concentrations of sulphate and nitrate were found in 2008 than in 2006 and 2007 what is probably related to the lowest amount of precipitation in 2008 (Fig. 2). Ammonium concentrations were also higher in 2008 than in the two previous years but the difference was not statistically significant. It is also evident that there was no significant difference in sulphate concentrations between the two study sites, neither between S-SO₄ concentrations in bulk open area precipitation, nor in S-SO₄ concentrations in throughfall (Fig. 2a). For nitrogen compounds (nitrate and ammonium), on the other hand, the results indicate significant differences between the two sites in bulk open field concentrations and in case of ammonium also in throughfall concentrations, with higher values having been found at Rájec (Figs. 2c, e). Significantly higher ($P < 10^{-4}$) at both sites were sulphate concentrations in throughfall within spruce stands than in bulk open area precipitation (Figs. 2a, c).

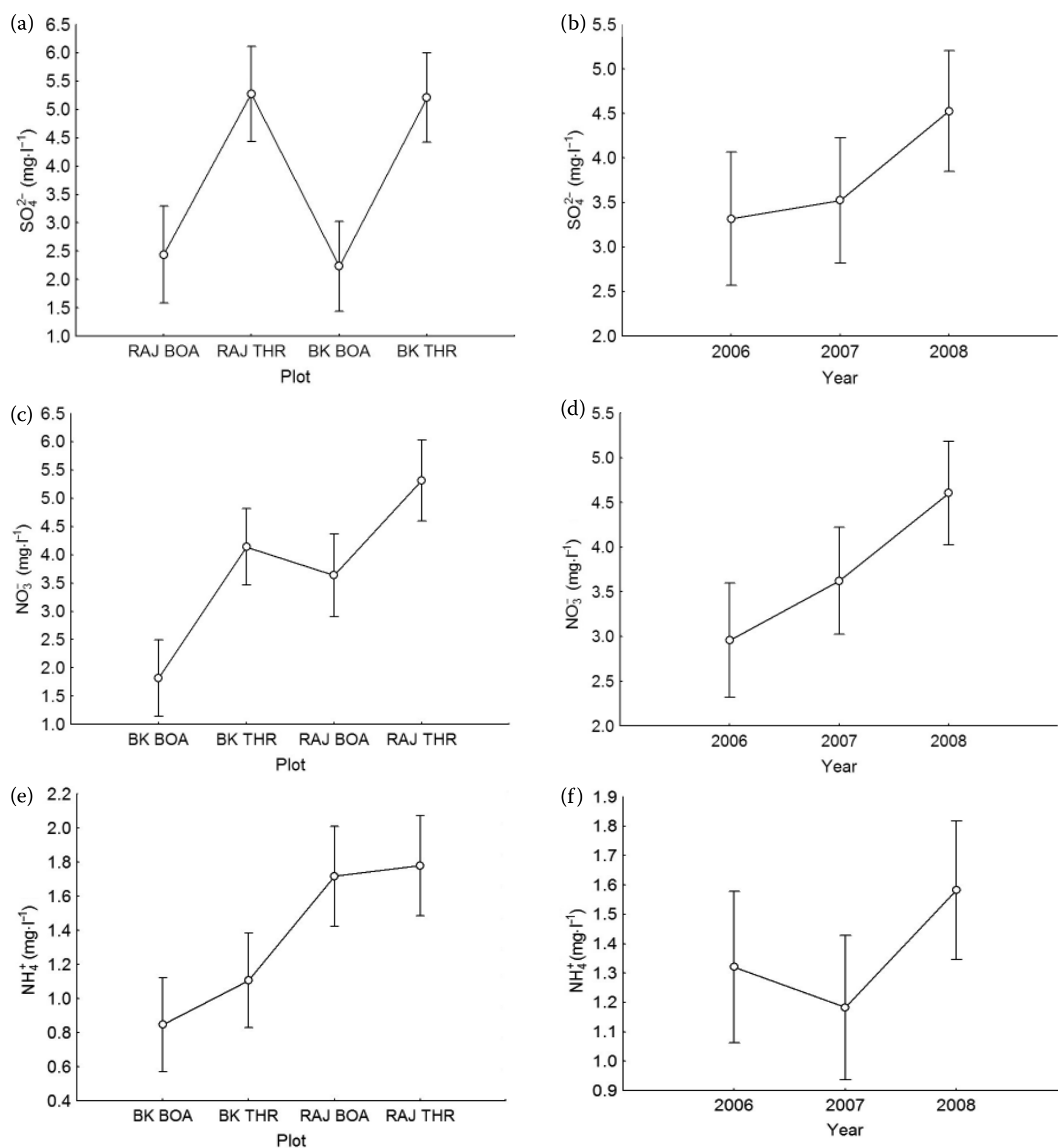
The relative differences between the element concentrations in bulk precipitation and throughfall are shown in the lower part of the Table 3. For sulphate ions the relative increase of concentration in throughfall (calculated as) was about 100% at both study sites during the three years studied. The relative increases of mean annual nitrate concentrations in throughfall in individual years were a bit lower at the both sites when compared with sulphate, namely: 55, 101, 105% and 65, 14 and 76% for

Bílý Kříž and Rájec, respectively during the three consecutive years (Table 3). The lowest concentration increase was observed for ammonium. In 2006 in Rájec the mean ammonium concentration in throughfall was even lower than the ammonium concentration in BOA precipitation.

Element fluxes in precipitation and throughfall

Annual bulk and throughfall fluxes of sulphur and nitrogen are depicted in the Fig. 3 with numerical values given in Table 4. Sulphur fluxes were markedly higher at Bílý Kříž than at Rájec both in the open area and in the throughfall in all years studied. The same holds for the fluxes of nitrate nitrogen in throughfall and in BOA precipitation (Fig. 3b). As concerns ammonium nitrogen, the fluxes were also higher at Bílý Kříž with some exceptions which occurred in 2006 (both BOA and throughfall flux was higher at Rájec) and in 2008 (BOA flux was higher at Rájec).

The throughfall fluxes of sulphur were, when considering the three years average, 2.1 times higher than open area fluxes at both study sites (see lower part of Table 4). The throughfall fluxes of nitrate were 1.9 times higher than open field fluxes at Bílý Kříž but only 1.6 times higher at Rájec (the values are three years averages). The throughfall fluxes of ammonium at Bílý Kříž were 1.3 times higher than open field fluxes. At Rájec the throughfall fluxes of ammonium were almost the same as the fluxes with BOA precipitation in all years but 2007 when the throughfall flux was about 1.2 times higher than open field flux. From the three years comparison of the two localities is evident that the soils under the spruce stand at Bílý Kříž received annually about 15.7 kg S·ha⁻¹ while soils at Rájec only 11.9 kg S·ha⁻¹ what makes difference of about 4 kg sulphur per ha and year. In case of sulphur the throughfall fluxes can serve as a good estimate of



	BK BOA			BK THR			RAJ BOA			RAJ THR		
	S-SO ₄	N-NO ₃	N-NH ₄	S-SO ₄	N-NO ₃	N-NH ₄	S-SO ₄	N-NO ₃	N-NH ₄	S-SO ₄	N-NO ₃	N-NH ₄
BK BOA	–	–	–	**	**	NS	NS	**	**	**	**	**
BK THR	**	**	NS	–	–	–	**	NS	*	NS	NS	*
RAJ BOA	NS	**	**	**	NS	*	–	–	–	**	*	NS
RAJ THR	**	**	**	NS	NS	*	**	*	NS	–	–	–
	2006			2007			2008					
	S-SO ₄	N-NO ₃	N-NH ₄	S-SO ₄	N-NO ₃	N-NH ₄	S-SO ₄	N-NO ₃	N-NH ₄	S-SO ₄	N-NO ₃	N-NH ₄
2006	–	–	–	NS	NS	NS	*	**	NS			
2007	NS	NS	NS	–	–	–	*	*	NS			
2008	*	**	NS	*	*	NS	–	–	–			

BOA – bulk open area precipitation, THR – throughfall, NS – not significant, * $P < 0.05$, ** $P < 0.01$ (Scheffé's post-hoc tests)

Fig 2. Results of two-way ANOVA analysis of sulphate, nitrate and ammonium concentrations measured in precipitation and throughfall at BÍlý Kříž and Rájec study sites during 2006–2008

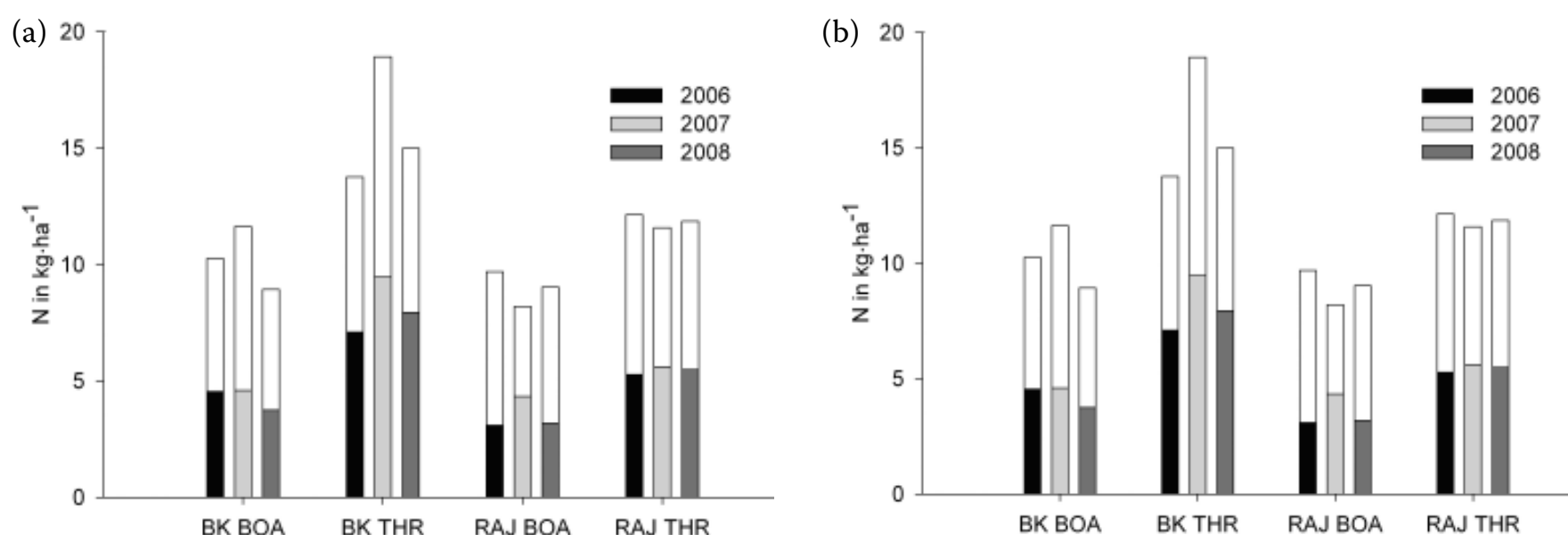


Fig 3. Annual deposition of sulphur (a) and nitrogen (b) at Bílý Kříž and Rájec study sites calculated for the years 2006–2008. Upper empty parts of bars in the second chart denote ammonium nitrogen; lower full parts of the bars denote nitrate nitrogen

total sulphur deposition (UN-ECE 2006). The annual input of inorganic nitrogen into the soil was also higher at Bílý Kříž with the average annual value of $15.9 \text{ kg} \cdot \text{ha}^{-1}$. In Rájec the average annual value was about $11.9 \text{ kg} \cdot \text{ha}^{-1}$.

DISCUSSION

Water fluxes

The negative values of interception could be explained by higher contribution of occult precipitation (fog and dew) to the throughfall at localities with more fog days. According to the TOLASZ (2007) the average annual number of days with fog exceeds 150 at the locality of Bílý Kříž. The contribution of fog as a water source to the water budget of forest ecosystems is documented by ZIMMERMANN et al. (1999). TESAŘ et al. (2002) have shown that the contribution of condensed fog water to the amount of bulk precipitation was about 9% in a catchment in the Šumava Mts. in the course of 1994–2000. SKY-

BOVA (2006) found that 22% of throughfall amounts came from occult precipitation in 2003–2004 in a spruce stand at Lysá hora mountain (1,324 m a.s.l.) in Beskids only 9 km from our experimental site.

Ion concentrations in bulk precipitation and throughfall

The concentrations of nitrogen and sulphur in bulk precipitation evaluated in this study (Table 3) were compared with values measured at the same sites in the past. KLIMO et al. (1996) evaluated the mean annual concentrations of sulphate and nitrate in bulk precipitation in Rájec for the period of 1976–1990. Average sulphate and nitrate concentrations in bulk precipitation were then $16.8 \text{ mg} \cdot \text{l}^{-1}$ and $7.1 \text{ mg} \cdot \text{l}^{-1}$, respectively. The distinct drop of average sulphate and nitrate concentrations to $2.0 \text{ mg} \cdot \text{l}^{-1}$ and $2.9 \text{ mg} \cdot \text{l}^{-1}$, respectively, found for 2006–2008 in our study corresponds well with the reduction of total emission of SO_2 and NO_x in the Czech Republic within the course of 1980–2000. In 2000 the sulphur dioxide and NO_x emissions, were 8 times and 2.4

Table 4. Annual element fluxes at Rájec and Bílý Kříž experimental study sites during 2006–2008 with open area (BOA) precipitation and with throughfall (THR)

Sampler/ plot	S- SO_4^{2-} ($\text{kg} \cdot \text{ha}^{-1}$)			N- NO_3^- ($\text{kg} \cdot \text{ha}^{-1}$)			N- NH_4^+ ($\text{kg} \cdot \text{ha}^{-1}$)			N _{inorg} ($\text{kg} \cdot \text{ha}^{-1}$)		
	2006	2007	2008	2006	2007	2008	2006	2007	2008	2006	2007	2008
BOA – Bílý Kříž	7.8	8.4	6.2	4.5	4.6	3.7	5.7	7.0	5.2	10.3	11.6	8.9
THR – Bílý Kříž	14.8	16.9	15.4	7.1	9.5	7.9	6.7	9.5	7.0	13.8	18.9	15.0
BOA – Rájec	3.7	4.2	3.3	3.1	4.3	3.2	6.7	3.9	5.8	9.8	8.2	9.0
THR – Rájec	8.1	8.3	6.7	5.3	5.6	5.5	6.8	6.0	6.4	12.1	11.6	11.9
The relative increase of annual element fluxes in throughfall when compared with bulk precipitation in%: ($100\% \times \text{THR}/\text{BOA}$) – 100												
Bílý Kříž	89	101	146	56	106	112	17	34	36	34	63	68
Rájec	117	96	105	70	29	72	1	55	9	23	41	31

times respectively, lower than those in 1980 according to HRUŠKA and CIENCIALA (2002), KOPÁČEK and VESELÝ (2005) and MAJER et al. (2005). KULHAVÝ et al. (2001) evaluated element concentrations in bulk precipitation at the Bílý Kříž site for the year 1999: the sulphate and nitrate concentrations were $2.9 \text{ mg}\cdot\text{l}^{-1}$ and $2.2 \text{ mg}\cdot\text{l}^{-1}$, respectively. The corresponding values found in our study were $1.9 \text{ mg}\cdot\text{l}^{-1}$ and $1.6 \text{ mg}\cdot\text{l}^{-1}$, respectively, which again agree with a 20–30% decrease of SO_2 concentrations in the air in the Czech Republic between 1999 and 2007 (MŽP 2008). The decrease in nitrate concentration at Bílý Kříž between 1999 and years of this study can only partly be explained from the overall decrease (12% between 199 and 2007 according to MŽP 2008) of NO_x concentrations in the air in the Czech Republic. Nitrogen oxides in general have a shorter lifespan in the lower atmosphere and their concentrations are more likely to be influenced by local sources of pollution in shorter time horizons.

Annual mean concentrations of sulphates, nitrates and ammonium in bulk open area precipitation measured at Rájec and Bílý Kříž fit in the range of values measured on higher elevated sites throughout the Europe as is apparent from the Table 5.

The 100% relative increase of sulphate concentration in throughfall vs BOA precipitation found for our study sites (Table 3) can be compared with the data published by KOPÁČEK et al. (2009) for the four mature (150 years old) spruce stands in the Bohemian Forest. At the altitudes of 1,000–1,330 m a.s.l. for the period 1998–2007 the relative increase of sulphur concentrations under the crowns was 50–100%. BERGER et al. (2008) study of a 65 years old spruce stand near Kreisbach (Austria) found a 129% relative increase of sulphur concentration under the crowns. Sulphate is a conservative anion which means it is neither taken up by canopy nor leached from the foliage (DE VRIES et al. 2001; BALESTRINI et al. 2007) or, more precisely said, the minor stomatal uptake of SO_2 is balanced by minute leaching of SO_4^{2-} from needles (CAPE et al. 1992; DRAAIJERS et al. 1996). The increase of sulphate concentration in throughfall can be fully assigned to the washing of dry deposits accumulated on the canopy between precipitation events. At both study sites the conditions influencing dry deposition (stand age, structure and composition, content of sulphur compounds in the air, humidity conditions) are similar and thus we found a similar increase of sulphate concentrations in throughfall.

While in case of sulphate, the concentrations in BOA precipitation were not significantly different at the two study sites, significantly higher concen-

trations of nitrate and ammonium were found in BOA precipitation at Rájec than at Bílý Kříž. This can be explained by the different character of the two study sites. The spruce stand in Rájec is embedded into agricultural landscape with three villages within the distance of 1.5 km from the stand and two local minor roads within 0.5 km. The spruce stand in Bílý Kříž is quite detached in a mountainous region. Agriculture, especially livestock production, is the dominant source of atmospheric NH_4 (ANEJA et al. 2001; EKL 2005; ERISMAN 2007; MŽP 2008) and also contributes significantly to nitrogen oxides emissions. The combustion of fossil fuels (i.e. transport and industry) produces the main part of atmospheric nitrogen oxides. From this point of view, the lower concentrations of ammonia nitrogen at Bílý Kříž are understandable.

The biogeochemical behavior of inorganic N in canopy differs both between the sites and between the reduced and oxidized form of nitrogen. While nitrate concentrations were significantly higher in throughfall vs BOA precipitations at both sites, only insignificant increases in throughfall concentrations were observed for ammonium. It suggests assimilation of NH_4^+ by canopy foliage. $\text{NH}_4\text{--N}$ retention in coniferous forest canopy was described e.g. by PIIRAINEN et al. (1998) and HOULE et al. (1999). Spruce canopies have been shown to be active sinks for inorganic N, and to have the highest biomass of epiphytic lichens (capable to assimilate nitrogen compounds) compared to pine and deciduous canopies (LOVETT 1992; HOULE et al. 1999). The higher efficiency of the spruce forest in Rájec in retaining nitrogen which (see ANOVA results Figs. 2c, e) could be explained by more favorable climatic conditions in Rájec which support metabolic activities of the microorganisms assimilating nitrogen compounds.

Element fluxes in precipitation and throughfall

In case of sulphur the throughfall fluxes can serve as a good estimate of total sulphur deposition (DE VRIES et al. 2001). The total sulphur deposition was greater at Bílý Kříž (due to higher throughfall amounts (Table 4), because throughfall concentrations were similar at the both plots. Also, the BOA and throughfall fluxes of N-NO_3 were higher at Bílý Kříž in all years studied despite the fact that corresponding concentrations were higher at Rájec during the whole period. The throughfall fluxes of sulphur were 2.0 to 2.4 times higher than open area fluxes at both study sites (see lower part of Table 4). These values are in accordance with values published by other authors. AUGUSTO et al. (2002) re-

viewed several papers bringing data from European forest sites and found that atmospheric deposition of sulphur had been 2 to 3 times higher in stands of *Picea abies* or *Pinus sylvestris* than in open areas.

The element fluxes at Bílý Kříž and Rájec were compared with bulk open area and throughfall fluxes measured in other parts of the Czech Republic at experimental plots with deposition measurement included in the ICP-Forests monitoring programme. The data from 12 localities, where deposition monitoring had been performed during for 2006–2007 were recently published in the annual report (BOHÁČOVÁ et al. 2009). The ICP intensive monitoring plots (level II plots) were installed in 1994–2004 to cover main tree species in their typical regions of the Czech Republic. It can be seen that the locality of Bílý Kříž belongs to the sites with higher sulphur flux with both bulk precipitation and throughfall.

Higher sulphur fluxes in the open field were measured in 2006 and 2007 at Jizerka in Jizerské Mts. (9.70 and 14.28 kg S·ha⁻¹·a⁻¹), Mísečky in Krkonoše Mts. (10.11 and 12.62 kg S·ha⁻¹·a⁻¹), and in 2006 also at Klepačka in Beskids (10.21 kg S·ha⁻¹·a⁻¹). Higher sulphur fluxes with throughfall were registered in 2006 and 2007 at Jizerka (21.32 and 27.13 kg S·ha⁻¹·a⁻¹), at Luisino údolí in Orlické Mts (34.49 and 34.13 kg S·ha⁻¹·a⁻¹) and at Klepačka only in 2006 (20.97 kg S·ha⁻¹·a⁻¹) (all data from BOHÁČOVÁ et al. 2009). Norway spruce is the main species in all stands with these high sulphur throughfall fluxes what affirms the higher filtering capacity of spruce canopy (HOJJATI et al. 2009). Nitrogen fluxes at Bílý Kříž are neither low nor high when compared with the ICP plots. Rájec belongs in the Czech Republic to the localities with lower load of sulphur and nitrogen.

Table 5. Annual element concentrations in bulk open field precipitation measured at various mountainous regions in Europe in comparison with values measured at Bílý Kříž and Rájec

	Concentration	Country (locality)
SO ₄ ²⁻ (mg·l ⁻¹)	1.81–2.01	Bílý Kříž (Czech Republic) ¹
	1.68–2.30	Rájec (Czech Republic) ²
	0.81–1.21	Czech Republic ³
	0.98–1.44	Czech Republic ⁴
	1.59–3.99	Germany ⁵
	1.69–4.57	Germany ⁶
	3.50–4.52	Switzerland ⁷
	3.82 (five-year average)	Poland ⁸
NO ₃ ⁻ (mg·l ⁻¹)	1.59–1.72	Bílý Kříž (Czech Republic) ¹
	2.08–3.49	Rájec (Czech Republic) ²
	1.42–1.59	Czech Republic ³
	1.54–1.95	Czech Republic ⁴
	1.12–2.63	Germany ⁵
	1.56–2.64	Germany ⁶
	2.39–3.06	Switzerland ⁷
	3.72 (five-year average)	Poland ⁸
NH ₄ ⁺ (mg·l ⁻¹)	0.63–0.71	Bílý Kříž (Czech Republic) ¹
	0.90–1.64	Rájec (Czech Republic) ²
	0.39–0.51	Czech Republic ³
	0.44–0.58	Czech Republic ⁴
	0.41–1.00	Germany ⁵
	0.63–1.23	Germany ⁶
	0.72–1.06	Switzerland ⁷
	1.47 (five-year average)	Poland ⁸

¹Bílý Kříž (Moravian-Silesian Beskids Mts.), 908 m a.s.l., 2006–2008; ²Rájec (Drahanská upland), 610 m a.s.l., 2006–2008; ³Bohemian Forest, 1,080 m a.s.l., 1998–2007 (KOPÁČEK et al. 2009); ⁴Bohemian Forest, 1,180 m a.s.l., 1998–2007 (KOPÁČEK et al. 2009); ⁵Erzgebirge – Oberbärenburg, 735 m a.s.l., 1993–2002 (ZIMMERMANN et al. 2006); ⁶Erzgebirge – Zinnwald, 877 m a.s.l., 1993–2002 (ZIMMERMANN et al. 2006); ⁷Southern Alps - Novaggio, 950 m a.s.l., 1997–2001 (THIMONIER et al. 2005); ⁸Dupniański Stream Catchment – Silesian Beskid Mts., 725 m a.s.l., 1999–2003 (MALEK, ASTEL 2005)

CONCLUSION

The comparative study of sulphur and nitrogen compounds concentrations in atmospheric precipitation and throughfall at two forest sites with young spruce monoculture in the Czech Republic has revealed some differences between the two sampling areas. While the differences between the sites in sulphate concentrations were not significant neither in bulk open area precipitation nor in throughfall, significant differences were observed in ammonium nitrogen and nitrate nitrogen concentrations. The concentrations of ammonium nitrogen were significantly higher at Rájec than at Bílý Kříž both in open area bulk precipitation and in throughfall. Concentrations of nitrate nitrogen were significantly higher in bulk open area precipitation at Rájec than at Bílý Kříž. The throughfall nitrate concentrations were also higher at Rájec but the difference was evaluated as non significant on the significance level of 0.05. Higher concentrations of nitrogen compounds in precipitations at Rájec can be explained by the influence of local agriculture and traffic on atmospheric pollution. The spruce canopy at the Rájec stand retained nitrogen more efficiently than that at Beskids.

Sulphur fluxes in the years 2006–2008 were markedly higher at Bílý Kříž than at Rájec both in the open area (6.2–8.4 kg S·ha⁻¹·a⁻¹ at Bílý Kříž vs 3.3–4.2 kg S·ha⁻¹·a⁻¹ at Rájec) and in the throughfall (14.8–16.9 kg S·ha⁻¹·a⁻¹ at Bílý Kříž vs 6.7–8.3 kg S·ha⁻¹·a⁻¹ at Rájec). The locality of Bílý Kříž belongs to the sites in the Czech Republic with higher sulphur deposition load with both bulk precipitation and throughfall. It could be caused by the fact that the NW wind flowing mainly in January and February brings to this site air polluting substances from the Ostrava industrial agglomeration.

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