

Evaluation of deposition fluxes in two mountain Norway spruce stands with different densities using the extended Canopy Budget Model

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ABSTRACT: The field study in a mountain forest at Bílý Kříž provides a comparison of element fluxes for two adjacent forest spruce stands of the same age (29 years in 2005) but with different stem densities. During five years (2001–2005), bulk and throughfall precipitation was sampled and analysed. Total deposition, dry deposition and canopy exchange fluxes were evaluated on the basis of the Canopy Budget Model. Highly significant differences in base cations, dissolved organic carbon, SO_4^{2-} , F^- , and Cl^- throughfall concentrations were found between the sparser and denser spruce stands. Throughfall, dry deposition and canopy exchange fluxes were also influenced by stand density. Annual throughfall fluxes of inorganic nitrogen were within $11.9\text{--}17.8\text{ kg N}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ on the sparser plot and within $15.4\text{--}20.6\text{ kg N}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ on the denser plot; annual throughfall fluxes of sulphur were within $15.3\text{--}20.3\text{ kg S}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ on the sparser plot and within $20.7\text{--}27.7\text{ kg S}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ on the denser plot. The critical load for nitrogen ($11.2\text{ kg N}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$) was exceeded on both plots in all evaluated years 2002–2005. Total annual inorganic nitrogen deposition was higher by up to 37.5% (in 2002) on the denser plot than on the sparser one.

Keywords: atmospheric deposition; thinning; throughfall; dry deposition; precipitation chemistry; total deposition

It was recognized in the 1970s that air pollution had serious ecological and economic consequences. The forest decline which affected forests in Europe and North America during the 1970s and 1980s was caused by several factors. The most important of them were an increased input of acidifying substances and inappropriate forest management. The adverse effect of increased deposition of nutritional N on forest and other ecosystems was also identified (ABER et al. 1989; BOBBINK et al. 1998). Countries of the UN Economic Commission for Europe (UNECE) developed a legal, organizational, and scientific framework to deal with these problems. The UNECE Convention on Long-range Transboundary Air Pollution (LRTAP) was the first international legally binding instrument to deal with problems of air pollution in Europe on a regional basis (www.unece.org/env/lrtap). Signed in 1979, it

was implemented in 1983. Emissions of air pollutants have been dramatically reduced since then. The Czech Republic achieved a substantial decrease in sulphur emissions in a relatively short time during the 1990s. Ongoing research aims to answer more and more precisely the basic question: what level of pollution is safe for the forests and other ecosystems. The concept of critical loads was developed by NILSSON and GRENNFELT (1988): The critical load is a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge. Within the LRTAP, model-based calculations have been reviewed and synthesized to set critical loads for atmospheric N deposition and acidity for various ecosystems (ACHERMANN, BOBBINK 2003; SPRANGER et al. 2004; DE BAKKER et al. 2007).

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Atmospheric deposition comprises two basic components: **wet deposition** (the flux of dissolved substances from the atmosphere with precipitation) and **dry deposition** (the flux of particles and gaseous substances from the atmosphere during dry periods) (definitions in ULRICH et al. 2006). Two methodological approaches have been developed by researchers to measure and assess atmospheric deposition components. One of them is based on micrometeorology (SEINFELD, PANDIS 1998; WESELY, HICKS 2000); the other is based on the monitoring and sampling of forest stand throughfall and open plot precipitation (DE VRIES et al. 2001; STAELENS et al. 2008). The second approach was chosen for the deposition monitoring programme within the framework of International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests and was used also in this study. Wet deposition could be determined directly if wet-only sampling devices are deployed. The collectors open up automatically only during the rain or snow events. For economic reasons permanently open collectors used to be applied instead of wet-only samplers (DRAAIJERS et al. 1998). In these cases bulk deposition is measured, which includes dry deposition trapped during dry periods. Alternatively, wet deposition can be estimated from the bulk deposition if the value of the average ratio of wet only to bulk deposition for given elements and relevant localities is known (e.g. THIMONIER et al. 2005). Throughfall is sampled by collectors placed under the canopy. The composition of throughfall depends on the wet deposition input, dry deposition washed from foliage and branches, and canopy exchange processes. Calculations of dry deposition fluxes, total element fluxes, canopy uptake and canopy leaching fluxes from the throughfall and bulk precipitation data were described by DE VRIES et al. (2001). Deposition fluxes of elements, and hence the accumulation of acidifying and other pollutants, depends on the site characteristics and is influenced by the terrain, vegetation, stand age, and the microclimate of the site (ERISMAN, DRAAIJERS 2003; DE SCHRIJVER et al. 2007; MALEK, ASTEL 2008). One factor that influences deposition fluxes in forested areas is forest stand density. Forest thinning can cause marked changes in the deposition pattern (BÄUMLER, ZECH 1997). In this paper the results of 5-year (2001–2005) monitoring of deposition fluxes in two adjacent Norway spruce stands with different stem density in the Moravian Silesian Beskydy Mts. are presented. The goals are to:

- quantify total deposition (*TD*), dry deposition (*DD*), and canopy exchange (*CE*) fluxes for in-

dividual elements on the basis of the extended Canopy Budget Model (DE VRIES et al. 2001),

- compare the deposition fluxes in the adjacent Norway spruce stands with different densities,
- compare the nitrogen deposition fluxes with empirical values of critical load given in Mapping Manual UNECE Convention on Long-range Transboundary Air Pollution (SPRANGER et al. 2004) and with the value of critical load calculated for the site by ZAPLETAL (2006).

MATERIAL AND METHODS

Study sites

The Bílý Kříž site is situated in the top part of the Moravian-Silesian Beskydy Mts. in the Czech Republic (49°30'N, 18°32'E; 908 m a.s.l.) (Fig. 1, Table 1). The bedrock is flysch with a predominance of Godula sandstone. The soil type is humo-ferric podzol with the mor-moder humus. The soils are of 50–60 cm depth, sandy loam to sandy clay loam, the soil texture being characteristic of sandy flysch sediments with 15–35% of clay fraction, highly skeletal in the depth.

The region is moderately cold and wet. Mean air temperature was 5.5°C, mean annual precipitation was 1,300 mm, the number of days with precipitation above 1 mm was 140–160, and the length of the growing season was 120–140 days during the period 1993–2003 (JANOŠ et al. 2004). Snow precipitation represents 16–24% of the annual precipitation (ČERVENÝ 1984). Mean relative air humidity at the site was about 82% during 1998–2007 (MARKOVÁ et al. 2009). Although the precipitation totals are rather high, occasional droughts occurred in the



Fig. 1. Position of the experimental site at Bílý Kříž, Beskydy Mts.
full square – experimental site

Table 1. Characteristics of the experimental site at Bílý Kříž (KULHAVÝ et al. 2001)

| Locality | Plot | Latitude | Longitude | Altitude (m) | Number of trees in 2000 (trees·ha ⁻¹) | Main species | FAO soil unit | Humus type |
|-----------|------|----------|-----------|--------------|---|--------------------------------|--------------------|------------|
| Bílý Kříž | FD | 49°30'N | 18°32'E | 908 | 2,600 | <i>Picea abies</i> (L.) Karst. | humo-ferric podzol | mor-moder |
| | FS | | | | 2,100 | | | |

area (HADAŠ 2007). The mild northwesterly wind in January and February brings air pollutants from the Ostrava industrial agglomeration. The region where Slovakia, Poland and the Czech Republic meet was one of the most polluted sites in Europe, especially by sulphur and nitrogen compounds (ZAPLETAL 2006). The 1st generation of spruce monoculture was prematurely felled after the frost disaster potentiated by emissions in 1978/1979 (RAŠKA 1985). The 2nd generation of spruce (*Picea abies* [L.] Karst.) was planted in 1981 using 4-year-old plants. The forest type at the site is acid *Abieto-Fagetum* with *Oxalis acetosella*, *Vaccinium myrtillus*, *Deschampsia flexuosa* and *Rubus hirtus* in the undergrowth. At the site, experimental plots were established in two contiguous stands with different densities (FD – dense stand, FS – sparse stand) each occupying the area of 2,500 m². The mean inclination of the slope where the stands are situated is 30° and its aspect is SE. The age of trees was 29 years in 2005. The development of stand densities during the years 1999–2005 is shown in Table 2. A more detailed description of stand characteristics can be found in POKORNY et al. (2008).

Sampling design

For throughfall and bulk precipitation sampling, funnel type collectors were used according

to BLOCK and BARTELS (1985) and NIEHUS and BRUGGEMANN (1995). The containers were made of polyethylene and they were inserted into thick-walled plastic pipes in order to shield the samples from direct solar radiation and to hold the funnels approximately 1 m above the ground. The upper edges of the funnels had sharp teeth to prevent contamination by bird droppings. The collection area of each funnel was 335 cm². There were 7 collectors randomly distributed at each plot. During winter seasons the number of collectors was reduced to 5 at each plot since the access to the research plots was difficult because of snow. Bulk atmospheric precipitation was sampled with one collector at the centre of a grassy open area situated about 100 m away from the stands. All samplers were installed in the spring of 1998. Samples were taken every month during winter and fortnightly during the rest of the year, resulting in 20–21 sampling events every year. During the winter the whole collectors, including the funnels, filters, and storage bottles, were replaced by clean ones. The amount of precipitation was measured in the laboratory after the snow and ice had melted. During the snow-free season the amount of precipitation was measured directly on the site and only 500 ml of each sample were taken to the laboratory in clean polyethylene bottles. The funnels and storage bottles were washed and rinsed with distilled water in the field and the filters were replaced.

Table 2. The density and leaf area index for the spruce stands FD and FS at Bílý Kříž in 1999–2005 (CHMI 2008)

| Year | Stand densities (trees·ha ⁻¹) | | | Leaf area index (m ² ·m ⁻²) | | |
|------|---|---------------------|-------|--|-------|-------|
| | FD | FS | FD-FS | FD | FS | FD-FS |
| 1999 | 2,600 | 2,100 | 500 | 10.84 | 7.95 | 2.89 |
| 2000 | 2,600 | 2,100 | 500 | 11.00 | 8.22 | 2.78 |
| 2001 | 2,600 | 1,880 [#] | 720 | 11.54 | 6.71 | 4.83 |
| 2002 | 2,500 ^{##} | 1,820 ^{##} | 680 | 11.68 | 7.69 | 3.99 |
| 2003 | 2,440 ^{##} | 1,820 | 620 | 12.34 | 9.14 | 3.20 |
| 2004 | 2,048 ^{##} | 1,652 ^{##} | 396 | 12.44 | 9.58 | 2.86 |
| 2005 | 2,044 | 1,652 | 392 | 11.82 | 10.01 | 1.81 |

[#]planned thinning, ^{##}natural thinning, FD – dense stand, FS – sparse stand

Sample preparation and chemical analyses

The samples were prepared the next day after the sampling. During winters, when samples contained snow and ice, it was necessary to wait until the ice melted. In any case the time between sample collection and sample preparation was shorter than 3 days. pH and conductivity were measured on each sample (methods are described in ČSN EN 27888 and in ČSN ISO 10523). Composite throughfall samples were prepared by mixing the samples per plot based on the original precipitation volumes in the collectors. The composite samples of throughfall and the sample from the open area were then filtered through a 0.45 µm MILIPORE membrane filter (Milipore Corporation, Billerica, MA, USA). After the filtration, each sample was divided into subsamples for individual analyses of different analytes. The subsamples for the determination of base cations (Na^+ , K^+ , Mg^{2+} , and Ca^{2+}) were measured following acidification by adding 0.5 cm³ of reagent-grade nitric acid per 100 cm³ within a week using the flame atomic absorption spectrophotometry for Ca^{2+} and Mg^{2+} and flame atomic emission spectrophotometry for Na^+ and K^+ (AA 30 F4 VARIAN spectrometer, air-acetylene flame). All anions, i.e. SO_4^{2-} , NO_3^- , NO_2^- , Cl^- and F^- , were analysed using ion exchange chromatography with KOH gradient elution [DX-600 ion chromatographic system equipped with a GP50 gradient pump, an ED50 electrochemical detector, an EG40 eluent generator and an IonPac[®] AS11-HC AS11 HR (250 × 2 mm) analytical column with an AG11 HR (50 × 2 mm) guard column, operated under PeakNet 6.0 software, all parts Dionex Corporation, Sunnyvale, CA, USA]. When anions could not be analysed immediately for technical reasons, the subsamples for the determination of anions were frozen until analyses could be performed. Dissolved organic carbon (DOC) was analysed using a TOC analyser (Shimadzu, Kyoto, Japan) with combustion at 680°C and subsequent infrared detection of CO_2 according to the standard ČSN EN 1484. For the determination of ammonium a manual spectrometric method was used based on the reaction of ammonium ions with salicylate and hypochlorite in the presence of sodium nitroprusside (ČSN ISO 7150-1).

Calculations and estimation of missing data

No special collectors for snow were used during winters and therefore precipitation volumes were incorrect in cases of heavy snow and/or frost. In these cases snow caps were formed on the tops of

samplers; snow was lost from the caps by wind erosion. Therefore fourteen percent of precipitation amounts had to be estimated. The regressions of precipitation amounts measured with our samplers and the amounts for the relevant periods computed from daily totals measured by the Czech Hydrometeorological Institute (CHMI) were used to calculate the correction. The station of CHMI was located directly on the site near the open area bulk precipitation collector (at the distance of 7 m). The amounts of water collected in the first sampling interval every year were divided into two parts: one belonging to the end of the previous year and the other belonging to the beginning of the particular year. The shares were calculated on the basis of precipitation totals belonging to these periods [daily totals were measured by CHMI (2008)] and data provided by Radek Pokorný.

Several times per year, during the summer, the collected amount of water was not sufficient to perform all analyses. In these cases the mean annual concentrations were used in calculations instead of the missing values. These situations occurred no more than twice a year. If a measured value was under the detection limit of the analytical method, a value equal to one half of this detection limit was used.

The mean annual concentrations of individual ions and the mean annual solution conductivities were calculated as volume weighted means using the formula:

$$\bar{c} = \frac{\sum_{i=1}^n c_i \times V_i}{\sum_{i=1}^n V_i} \quad (1)$$

where:

\bar{c} – volume weighted annual concentration in mg·l⁻¹ or conductivity in µS·cm⁻¹,

n – number of sampling events,

c_i – concentration of the ion in mg·l⁻¹ or conductivity of the sample in µS·cm⁻¹ for the i^{th} sampling event,

V_i – amount of water sampled in the i^{th} sampling event.

The mean pH values were calculated using the same formula but measured pH values were converted to concentrations of H^+ ion before calculation. The result, i.e. the mean annual concentration of H^+ ion, was then reconverted to pH.

Assessment of total deposition fluxes and dry deposition fluxes according to the Canopy Budget Model

Throughfall (TF) and bulk precipitation fluxes (BP) were calculated by multiplying the mean annu-

al concentration and the total amount of throughfall or bulk precipitation for each particular year.

Wet deposition (*WD*) data and annual water fluxes were taken from the web site of the Czech Hydrometeorological Institute (CHMI) (CHMI 2005). The CHMI uses an automatic pluviocollector for precipitation sampling. Their station at Bílý Kříž runs since 1989. Daily wet only precipitations were pooled to weekly samples to be analyzed.

Stemflow (*SF*) was not measured at the studied plots and had to be estimated from the annual throughfall fluxes. The relationships described in the Technical Report EC-UN/ECE (DE VRIES et al. 2001) were used:

$$SF_i = TF_i \times \frac{\alpha}{1 - \alpha} \quad (2)$$

where:

SF_i – stemflow ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

TF_i – throughfall ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

i – given ion (H^+ , Ca^{2+} , Mg^{2+} , K^+ , Na^+ , NH_4^+ , NO_3^- , SO_4^{2-} , Cl^-),

α – empirical value calculated as a function of stand age.

For our 25–29 years old spruce stand:

$$\alpha = 0.31 - 0.0034 \times \text{age} \quad (3)$$

Total deposition fluxes (*TD*), canopy exchange fluxes (*CE*), and dry deposition fluxes (*DD*) were calculated on the basis of the extended canopy budget model (DE VRIES et al. 2001, 2003). This model works on the assumption that the total deposition flux of a particular ion is given by the equation (4):

$$TD = TF + SF \pm CE = TF + SF - Cle + CU = WD + DD \quad (4)$$

where:

TD – total deposition fluxes ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

SF – stemflow ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

TF – throughfall ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

CE – canopy exchange fluxes ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

Cle – canopy leaching ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

CU – canopy uptake ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

DD – dry deposition fluxes ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

WD – wet deposition ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$).

Canopy exchange of Na^+ , Cl^- , and SO_4^{2-} ions is considered to be negligible (RAGSDALE et al. 1992). For these ions total depositions are set equal to the sum of throughfall and stemflow fluxes:

$$TD_i = TF_i + SF_i \quad (5)$$

where:

TD_i – total deposition fluxes of the ion i ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

TF_i – throughfall of the ion i ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

SF_i – stemflow of the ion i ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

i – stands for Na^+ , SO_4^{2-} or Cl^- .

Total depositions of base cations (BC: Ca^{2+} , Mg^{2+} and K^+) are estimated by multiplying the bulk depositions of those cations with the ratio of the sodium input by both throughfall and stemflow according to the formula:

$$TD_i = \frac{TF_{\text{Na}} + SF_{\text{Na}}}{BP_{\text{Na}}} \times BP_i \quad (6)$$

where:

TD_i – total deposition fluxes of the ion i ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

TF_{Na} – throughfall of sodium ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

SF_{Na} – stemflow of sodium ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

BP_{Na} – flux of sodium in bulk precipitation ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

BP_i – flux of ion i in bulk precipitation ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

i – stands for Ca^{2+} , Mg^{2+} , or K^+ .

Canopy exchange (canopy leaching in case of Ca^{2+} , Mg^{2+} , and K^+) is then computed as the difference between the sum of the particular ion in throughfall and stemflow minus its total deposition:

$$Cle_i = TF_i + SF_i - TD_i \quad (7)$$

where:

Cle_i – canopy leaching of ion i ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

TF_i – throughfall of the ion i ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

SF_i – stemflow of the ion i ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

TD_i – total deposition fluxes of the ion i ($\text{mol} \cdot \text{ha}^{-1} \cdot \text{yr}^{-1}$),

i – stands for Ca^{2+} , Mg^{2+} , or K^+ .

Total canopy uptake of NH_4^+ and H^+ is assumed to be equal to the total canopy leaching of Ca^{2+} , Mg^{2+} , and K^+ , taking place through ion exchange, while subtracting the leaching of weak acids (*WA*), i.e. the sum of bicarbonate and organic acids anions:

$$CU_{\text{NH}_4^+} + CU_{\text{H}^+} = Cle_{\text{Ca}^{2+}} + Cle_{\text{Mg}^{2+}} + Cle_{\text{K}^+} - Cle_{\text{WA}} \quad (8)$$

For calculation of NH_4^+ and H^+ uptake, I assumed (according DE VRIES et al. 2001) that H^+ exchange capacity is six times larger than NH_4^+ :

$$CU_{\text{H}^+} = \frac{6 \times TF_{\text{H}^+}}{TF_{\text{NH}_4^+} + 6 \times TF_{\text{H}^+}} \times (Cle_{\text{Ca}^{2+}} + Cle_{\text{Mg}^{2+}} + Cle_{\text{K}^+} - Cle_{\text{WA}}) \quad (9)$$

The leaching of weak acids is calculated by subtracting their TD from their measured TF and SF .

The dry deposition of weak acids is assumed to equal the bulk deposition (total deposition equals twice the bulk deposition) (ULRICH et al. 2006).

$$Cle_{WA} = TF_{WA} + SF_{WA} - 2 \times BP_{WA} \quad (10)$$

WA stands for weak acids, i.e. the sum of organic acid ($RCOO^-$) concentration and bicarbonate (HCO_3^-).

Total deposition of protons is (as results from Eq. 4):

$$TD_{H^+} = TF_{H^+} + SF_{H^+} + CU_{H^+} \quad (11)$$

The inclusion of weak acid leaching also requires an estimate of the concentration of weak acids in both bulk deposition and throughfall, because the concentration of bicarbonate and organic acids was not measured directly. This estimation of the weak acid concentration was based on the method described by DE VRIES et al. (2001): the HCO_3^- concentration was derived from the sample pH and from assumed atmospheric CO_2 pressure (Eq. 12), and $RCOO^-$ derived from DOC (Eq. 13):

$$HCO_3^- = \frac{K_{CO_2} \times pCO_2}{H^+} \quad (12)$$

where :

HCO_3^- – organic acid concentration in $\mu mol \cdot l^{-1}$,

$K_{CO_2} = 10^{-7.8} mol^2 \cdot l^{-2} \cdot bar^{-1}$ (i.e. dissociation constant of CO_2),

$pCO_2 = 0.0003 bar$ (i.e. partial CO_2 pressure),

H^+ – proton concentration ($mol \cdot l^{-1}$).

$$RCOO^- = m \times DOC \times \frac{K_a}{H^+ + K_a} \quad (13)$$

where:

$RCOO^-$ – organic acid functional group concentration ($\mu mol \cdot l^{-1}$),

$m = 5.5 mol \cdot kg^{-1}$ (i.e. concentration of acidic functional groups on dissolved organic carbon),

DOC – dissolved organic carbon concentration (in $mg \cdot l^{-1}$),

K_a – estimated dissociation constant for organic acid in $mol \cdot l^{-1}$ [Eq. (14)],

H^+ – proton concentration ($mol \cdot l^{-1}$).

$$pK_a = a + b \times pH - c \times pH^2 \quad (14)$$

where: $a = 0.96$, $b = 0.90$, $c = 0.039$.

Dry deposition fluxes (DD) for all elements were calculated as:

$$DD_i = TD_i - WD_i \quad (15)$$

where:

DD_i – total deposition fluxes of the ion i ($mol \cdot ha^{-1} \cdot yr^{-1}$),

TD_i – total deposition fluxes of the ion i ($mol \cdot ha^{-1} \cdot yr^{-1}$),

WD_i – wet deposition fluxes of the ion i ($mol \cdot ha^{-1} \cdot yr^{-1}$),

i – stands for H^+ , Ca^{2+} , Mg^{2+} , K^+ , Na^+ , NH_4^+ , NO_3^- , SO_4^{2-} , Cl^- .

Statistics

Basic statistical calculations (means, standard deviations, regressions) were done by Microsoft Office Excel 2007. The Wilcoxon signed-rank test was performed to test whether ion concentrations and conductivities were significantly different among plots. The non-parametric test was used because the distribution of differences was not normal according to the Shapiro and Wilk's test for normality. These analyses were done using STATISTICA 8 (StatSoft, Tulsa, USA).

RESULTS

Precipitation totals and canopy interception

Annual precipitation totals calculated from water amounts captured by the bulk open field collector varied between 1,014 and 1,616 mm during the 2001–2005 period (Table 3). Monthly average precipitation measured at the meteorological station is depicted in Fig. 2. July was the month richest in precipitation during 2001–2005 with the monthly total of approximately 168 mm. High precipitation totals were measured also in January and Febru-

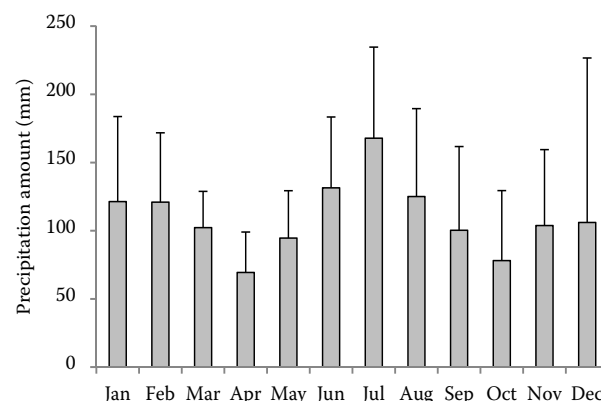


Fig. 2. Average monthly values of rainfall (mm) calculated for the period 2001–2005 from values measured at the CHMI meteorological station at Bílý Kříž

Error bars represent standard deviations at the CHMI meteorological station at Bílý Kříž

Table 3. Annual precipitation and volume-weighted mean annual element concentrations, pH and conductivity at 20°C in wet, bulk open field and throughfall precipitation, Bílý Kříž experimental forest site

| Year | Collector | Amount of precipitation (mm) | pH | Conductivity at 20°C ($\mu\text{S}\cdot\text{cm}^{-1}$) | H ⁺ Ca ²⁺ Mg ²⁺ Na ⁺ K ⁺ F ⁻ Cl ⁻ SO ₄ ²⁻ NO ₂ ⁻ NO ₃ ⁻ NH ₄ ⁺ | | | | | | | | | | |
|------|-----------|------------------------------|------|---|---|------|------|------|------|------|------|-------|-----|------|------|
| | | | | | (μeqv·l ⁻¹) | | | | | | | | | | |
| 2001 | WET | 1,520 | 4.54 | 19.3 | 28.8 | 10.5 | 3.3 | 5.7 | 2.8 | 0.50 | 9.0 | 41.8 | NA | 26.8 | 29.9 |
| | BP | 1,599 | NA | 18.9 | NA | 15.4 | 4.0 | 8.7 | 5.1 | NA | 11.6 | 46.6 | 0.1 | 26.9 | 30.8 |
| | TF-FS | 1,509 | NA | 25.5 | NA | 28.5 | 9.8 | 11.2 | 17.0 | NA | 14.7 | 70.1 | 0.1 | 35.0 | 33.8 |
| | TF-FD | 1,747 | NA | 28.8 | NA | 39.9 | 16.2 | 15.3 | 34.4 | NA | 21.3 | 98.8 | 0.3 | 40.5 | 34.8 |
| 2002 | WET | 1,275 | 4.60 | 18.8 | 25.1 | 14.0 | 2.5 | 4.3 | 1.8 | 1.1 | 8.2 | 45.6 | NA | 27.9 | 33.8 |
| | BP | 1,319 | 4.84 | 16.3 | 14.5 | 25.6 | 6.4 | 7.8 | 4.8 | 1.0 | 10.0 | 46.0 | 0.1 | 26.5 | 35.1 |
| | TF-FS | 1,218 | 4.76 | 25.9 | 17.4 | 43.3 | 17.1 | 11.0 | 25.2 | 2.0 | 16.6 | 81.6 | 0.1 | 36.2 | 33.7 |
| | TF-FD | 1,389 | 4.88 | 28.6 | 13.1 | 58.8 | 24.6 | 14.8 | 48.2 | 4.6 | 29.0 | 110.3 | 0.2 | 46.4 | 32.6 |
| 2003 | WET | 1,050 | 4.61 | 21.9 | 24.5 | 11.7 | 3.5 | 7.4 | 2.0 | 2.3 | 13.5 | 49.9 | NA | 36.4 | 48.0 |
| | BP | 1,014 | 4.78 | 19.4 | 16.6 | 27.4 | 8.4 | 9.3 | 8.8 | 1.6 | 18.5 | 56.8 | 0.3 | 37.2 | 36.6 |
| | TF-FS | 894 | 4.70 | 31.7 | 20.1 | 70.1 | 25.6 | 15.7 | 37.4 | 2.8 | 25.0 | 106.8 | 0.2 | 54.3 | 46.6 |
| | TF-FD | 1,001 | 4.64 | 40.2 | 22.8 | 78.5 | 39.4 | 18.3 | 67.7 | 3.9 | 36.3 | 150.7 | 0.2 | 62.5 | 47.9 |
| 2004 | WET | 1,214 | 4.52 | 20.1 | 30.2 | 7.8 | 3.3 | 8.0 | 2.3 | 1.4 | 13.8 | 38.6 | NA | 29.3 | 28.9 |
| | BP | 1,180 | 4.86 | 18.2 | 13.7 | 39.5 | 4.8 | 15.2 | 8.2 | 2.9 | 22.9 | 50.9 | 0.2 | 32.1 | 37.2 |
| | TF-FS | 1,093 | 4.81 | 29.9 | 15.4 | 60.3 | 25.2 | 19.5 | 35.5 | 5.1 | 33.9 | 100.4 | 0.2 | 53.9 | 43.0 |
| | TF-FD | 1,220 | 4.83 | 29.7 | 15.0 | 63.5 | 23.0 | 16.3 | 42.3 | 3.4 | 30.2 | 105.8 | 0.1 | 51.2 | 38.8 |
| 2005 | WET | 1,582 | 4.62 | 17.6 | 24.0 | 8.3 | 2.8 | 7.2 | 1.8 | 1.1 | 10.2 | 35.4 | NA | 27.3 | 31.4 |
| | BP | 1,616 | 4.95 | 16.7 | 11.3 | 38.0 | 9.5 | 11.8 | 9.0 | 3.3 | 17.4 | 45.4 | 0.2 | 26.7 | 32.4 |
| | TF-FS | 1,536 | 4.86 | 26.4 | 13.7 | 71.3 | 26.8 | 13.1 | 29.5 | 2.4 | 21.4 | 82.5 | 0.1 | 42.7 | 39.8 |
| | TF-FD | 1,643 | 4.77 | 29.8 | 16.9 | 72.1 | 29.5 | 14.0 | 37.1 | 3.1 | 24.4 | 99.1 | 0.1 | 44.0 | 45.5 |

ary (about 121 mm on average). April and October were low in precipitation with monthly totals of 69 and 79 mm, respectively. The greatest variability in monthly totals was recorded for December during the studied period (coefficient of variation 114%, Fig. 2).

In 2001, 2002 and 2005 the amounts of water collected in the bulk open field collector were higher than the amounts measured in the automatic wet-

only precipitation sampler, in 2003 and 2004 the bulk amounts were lower with the difference being within 2–5% in all cases (Fig. 3 and Table 3).

Throughfall amounts were different for the sparse (FS) and dense (FD) plot (Fig. 3 and Table 3). Rainfall interception amounted to almost 100 mm (7.5%) of precipitation per year on the sparse plot but throughfall was higher than the atmospheric precipitation totals on the denser plot in all but one (2003) year.

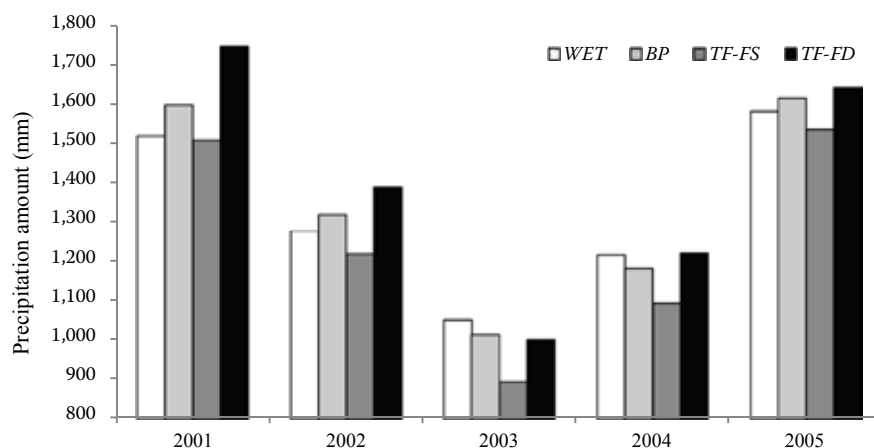


Fig. 3. The amounts of wet (WET), bulk open field (BP) and throughfall (TF) precipitation in mm

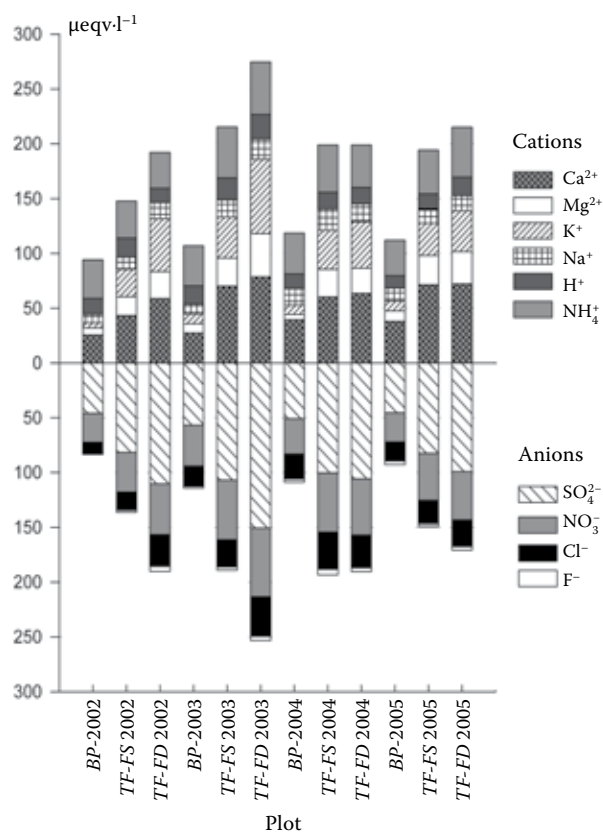


Fig. 4. Volume-weighted mean concentrations of cations and anions in bulk precipitation (*BP*) and throughfall on the sparser (*TF-FS*) and denser (*TF-FD*) plots at Bílý Kříž during 2002–2005

Chemical composition of bulk precipitation and throughfall

The *BP* precipitation (Table 3) was slightly acid: pH 4.8–4.9 and its mean annual electrical conductivity as an overall measure of dissolved ion amounts was 16–19 $\mu\text{S}\cdot\text{cm}^{-1}$. The highest concentrations in *BP* precipitation were SO_4^{2-} , NO_3^- and NH_4^+ (Fig. 4). Among anions concentrations were decreasing in the sequence: $\text{SO}_4^{2-} > \text{NO}_3^- > \text{Cl}^-$, among cations dominated NH_4^+ in 2001–2003 followed by Ca^{2+} , in 2004–2005 the prevailing cation was Ca^{2+} followed by NH_4^+ (Fig. 4, Table 3). Concentrations were higher in throughfall than in bulk open field precipitation for almost all elements (most markedly for K^+ and Mg^{2+}), due to the enrichment of precipitation during the passage through the crowns, except for NH_4^+ in 2002 on both plots and for H^+ in 2002 on the denser plot (Table 3, Fig. 4). For example, enrichment factor for K^+ on the denser plot was 4.1–10.0, on the sparser plot 3.3–5.3, enrichment factor for Mg^{2+} on the denser plot was 3.1–4.8, on the sparser plot 2.5–5.3, while enrichment factor for H^+ was as low as 0.9–1.5 on the denser plot and 1.1–1.2 on the sparser plot (as can be calculated from data in Table 3). A comparison of element concentrations in biweekly samples of bulk precipitation on the open plot and throughfall precipitation on the sparse (*FS*) and dense (*FD*) plots fluxes using the

Table 4. Volume-weighted means of ion concentrations and conductivity in bulk precipitation in the open area (*BP*) and in throughfall on the sparse (*TF-FS*) and dense (*TF-FD*) plots at Bílý Kříž for the studied five years 2001–2005

| Parameter | <i>BP</i> | <i>TF-FS</i> | <i>TF-FD</i> | <i>n</i> |
|---|--------------------|--------------------|--------------------|----------|
| pH | 4.86 | 4.79 | 4.78 | |
| H^+ ($\mu\text{eq}\cdot\text{l}^{-1}$) | 13.70 ^a | 16.22 ^a | 16.58 ^a | 72 |
| Conductivity ($\mu\text{S}\cdot\text{cm}^{-1}$) | 17.82 ^a | 27.45 ^b | 30.79 ^c | 95 |
| C-DOC | 1.64 ^a | 4.26 ^b | 5.60 ^c | 95 |
| Ca^{2+} | 0.58 ^a | 1.07 ^b | 1.22 ^c | 88 |
| Mg^{2+} | 0.08 ^a | 0.25 ^b | 0.31 ^c | 88 |
| Na^+ | 0.24 ^a | 0.32 ^b | 0.36 ^c | 88 |
| K^+ | 0.28 ^a | 1.09 ^b | 1.72 ^c | 88 |
| F^- ($\text{mg}\cdot\text{l}^{-1}$) | 0.03 ^a | 0.04 ^b | 0.05 ^c | 72 |
| Cl^- | 0.56 ^a | 0.76 ^b | 0.97 ^c | 89 |
| SO_4^{2-} | 2.33 ^a | 4.13 ^b | 5.27 ^c | 89 |
| NO_2^- | 0.01 ^a | 0.01 ^a | 0.01 ^a | 89 |
| NO_3^- | 1.81 ^a | 2.68 ^b | 2.94 ^b | 89 |
| NH_4^+ | 0.61 ^a | 0.70 ^b | 0.71 ^b | 88 |

n – number of compared pairs for the relevant parameter, values within rows followed by different letters are significantly different ($P < 0.01$) when tested by the Wilcoxon signed-rank test

Table 5. Annual element fluxes wet-only (*WET*) and bulk (*BP*) in the open field, and under the canopy on the *FS* (*TF-FS*) and *FD* (*TF-FD*) plots at Bílý Kříž (kg·ha⁻¹·year⁻¹)

| Year | Collector | C-DOC | H ⁺ | Ca ²⁺ | Mg ²⁺ | Na ⁺ | K ⁺ | F ⁻ | Cl ⁻ | S-SO ₄ ²⁻ | N-NO ₂ ⁻ | N-NO ₃ ⁻ | N-NH ₄ ⁺ | Inorg N |
|------|--------------|-------|----------------|------------------|------------------|-----------------|----------------|----------------|-----------------|---------------------------------|--------------------------------|--------------------------------|--------------------------------|---------|
| 2001 | <i>WET</i> | NA | 0.43 | 3.2 | 0.6 | 2.0 | 1.6 | 0.20 | 4.9 | 10.2 | NA | 5.7 | 6.3 | 12.0 |
| | <i>BP</i> | 21.3 | NA | 4.9 | 0.8 | 3.2 | 3.2 | NA | 6.6 | 11.9 | 0.02 | 6.0 | 6.9 | 12.9 |
| | <i>TF-FS</i> | 42.4 | NA | 8.6 | 1.8 | 3.9 | 10.0 | NA | 7.9 | 17.0 | 0.02 | 7.4 | 7.1 | 14.6 |
| | <i>TF-FD</i> | 79.5 | NA | 14.0 | 3.4 | 6.2 | 23.5 | NA | 13.2 | 27.7 | 0.08 | 9.9 | 8.5 | 18.5 |
| 2002 | <i>WET</i> | NA | 0.32 | 3.6 | 0.4 | 1.3 | 0.9 | 0.20 | 3.7 | 9.3 | NA | 5.0 | 6.0 | 11.0 |
| | <i>BP</i> | 19.9 | 0.19 | 6.8 | 1.0 | 2.4 | 2.5 | 0.26 | 4.7 | 9.7 | 0.02 | 4.9 | 6.5 | 11.4 |
| | <i>TF-FS</i> | 49.2 | 0.21 | 10.6 | 2.5 | 3.1 | 12.0 | 0.46 | 7.2 | 15.9 | 0.02 | 6.2 | 5.8 | 11.9 |
| | <i>TF-FD</i> | 78.6 | 0.18 | 16.4 | 4.2 | 4.7 | 26.2 | 1.20 | 14.3 | 24.6 | 0.03 | 9.0 | 6.3 | 15.4 |
| 2003 | <i>WET</i> | NA | 0.26 | 2.5 | 0.5 | 1.8 | 0.8 | 0.45 | 5.0 | 8.4 | NA | 5.4 | 7.1 | 12.4 |
| | <i>BP</i> | 20.8 | 0.17 | 5.6 | 1.0 | 2.2 | 3.5 | 0.31 | 6.7 | 9.2 | 0.04 | 5.3 | 5.2 | 10.5 |
| | <i>TF-FS</i> | 41.7 | 0.18 | 12.6 | 2.8 | 3.2 | 13.1 | 0.48 | 7.9 | 15.3 | 0.03 | 6.8 | 5.8 | 12.7 |
| | <i>TF-FD</i> | 71.0 | 0.23 | 15.8 | 4.8 | 4.2 | 26.5 | 0.73 | 12.9 | 24.2 | 0.04 | 8.8 | 6.7 | 15.5 |
| 2004 | <i>WET</i> | NA | 0.37 | 1.9 | 0.5 | 2.2 | 1.1 | 0.32 | 5.9 | 7.5 | NA | 5.0 | 4.9 | 9.9 |
| | <i>BP</i> | 21.0 | 0.16 | 9.4 | 0.7 | 4.1 | 3.8 | 0.65 | 9.6 | 9.6 | 0.02 | 5.3 | 6.2 | 11.5 |
| | <i>TF-FS</i> | 61.9 | 0.17 | 13.2 | 3.3 | 4.9 | 15.2 | 1.05 | 13.1 | 17.6 | 0.02 | 8.3 | 6.6 | 14.9 |
| | <i>TF-FD</i> | 68.0 | 0.18 | 15.5 | 3.4 | 4.6 | 20.2 | 0.78 | 13.1 | 20.7 | 0.02 | 8.7 | 6.6 | 15.4 |
| 2005 | <i>WET</i> | NA | 0.38 | 2.6 | 0.5 | 2.6 | 1.1 | 0.33 | 5.7 | 9.0 | NA | 6.0 | 6.9 | 13.0 |
| | <i>BP</i> | 27.4 | 0.18 | 12.3 | 1.9 | 4.4 | 5.7 | 1.02 | 10.0 | 11.8 | 0.03 | 6.0 | 7.3 | 13.4 |
| | <i>TF-FS</i> | 70.8 | 0.21 | 21.9 | 5.0 | 4.6 | 17.7 | 0.70 | 11.7 | 20.3 | 0.02 | 9.2 | 8.6 | 17.8 |
| | <i>TF-FD</i> | 95.1 | 0.28 | 23.8 | 5.9 | 5.3 | 23.8 | 0.98 | 14.2 | 26.1 | 0.03 | 10.1 | 10.5 | 20.6 |

NA – data not available, WET values data (CHMI 2008)

Wilcoxon test revealed highly significant differences for the base cations (Ca²⁺, Mg²⁺, Na⁺, K⁺), for dissolved organic carbon and for the anions SO₄²⁻, F⁻ and Cl⁻ (Table 4). Insignificant differences among all plots (FD, FS and the open area) were found for H⁺ and for NO₂⁻ concentrations. The differences between concentrations of NH₄⁺ and NO₃⁻ on *FD* and *FS* plots were not significant either.

Element fluxes

The calculated element fluxes are shown in Table 5. The values of wet-only deposition fluxes were lower than the bulk open field deposition fluxes with a few exceptions: namely H⁺ in 2002–2005 and NO₃⁻ fluxes in 2002 and 2003, NH₄⁺ flux in 2003 and F⁻ flux in 2003.

Table 6. Relative increase (%) in element fluxes on the dense plot (*FD*) in comparison with the sparse plot (*FS*) at Bílý Kříž

| Period | H ⁺ | C-DOC | Ca ²⁺ | Mg ²⁺ | Na ⁺ | K ⁺ | F ⁻ | Cl ⁻ | S-SO ₄ ²⁻ | N-NO ₃ ⁻ | N-NH ₄ ⁺ | Inorg N |
|-----------|----------------|-------|------------------|------------------|-----------------|----------------|----------------|-----------------|---------------------------------|--------------------------------|--------------------------------|---------|
| 2001 | NA | 87.5 | 62.1 | 91.2 | 58.1 | 134.4 | NA | 67.7 | 63.2 | 33.9 | 19.1 | 27.0 |
| 2002 | 14.2 | 59.7 | 54.8 | 64.7 | 54.3 | 117.9 | 164.1 | 98.8 | 54.3 | 46.3 | 10.2 | 28.9 |
| 2003 | 27.2 | 70.2 | 25.4 | 72.4 | 30.4 | 103.0 | 53.2 | 62.6 | 57.9 | 28.7 | 15.1 | 22.4 |
| 2004 | 8.7 | 9.8 | 17.5 | 2.0 | -6.4 | 33.0 | -25.5 | -0.7 | 17.7 | 5.9 | 0.6 | 3.5 |
| 2005 | 32.6 | 34.4 | 8.2 | 18.0 | 14.2 | 34.5 | 40.0 | 22.1 | 28.6 | 10.1 | 22.1 | 15.9 |
| 2001–2005 | 13.6 | 52.3 | 33.6 | 49.6 | 30.1 | 84.6 | 57.9 | 50.1 | 44.3 | 25.0 | 13.4 | 19.6 |

NA – data not available, values calculated for individual elements as: $RI_x = \left(\frac{\sum_{i=1}^n (TF(FD)_x / TF(FS)_x)}{n} \right) \times 100 - 100\%$

Table 7. Total deposition fluxes (*TD*) and canopy exchange fluxes (*CU* – canopy uptake, *Cle* – canopy leaching) for the *FS* and *FD* plots at Bílý Kříž (kg·ha⁻¹·year⁻¹)

| Year | Plot | H ⁺ | | Ca ²⁺ | | Mg ²⁺ | | Na ⁺ | | K ⁺ | | Cl ⁻ | | S-SO ₄ ²⁻ | | N-NO ₃ ⁻ | | N-NH ₄ ⁺ | | Inorg N | |
|---------------|-----------|----------------|-----------|------------------|------------|------------------|------------|-----------------|------------|----------------|------------|-----------------|------------|---------------------------------|------------|--------------------------------|------------|--------------------------------|------------|-----------|-----------|
| | | <i>TD</i> | <i>CU</i> | <i>TD</i> | <i>Cle</i> | <i>TD</i> | <i>Cle</i> | <i>TD</i> | <i>Cle</i> | <i>TD</i> | <i>Cle</i> | <i>TD</i> | <i>Cle</i> | <i>TD</i> | <i>Cle</i> | <i>TD</i> | <i>Cle</i> | <i>TD</i> | <i>Cle</i> | <i>TD</i> | <i>CU</i> |
| 2001 | <i>FS</i> | | | 7.7 | 3.4 | 1.2 | 1.1 | 5.0 | 5.0 | 5.0 | 7.9 | 10.1 | 10.1 | 21.9 | 21.9 | NA | NA | NA | NA | NA | NA |
| | <i>FD</i> | | | 12.2 | 5.8 | 1.9 | 2.5 | 8.0 | 8.0 | 7.9 | 22.4 | 17.0 | 17.0 | 35.7 | 35.7 | NA | NA | NA | NA | NA | NA |
| 2002 | <i>FS</i> | 0.56 | 0.29 | 11.3 | 2.2 | 1.7 | 1.5 | 4.0 | 4.0 | 4.1 | 11.3 | 9.2 | 9.2 | 20.5 | 20.5 | 8.2 | 0.2 | 1.3 | 1.3 | 16.8 | 1.5 |
| | <i>FD</i> | 0.71 | 0.47 | 17.5 | 3.5 | 2.6 | 2.7 | 6.1 | 6.1 | 6.3 | 27.3 | 18.3 | 18.3 | 31.6 | 31.6 | 12.2 | 0.6 | 2.7 | 2.7 | 23.1 | 3.4 |
| 2003 | <i>FS</i> | 0.54 | 0.31 | 10.7 | 5.4 | 2.0 | 1.6 | 4.1 | 4.1 | 6.6 | 10.1 | 10.1 | 10.1 | 19.6 | 19.6 | 9.0 | 0.3 | 1.7 | 1.7 | 18.1 | 2.0 |
| | <i>FD</i> | 0.87 | 0.58 | 13.9 | 6.3 | 2.6 | 3.6 | 5.4 | 5.4 | 8.7 | 25.2 | 16.5 | 16.5 | 30.9 | 30.9 | 11.8 | 0.6 | 2.8 | 2.8 | 23.2 | 3.4 |
| 2004 | <i>FS</i> | 0.51 | 0.30 | 14.2 | 2.7 | 1.0 | 3.2 | 6.2 | 6.2 | 5.7 | 13.6 | 16.7 | 16.7 | 22.4 | 22.4 | 10.9 | 0.4 | 1.9 | 1.9 | 21.2 | 2.3 |
| | <i>FD</i> | 0.70 | 0.47 | 13.3 | 6.5 | 1.0 | 3.4 | 5.8 | 5.8 | 5.4 | 20.4 | 16.6 | 16.6 | 26.4 | 26.4 | 11.8 | 0.6 | 2.8 | 2.8 | 23.0 | 3.4 |
| 2005 | <i>FS</i> | 0.74 | 0.47 | 16.5 | 11.3 | 2.5 | 3.8 | 5.9 | 5.9 | 7.6 | 14.8 | 14.8 | 14.8 | 25.8 | 25.8 | 12.2 | 0.6 | 3.2 | 3.2 | 26.3 | 3.8 |
| | <i>FD</i> | 0.92 | 0.56 | 18.9 | 11.2 | 2.9 | 4.6 | 6.7 | 6.7 | 8.7 | 21.5 | 18.1 | 18.1 | 33.1 | 33.1 | 13.4 | 0.6 | 3.5 | 3.5 | 30.2 | 4.1 |
| <i>RI</i> (%) | | 36.9 | 56.6 | 30.1 | 57.5 | 30.1 | 70.6 | 30.1 | 30.1 | 30.1 | 113.9 | 50.1 | 50.1 | 44.3 | 44.3 | 24.6 | 80.0 | 58.5 | 58.5 | 22.0 | 61.9 |

NA – data not available, \overline{RI}_x – mean relative increase of flux values on the denser plot calculated for element *x* for the number of studied years *n* as:

$$\overline{RI}_x = \left(\frac{\sum_{i=1}^n (TF(FD)x_i / TF(FS)x_i)}{n} \times 100 \right) - 100\%$$

Throughfall fluxes were strongly influenced by stand density. The differences are most pronounced for K⁺. The lowest differences can be seen in nitrogen compounds and H⁺. The differences in throughfall fluxes between the *FS* and *FD* plot were not the same in individual years (Table 6), because both stands went through changes (growth, crown developments, decrease of the trees number on both plots). The greatest relative increase of annual deposition on the denser plot was observed for the first three years of the research period when the differences between the stand densities were 620 to 720 trees·ha⁻¹. The values of dry deposition fluxes, total deposition fluxes and canopy exchange fluxes calculated according to the Canopy Budget Model for each sampling year are given in Tables 7 and 8.

Total deposition fluxes, dry deposition fluxes and canopy exchange fluxes were higher on the denser plot for all elements in 2001–2003 (Tables 7 and 8) when the difference in stand densities between the two plots was 620–720 trees·ha⁻¹ (Table 2) and the difference in leaf area indices was 3.20–4.83. At the beginning of 2004 both stands were damaged by wind and snow and many trees had to be removed. The difference between the stand densities on studied plots decreased. Results in Tables 7 and 8 reflect the situation: the estimates of calcium, magnesium, sodium, potassium and chloride total and dry depositions were almost the same for both forested plots in 2004. The share of dry deposition in total element fluxes was more than 50% for all elements and was always higher on the denser (*FD*) plot (Table 8).

Empirical critical loads of nitrogen and its exceedances

In the Manual on methodologies and criteria for modelling and mapping critical loads (SPRANGER et al. 2004) the empirical critical load of 10–20 kg N·ha⁻¹·yr⁻¹ for nitrogen deposition is considered to be the quite reliable value for temperate forest ecosystems. At Bílý Kříž the nitrogen critical load lies closer to the lower value of this range. According to ZAPLETAL (2006 and personal communication) the critical load for nitrogen at this site was 11.2 kg N·ha⁻¹·yr⁻¹ (calculated using the simple mass balance method). This threshold was clearly exceeded on both plots in the years 2001–2005 (Table 5). Estimated total nitrogen deposition (Table 7) was up to

Table 8. Dry deposition fluxes (DD in $\text{kg}\cdot\text{ha}^{-1}\cdot\text{year}^{-1}$) for the FS and FD plots and the share of DD in TD (calculated as $100 DD/TD$ in %) on the FD and FS plots at Bílý Kříž

| Year | Plot | Ca^{2+} | | Mg^{2+} | | Na^+ | | K^+ | | Cl^- | | S-SO_4^{2-} | | N-NO_3^- | | N-NH_4^+ | | Inorg N | |
|----------|------|------------------|-------|------------------|-------|---------------|-------|--------------|-------|---------------|-------|----------------------|-------|-------------------|-------|-------------------|-------|---------|-------|
| | | DD | share | DD | share | DD | share | DD | share | DD | share | DD | share | DD | share | DD | share | DD | share |
| 2001 | FS | 4.5 | 58.6 | 0.6 | 50.2 | 3.0 | 60.2 | 3.4 | 68.1 | 5.2 | 51.7 | 11.7 | 53.5 | NA | NA | NA | NA | NA | NA |
| | FD | 9.0 | 73.8 | 1.3 | 68.5 | 6.0 | 74.9 | 6.3 | 79.8 | 12.1 | 71.2 | 25.5 | 71.5 | NA | NA | NA | NA | NA | NA |
| 2002 | FS | 7.7 | 68.3 | 1.3 | 76.7 | 2.7 | 67.1 | 3.2 | 78.1 | 5.5 | 59.9 | 11.1 | 54.5 | 3.2 | 38.8 | 2.7 | 31.2 | 5.9 | 34.8 |
| | FD | 13.9 | 79.4 | 2.2 | 84.9 | 4.8 | 78.7 | 5.4 | 85.8 | 14.6 | 79.8 | 22.3 | 70.5 | 7.2 | 59.2 | 4.9 | 45.0 | 12.1 | 52.5 |
| 2003 | FS | 8.2 | 76.9 | 1.5 | 77.2 | 2.4 | 56.9 | 5.8 | 87.7 | 5.1 | 50.3 | 11.2 | 57.1 | 3.7 | 40.7 | 2.1 | 22.7 | 5.7 | 31.6 |
| | FD | 11.4 | 82.3 | 2.1 | 82.5 | 3.6 | 67.0 | 7.9 | 90.5 | 11.4 | 69.4 | 22.5 | 72.9 | 6.5 | 54.7 | 4.3 | 38.1 | 10.8 | 46.5 |
| 2004 | FS | 12.3 | 86.6 | 0.6 | 53.3 | 4.0 | 64.3 | 4.6 | 80.7 | 10.8 | 64.6 | 14.9 | 66.5 | 5.9 | 54.3 | 5.4 | 52.4 | 11.3 | 53.4 |
| | FD | 11.4 | 85.7 | 0.5 | 50.1 | 3.6 | 61.9 | 4.3 | 79.4 | 10.7 | 64.4 | 18.8 | 71.5 | 6.8 | 57.5 | 6.3 | 56.3 | 13.1 | 56.9 |
| 2005 | FS | 13.9 | 84.0 | 2.0 | 78.9 | 3.3 | 55.3 | 6.5 | 85.6 | 9.0 | 61.1 | 16.8 | 65.2 | 6.2 | 50.6 | 7.1 | 50.6 | 13.3 | 50.6 |
| | FD | 16.2 | 86.0 | 2.3 | 81.5 | 4.1 | 60.9 | 7.6 | 87.4 | 12.3 | 68.2 | 24.1 | 72.9 | 7.4 | 55.0 | 9.8 | 58.5 | 17.2 | 56.9 |
| Avg FS | | 9.3 | 74.9 | 1.2 | 67.3 | 3.1 | 60.8 | 4.7 | 80.0 | 7.1 | 57.5 | 13.1 | 59.3 | 4.7 | 46.1 | 4.3 | 39.2 | 9.1 | 42.6 |
| Avg FD | | 12.4 | 81.4 | 1.7 | 73.5 | 4.4 | 68.6 | 6.3 | 84.6 | 12.2 | 70.6 | 22.7 | 71.9 | 7.0 | 56.6 | 6.3 | 49.5 | 13.3 | 53.2 |

dry deposition fluxes were assessed according to the chapter Canopy Budget Model, Avg FS and Avg FD are five-year averages (or four-year averages in the case of nitrogen compounds) calculated for FS and FD plot, respectively; NA – data not available

28–37% higher on the denser plot in 2002 and 2003 when the differences in stand densities were 680 and $620 \text{ trees}\cdot\text{ha}^{-1}$. That means the soil under the stand with higher density was more endangered by elevated nitrogen input.

DISCUSSION

The pattern of interannual variations in precipitation is usual for the site (TOLASZ 2007).

The fact that throughfall volumes on the denser plot were higher than the atmospheric precipitation totals in four out of the five monitored years could be explained by the contribution of cloud or fog water collected by forest canopies. It is known that approximately 10 to 25% of annual precipitation is lost by canopy interception, depending on evaporation power of the air, storm characteristics, and vegetation (CHANG 2003). Nevertheless, fog condensed in forest canopy can contribute significant amounts of water to the ecosystem budget especially in the areas with high annual numbers of foggy days (HUTLEY et al. 1997; ZIMMERMANN et al. 1999; DEFELICE 2002; LIU et al. 2004; BRUIJNZEEL et al. 2005). The studied locality belongs to the places with the highest average annual number of foggy days (more than 150 according to TOLASZ et al. 2007). It seems that in the denser stand, canopy condensation was able to refill the amount of water lost by canopy interception in all studied years except the driest one – 2003. The difference between

interceptions on the denser and sparser plot was caused by different microclimatic conditions in the two forest stands (MATEJKA et al. 2004).

The composition of bulk precipitation is influenced by the climate, by the long distance transport of air masses, by local weather conditions, local emission sources and by the amount of aerosols in the air. Values measured at the Bílý Kříž site can be compared with values reported for other parts of the Czech Republic. During the studied period 2001–2005 the amount and chemical composition of bulk open field and throughfall precipitation were monitored on several ICP plots in the Czech Republic, however on some of these plots the monitoring period did not include all five years. The data are shown in yearbooks published by BOHÁČOVÁ et al. (2003, 2004, 2007). The prevalent cation at most sites was also NH_4^+ , however, on Březka, Všetec and Želivka plots Ca^{2+} concentrations were higher than NH_4^+ concentrations. The prevalent anion in BP precipitation on most Czech ICP plots listed in the above-mentioned yearbooks was SO_4^{2-} or NO_3^- (at Želivka, Medlovice and Březka). Sulphate concentrations in BP precipitation at Bílý Kříž were rather high in comparison with those determined on ICP plots throughout the Czech Republic. The only site with higher SO_4^{2-} concentrations during the whole period was the plot at Medlovice. The greatest similarity in the composition of BP precipitation can be seen between Bílý Kříž and Mísečky. These two plots lay at a similar altitude and also the amounts of precipitation per year were of comparable volume. ZAPLETAL et al.

(2007) described the composition of BP and TF at the Červenohorské sedlo site (50°07'N, 17°09'E, 1,013 m a.s.l.) for the period 1999–2002. The prevailing cation in BP was also NH_4^+ followed by Ca^{2+} . The main anions were SO_4^{2-} and NO_3^- , but the SO_4^{2-} concentrations in 2001 and 2002 were more than twice higher when compared with the SO_4^{2-} concentrations in BP at Bílý Kříž during the same years. The precipitation totals were comparable at both sites. While comparing the values measured at Bílý Kříž with concentrations of elements in bulk precipitation measured at other forest sites in Europe, the difference in concentrations of Na^+ and Cl^- ions between sites located near the sea and those in inland is evident. Localities influenced by the sea, e.g. Monte Rufeno in Italy, Melle in the north of Belgium, Heiliges Meer in Northwest Germany or Vilsandi in Estonia, have markedly higher concentrations of Na^+ and Cl^- in bulk precipitation (STAELENS et al. 2005; HERRMANN et al. 2006; PAJUSTE et al. 2006; BALESTRINI et al. 2007) than the values measured at Bílý Kříž (i.e. 10.0–22.9 $\mu\text{ekv Cl}^-\cdot\text{l}^{-1}$ and 7.8–15.2 $\mu\text{ekv Na}^+\cdot\text{l}^{-1}$). The mean Na^+ to Cl^- ratio in wet-only precipitation was 0.60 (0.53–0.71 in individual years 2001–2005), in bulk precipitation it was 0.68 (0.50–0.78 in individual years). This could indicate Cl^- deposition from anthropogenic sources because the theoretical value of the ratio for sea-water is 0.86. The content of ions in bulk precipitation is higher than in most areas in the Swiss Alps (THIMONIER et al. 2005) except Novaggio in the Southern Alps where the concentrations were very similar for all ions. Comparable values of ion concentrations in bulk precipitation were published also for Kreisbach in Lower Austria (BERGER et al. 2008).

The comparison of annual concentrations of studied elements on the denser and sparser plot conforms the Canopy Budget Model where canopy uptake of inorganic nitrogen compounds is considered. The uptake fluxes of nitrogen compounds in denser stand are higher than in sparse stand and are able to balance the increase in dry deposition flux caused by denser canopy. The same holds for protons which are accepted by crowns in exchange for basic elements – Eq. (13). BÄUMLER and ZECH (1997) also studied differences in throughfall element concentrations for two mixed mountain forest stands in Tegernsee Alps (Germany) with different tree densities (the difference in basal area was 40%). They found highly significant differences in conductivity, total organic carbon, Ca^{2+} , K^+ , Mg^{2+} , SO_4^{2-} , and NH_4^+ and NO_3^- concentrations. The differences in H^+ , Na^+ and HPO_4^{2-} were not significant. The difference between BÄUMLER and ZECH's (1997) and our findings for N-NH_4^+ and N-NO_3^-

concentrations in sparse and dense stand could be explained by the difference in annual input of nitrogen. Substantially higher annual input of NO_3^- and NH_4^+ with bulk precipitation was measured at the Bäumler's site: 59.4 kg NO_3^- per ha and year and 15.1 kg NH_4^+ per ha and year nitrate and ammonium (values measured in the Beskids were 24.4 kg NO_3^- and 8.3 kg NH_4^+ per ha and year).

The values of wet-only deposition fluxes should always be lower than bulk open field deposition fluxes, because the samples from wet-only samplers do not include the contribution of dry deposition caught by sampler vessels. The wet-only fluxes of H^+ at Bílý Kříž were higher than the bulk open field fluxes of H^+ but it is necessary to bear in mind that the amount of free H^+ (or rather H_3O^+ ions measured as pH) in any solution is given by the amount and kind of acidic and base solution components and their balance. This balance was shifted towards lower acidity in the bulk precipitation samples from Bílý Kříž by buffering compounds contained in samples. STAELENS et al. (2005) studied bulk and wet-only deposition at two adjacent sites in Melle (Belgium) and also found the bulk H^+ deposition significantly lower than the wet-only deposition. Higher flux of H^+ with wet-only precipitation in comparison with bulk precipitation was measured also by BALESTRINI et al. (2007) at two (out of five) studied sites in Italy. In the case of F^- in 2003, the higher value of wet-only deposition in our study is difficult to explain. Perhaps the higher uncertainty related to the measurement of very low F^- concentrations could cause this result. As concerns NO_3^- in 2002–2003 and NH_4^+ in 2003, the explanation of higher wet-only fluxes could lie in the fact that the sampling interval was one week for wet-only sampler (run by the Czech Hydro-meteorological Institute) and a fortnight or a month (during the winter) for all other samplers, what can play a certain role in the case of nitrogen compounds which are assimilated by microorganisms. Concentrations of nitrogen compounds in water samples were measured in filtered, i.e. microbial-free, samples. To assess *TD*, *DD*, and *CE* of individual elements Canopy Budget Model was applied which is based on several simplifying assumptions. The reliability of the presented results depends on the validity of these assumptions. The most important is the negligible interaction of Na^+ with the forest canopy. STAELENS et al. (2007) reported Na^+ leaching from beech canopy during a short period of leaf emergence and BALESTRINI et al. (2007) suggested possible leaching of Na^+ from the *Quercus cerris* tissue. The possibility of Na^+ leaching from coniferous canopy was (according to my knowledge) described only by REINERS and OLSON (1984) in balsam fir. On

the other hand, studies published by RAGSDALE et al. (1992), HULTBERG and FERM (1995), STACHURSKI and ZIMKA (2000) justify the employment of Na^+ as the tracer ion which is neither absorbed nor leached from the canopy. In the calculations, I considered also the interactions of Cl^- and sulphur compounds with canopy to be negligible. It does not mean that the uptake of these elements is absent but we can suppose that the uptake is balanced by leaching from the canopy (LINDBERG, LOWETT 1992). The greatest uncertainties in calculations arise when fluxes of nitrogen compounds are evaluated as was shown by VAN LEUWEN et al. (2000). Still the Canopy Budget Model is a useful tool and its implementation enables to derive some conclusions from the presented results. *DD*, *Cle*, and *CU* of elements are positively influenced by stand density. In the case of inorganic nitrogen compounds the increase in *DD* in the denser stand was only partially compensated by increased *CU* at the studied site.

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