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Pollution control enhanced spruce growth in the "Black Triangle" near the Czech–Polish border



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HIGHLIGHTS

GRAPHICAL ABSTRACT

- Long-term growth changes of Norway spruce are evaluated for the "Black Triangle."
- The ring width variations of Norway spruce reflect May–July temperatures.
- Acid deposition reduced the growthtemperature relationships of Norway spruce.
- This study suggests a complex interplay of multiple factors on forest decline.
- Our results prove a recovery of forest growth in the 1990s.

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ABSTRACT

Norway spruce (*Picea abies* (L.) Karst.) stands in certain areas of Central Europe have experienced substantial dieback since the 1970s. Understanding the reasons for this decline and reexamining the response of forests to acid deposition reduction remains challenging because of a lack of long and well-replicated tree-ring width chronologies. Here, spruce from a subalpine area heavily affected by acid deposition (from both sulfur and nitrogen compounds) is evaluated. Tree-ring width measurements from 98 trees between 1000 and 1350 m above sea level (a.s.l.) reflected significant May–July temperature signals. Since the 1970s, acid deposition has reduced the growth–climate relationship. Efficient pollution control together with a warmer but not drier climate most likely caused the increased growth of spruce stands in this region, the so-called "Black Triangle," in the 1990s.

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1. Introduction

In the second half of the 20th century, acidic air pollution was a serious concern across most of Europe (Stern, 2005). The increase in global emissions, mainly sulfur dioxide (SO₂) and nitrogen oxides (NO_x), was unprecedented after World War II due to economic expansion (Grübler, 2002; Smith et al., 2011). The SO₂ and NO_x emissions in the Czech Republic increased sharply after approximately 1950 (Dignon and Hameed, 1989; Smil, 1990) until the 1980s (Kopáček and Veselý, 2005). The emission loads were highest in the area known as the "Black Triangle," that is, the Czech–Polish–German border region (Grübler, 2002). This region was characterized by large coal resources, numerous power plants (Kopáček and Veselý, 2005), and topography that favored the occurrence of prolonged inversion events. In view of these ecological impacts, the United Nations Environment Programme (UNEP) officially designated this area as an "ecological disaster zone" (Grübler, 2002).

Desulfurization of brown coal power plants as well as the political and economic changes in the early 1990s caused anthropogenic emissions to reduce considerably (Kopáček and Veselý, 2005). According to the Gothenburg Protocol, SO₂ and NO_x emissions in the Czech Republic should have been reduced by 85% and 61%, respectively, in 2010 compared with the 1990 baseline. By 2007, SO₂ emissions had already decreased by 88% and NO_x emissions by 62% (Helliwell et al., 2014); therefore, the target emissions had been successfully surpassed. The reduction in SO₂ emissions in the Czech Republic has been one of the most dramatic examples of pollution reduction in Europe (Vestreng et al., 2007).

However, the high and persistent levels of sulfur dioxide pollution led to extensive forest decline (Vávrová et al., 2009), with most of the damage occurring in high-elevation conifer ecosystems (Vacek et al., 2013; Rydval and Wilson, 2012; Elling et al., 2009). Tree injury was typically observed during the winter periods due to frequent episodes of harsh meteorological conditions and the high SO₂ concentration in the air (Lomský et al., 2012). One of the most common woody species growing in high-elevation areas is Norway spruce (Picea abies (L.) Karst.), which covers an extremely broad ecological spectrum (Hertel and Schöling, 2011), including many ecotones outside its natural range (Tollefsrud et al., 2008). In recent decades, spruce tree-ring width (TRW) reductions and poor crown condition during peak pollution have been reported several times (e.g., Vacek et al., 2013; Rydval and Wilson, 2012; Akselsson et al., 2004; Kroupová, 2002; Kandler and Innes, 1995; Sander et al., 1995). The decline in anthropogenic emissions led to Norway spruce growth recovery. However, the possible explanations for this phenomenon are unclear. These changes have been mostly associated with rapid reductions in SO₂ emissions (e.g., Hauck et al., 2012; Elling et al., 2009), higher N availability (e.g., Laubhann et al., 2009), the combined impact of rapid reductions in atmospheric SO₂ and NO_x, and significantly higher mean temperatures during the growing season (e.g., Bošel'a et al., 2014a). Whether and how forest ecosystems responded to the drastic pollution controls initiated in the early 1990s has not been investigated yet.

Here, TRW measurements are used to evaluate the long-term behavior of Norway spruce from the "Black Triangle," the most polluted part of Central Europe. The aim of this study is to further investigate the forest decline beginning in the 1980s and the subsequent TRW recovery. Our study is primarily motivated by the following two hypotheses: (1) The combined effects of the extremely warm and dry summer in 1976 together with the exceptionally cold winter in 1978/1979 contributed to the decreased growth of Central European Norway spruce from the late 1970s to the early 1980s. (2) The subsequent growth recovery was driven not only by reductions in SO₂ and NO_x emissions starting in the late 1980s but also by the overall effects of increasing temperatures. Our results may prove beneficial in determining the future forest productivity under the predicted global climate change. Such information should be considered further in forest management strategies.

2. Materials and methods

2.1. Study area

During the second half of the last century, the Krkonoše mountain range along the Czech-Polish border was one of the main foci of air pollution in Central Europe (Fig. 1). This region belongs to the Bohemian Massif. The main ridge of the mountains (35 km long) spreads from east to west, forming a watershed between the North Sea and the Baltic Sea. The biogeographic conditions, termed arctic-alpine tundra (Soukupová et al., 1995), are characterized by frequent weather changes and long, extremely cold, and damp winters with abundant snow cover (Hejcman et al., 2006). The lowest mean temperatures are observed in January, and the warmest months are June and August. Annual precipitation totals vary from 1300 to 1450 mm (Table 1). The highest precipitation totals occur in July and the lowest in spring, especially in April, when the highest peaks are still snow covered. A continuous snow cover persists from November to the beginning of May (5-6 months), with a maximum snow depth observed in March or in the beginning of April. The mean snowpack thickness reaches values of approximately 1.8 m (Hejcman et al., 2006). The main ridge is well exposed to wind. The predominant winds in Krkonoše blow from the west or southwest (Kerzelová, 1983).



Fig. 1. (*A*) Location of the study area in Europe and a detailed overview of the study sites in the Krkonoše Mountains. (*B*) Natural distribution of Norway spruce (*Picea abies* (L.) Karst.) (http://www.euforgen.org), together with the core region of sulfur deposition >2500 mg S m⁻² a⁻¹ in 1985 (hatched area reproduced from www.emp.int).

Table 1

Characteristics of the five tree-ring sampling sites (site codes: L, lower altitude below 1100 m a.s.l.; M, middle altitude between 1100 and 1300 m a.s.l.; and U, upper altitude above 1300 m a.s.l.).

Site code	Site	Coordinates	Altitude (m a.s.l.)	Aspect	Slope (°)	Prevailing soil types	Vegetation zone	Edaphic category	Avg. temp. (°C)	Avg. prec. (mm)
1L	Bílá Voda	50° 47′ 175″ N 15° 27′ 314″ E	1009	S	10	Podzolic soil	Beech-Spruce	Lapidosa acidophila	3.9	1401
2M	Mumlavská hora	50° 47′ 56″ N 15° 27′ 53″ E	1185	SW	5	Podzolic soil	Spruce	Paludosa oligotrophica	3.2	1421
3M	Alžbětinka	50° 45′ 34″ N 15° 31′ 15″ E	1192	NW	14	Leptosol, podzolic soil	Spruce	Humilis	3.3	1340
4M	Modrý důl	50° 43′ 13″ N 15° 42′ 25″ E	1237	S	22	Leptosol, podzolic soil	Spruce	Humilis	3.5	1413
5U	Pašerácký chodníček	50° 44′ 25″ N 15° 45′ 56″ E	1317	SW	18	Leptosol, podzolic soil	Spruce	Humilis	2.6	1414

Our study sites cover montane (800–1200-m) and subalpine (1200–1450-m) vegetation zones. The tree line is located approximate-ly 1250–1350 m above sea level (a.s.l.). Compared with timberline trees, tree-line trees, in general, grow in colder environments and experience shorter vegetation periods, leading to overall reduced TRW and even a higher proportion of missing rings (Körner, 1998; Treml et al., 2012). Norway spruce (*P. abies* (L.) Karst.) occupies 82.6% of the area and dominates the even-aged conifer forest at middle to higher elevations. Dwarf mountain pine stands prevail at the subalpine vegetation level, where many relict or endemic species such as cloudberry are also present.

2.2. Tree-ring sampling and chronology development

Sampling sites were selected in the northwestern and northeastern parts of the mountain range. These areas were most threatened (Vacek et al., 2013). The selected sites cover a wide altitudinal range (Table 1 and Fig. 1). Because the between-tree variability within a site is much higher than the within-tree variability around the stem (Bošel'a et al., 2014b), one core per tree was extracted at breast height (1.3 m) using a Pressler borer (Haglof Company Group) with a 5-mm inner diameter. To avoid compression wood, the cores were sampled in a direction parallel to the slope. All samples were measured using a VIAS TimeTable measuring system (measuring length 78 cm; resolution <1/100 mm) devised by SCIEM. The TRW series were measured (an accuracy of 0.01 mm) and synchronized using PAST4 (Knibbe, 2004). The individual TRW series obtained were cross-dated. Missing and false rings were corrected using PAST4 (Knibbe, 2004) and COFECHA (Grissino-Mayer, 2001). The degree of similarity between the TRW series was assessed using correlation coefficients (p < 0.01), the coefficient of agreement (p < 0.05; Eckstein and Bauch, 1969), and an optical comparison of both series, which is crucial for the final dating (Rybníček et al., 2010a). Well-correlated TRW series were used to create composite TRW chronology.

To remove non-climatic, age-related growth trends from the raw TRW series as well as other non-climatic factors (e.g., competition), cubic smoothing splines with 50% frequency cutoff at 100 years were applied (Cook and Peters, 1981) using ARSTAN software (Cook and Krusic, 2005). This standardization method was selected because of its ability to preserve interannual to multi-decadal growth variations (Büntgen et al., 2008). TRW indices were calculated as residuals from estimated growth curves after applying an adaptive power transformation to the raw measurement series (Cook and Peters, 1997). The mean chronologies were calculated using bi-weight robust means, and their signal strength was assessed using the inter-series correlation (Rbar) and the expressed population signal (EPS; Wigley et al., 1984). The similarity among the indexed TRW chronologies was assessed using hierarchical cluster analysis.

2.3. Climate data

Climatic data were derived by interpolating from a set of nearby weather stations with locally weighted regressions and the effect of altitude. The original station series measurements were subjected to quality control and homogenization using ProClimDB (Štěpánek, 2007; Štěpánek et al., 2009). These measurements included 268 meteorological stations and 787 precipitation stations representing the territory of the Czech Republic. All observations of the weather variables were tested for outliers and breaks through a detailed homogenization sequence, and the gaps in missing data were filled (Štěpánek et al., 2009, 2011). All weather elements could be interpolated by the highdensity network for the sampling sites, particularly focusing on the features of the mountainous terrain (Štěpánek et al., 2011). The database for the research area included data on the daily average, and minimum and maximum temperatures (T_{avg} , T_{min} and T_{max} , respectively); daily sum of global radiation; mean daily wind speed; mean daily relative humidity; and daily sum of precipitation. With these input parameters, the effective global radiation (EGR) value for each site was calculated as the sum of the global radiation on frost- and snow-free days with mean daily temperatures above 5 °C and no water stress. Most of the key climate factors can be simultaneously considered using this complex indicator (e.g., Trnka et al., 2011). The SoilClim (Hlavinka et al., 2011) model was used to estimate the daily values of the relative soil water content (AWR) for the top 1.3 m, which was used as one of the water availability proxies. All climate data cover the period from 1961 to 2011.

2.4. Sulfur and nitrogen deposition measurements

Since November 1993, the monthly cumulative samples of precipitation (bulk deposition, two samplers) and spruce canopy throughfall have been collected from the Krkonoše Mountains near the 4M site (Table 1 and Fig. 1). Rain was sampled using polyethylene collectors placed 1.5-2 m above the ground and protected against light, bird perching, and sample evaporation (samples were collected in a bottle connected to the collector funnel by a 1-mm-diameter tube). Highdensity polyethylene cylinders situated 2-2.5 m above the ground were used to sample snow. The samples from individual collectors were combined to yield a single composite sample for each site and weighed to determine the amount of water for deposition calculations. The samples were prefiltered through a 200-µm polyethylene sieve to remove coarse particles, either during collection (rain collectors were equipped with a sieve) or immediately after melting the snow from the winter collectors. Then, the samples were stored in the dark at 4 °C before analysis. The sulfate (SO_4^{2-}) and nitrate (NO_3^{-}) concentrations were measured using ion chromatography. The total S deposition was calculated from throughfall, and N deposition from bulk precipitation.

2.5. Sulfur and nitrogen deposition estimates

A long time series of parallel data was obtained from the marked temporal correlation between measured S and N depositions and their respective emission rates during the last two decades, such that the significant relationships between these variables could be calculated. Consequently, these data could be used to estimate long-term deposition sequences based on reconstructed emission trends (Schöpp et al., 2003; Kopáček and Veselý, 2005; Kopáček et al., 2012). Relative changes



Fig. 2. Measured sulfur (white) and nitrogen (black) deposition near the 4M site since 1994.

in the reconstructed deposition sequences are generally valid for relatively large regions (Schöpp et al., 2003). To determine historical S deposition for the period before 1994, a statistical method based on this temporal coherence between the measured S deposition and the respective Central European emission rates of SO₂ was used (Kopáček et al., 2012). Briefly, SO₂ emissions were calculated from fuel consumption (hard coal, brown coal, lignite, and oil), the burning of wood and natural gas, and emissions from smelting. Because the annual mean precipitation SO₄, as well as throughfall SO₄, showed an overall marked temporal coherence, concentration could be used to represent with respect to the emission rates. SO₄ in bulk and throughfall precipitation and the SO₂ emission rate were found to be significantly correlated $(R^2 = 0.73, p < 0.05)$. This linear regression equation was used to calculate the annual SO₄ concentration for precipitation from the emission rates for the whole period of emission reconstruction (1920-2011). Deposition was calculated as a product of estimated SO₄ concentration and measured (1961-2011) precipitation depth.

Since 1994, the S throughfall deposition measured near the 4M site reduced by 71%, from 42 (1994–1995) to 12 kg S ha⁻¹ year (2010–2011), and bulk deposition by 76% (Fig. 2). Within the same period, the SO₂ emissions decreased by 73%; therefore, the annual decline in the SO₄ concentration in precipitation was closely related to the reduction of SO₂ emissions ($R^2 = 0.72$, p < 0.05). Based on records of SO₂ emissions (Kopáček and Veselý, 2005), the estimated S deposition peaked in the 1980s, with a S throughfall deposition of 53 kg S ha⁻¹ per year (1980–1985). The measured S throughfall deposition for 2007–2011 (10 kg S ha⁻¹ per year) was lower than the estimated S deposition for the period 1961–1965 (38 kg S ha⁻¹ per year) (Fig. 3).

The measured N deposition in bulk precipitation decreased significantly (p < 0.05) beginning in 1994 (Fig. 2). However, the N deposition decline was less pronounced than that of S deposition, that is, from 18 kg N ha⁻¹ per year (1994–1995) to 7 kg N ha⁻¹ per year (2010–2011). Similar to the S deposition estimates, reconstructed N bulk deposition showed a similar long-term trend to the respective NO_x and NH₃ emissions. The estimated N bulk deposition peaked in the 1980s, with a N deposition of 28 kg N ha⁻¹ per year (1980–1985). The measured N bulk deposition for 2007–2011 (9 kg N ha⁻¹ per year) was lower than the estimated N deposition for the period 1961–1965 (19 kg N ha⁻¹ per year) (Fig. 4).

2.6. Growth-climate response analysis

DendroClim2002 was used to model TRW based on climatic characteristics (Biondi and Waikul, 2004). The residual spruce chronology and climatic time series from 1961–2011 were used to calculate the correlation coefficients between the TRW indices and climatic drivers. The correlation coefficients were calculated for a seasonal window from May of



Fig. 3. (A) Regression between measured and modeled S deposition. (B) Reconstructed (black line) and measured (points) S throughfall deposition since the 1920s. Deposition before 1961 was calculated using average annual precipitation.



Fig. 4. (A) Regression between measured and modeled N deposition. (B) Reconstructed (black line) and measured (points) N bulk deposition since the 1920s. Deposition before 1961 was calculated using average annual precipitation.



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Fig. 5. (*A*) Site similarities of TRW index chronologies indicated using hierarchical cluster analysis. Strips show negative pointer years for the investigated period (1961–2011) with >40% (light gray) and 75% (dark gray) reduction. (*B*) Sample replication for the Krkonoše Mts.

the previous year until August of the year of the tree-ring formation (referred to as "the given year"). This interval should have the maximum impact on the TRW (Treml et al., 2014). In addition to monthly values, the seasonal means or sums of the previous and current years were analyzed. To determine the temporal variability in the growthclimate relationship, correlation coefficients were also computed for 13-year moving windows using Statistica (Statsoft, Inc.; Prague, Czech Republic). Temperature means (May–July), precipitation totals (March), and sulfur and nitrogen depositions were selected for the correlation analysis. The negative pointer years were analyzed for the period replicated by at least 20 TRW series (from 1859 to the present). A reduction in TRW that was 40% greater than the average TRW in the previous 4 years and that was found in at least 40% of the trees was considered (Schweingruber et al., 1990).

3. Results

3.1. TRW chronologies

The TRW chronologies from the five sites vary in length (from 65 to 228 tree rings) and cover a wide altitudinal range in the Krkonoše Mountains. The high reliability of all chronologies was confirmed by the Rbar (>0.61) and EPS (>0.90) values, which remained above the threshold of 0.85 (Wigley et al., 1984) for the full period of the analysis. Replication decreased backward in time, but it did not drop below the 30 series during the studied period (Fig. 5b). A higher number of missing tree rings were observed at higher altitudes, where stronger air pollution stress is expected (Table 2). Most of the missing rings (78%) were identified in the period 1980–1984.

Hierarchical cluster analysis confirmed that all indexed TRW chronologies were highly similar (Fig. 5a), which is also apparent from a visual assessment. The similarity of the chronologies was verified in

Table 2

Basic characteristics of the TRW chronologies from all sites.

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Sites	Raw measurement statistics									Master chronology			
	Series	MSL	AGR	SD	Rbar	EPS	AC1	MR	Start	End	Length		
1L	23	58	2.55	1.48	0.70	0.95	0.89	0.00	1946	2010	65		
2M	22	113	1.25	0.61	0.64	0.92	0.85	1.45	1784	2011	228		
3M	17	149	1.19	0.54	0.69	0.93	0.84	0.82	1837	2011	175		
4M	17	125	1.55	0.81	0.61	0.90	0.87	1.65	1847	2011	165		
5U	19	119	1.08	0.50	0.65	0.94	0.83	0.55	1814	2011	198		

MSL, mean segment length; AGR, average growth rate (mm); SD, standard deviation; Rbar, inter-series correlation; EPS, expressed population signal (minimum EPS in the studied period); AC1, first autocorrelation; and MR, mean no. of missing rings tree⁻¹.

all the common negative extremes, which were also captured by analyzing the negative pointer years (Fig. 5a, gray strips). The negative pointer years were analyzed for all series from the Krkonoše Mountains. The greatest decline in the TRW began in 1979 and lasted until 1982. In 1980, 99% of all trees responded negatively. Other considerable reductions in TRW occurred in 1974 (78% of trees) and 1996 (78% of trees).

3.2. TRW response to climate variability

The similarities among the different climatic variables are summarized in Fig. 6a. The best independent variables related to TRW above 1000 m a.s.l. were mean temperature and EGR for the May–July period (Fig. 6b). The mean monthly temperatures influenced TRW mainly at the beginning of the growing season (May) and during the summer (July). This pattern suggests that higher spring and summer temperatures of the given year are positively related to TRW. The EGR emerged as an important indicator of tree behavior for almost the entire duration of the growing season. TRW is significantly controlled by EGR, particularly at the beginning of cambial activity. This effect gradually decreases during the growing season.

The precipitation totals and AWR are not as important for TRW formation in mountain areas, as confirmed by this study. The effect of precipitation was positive in the months preceding the growing season (significant only in March) and negative during the growing season (Fig. 6b). An analysis of the TRW response to AWR indicated a similar negative effect (significant only in May), except for a positive effect before the growing season. The previous growing season was less important for spruce TRW. Significant (positive) correlations were calculated only for AWR during the summer months (Fig. 6b).

3.3. Sulfur and nitrogen deposition effect on TRW

During the time of the highest sulfur deposition, the TRW reached its lowest values (Fig. 7). The TRW indices were reduced by >50% during the 1980s compared with their long-term mean (1850–2011). After the decline in sulfur deposition, the mean TRW indices started to increase, even exceeding the values from the period before the extreme emission load.

Based on the correlations among the climatic variables (Fig. 6a), we selected the May–July temperature means and March precipitation totals for the moving correlation analysis. The mean temperature from May to July was the most important factor for TRW (Fig. 6b). The correlation coefficients within the 13-year moving window emphasized a dominant positive effect of temperature on spruce TRW, similar to the effect of precipitation in March. However, this temperature effect was



Fig. 6. (*A*) A correlation table among climatic variables. The marked values (*) are statistically significant ($\alpha = 0.01$). (*B*) Comparison of the correlation coefficients of the individual residual TRW index chronologies with the mean monthly temperature (T_{avg}), index of effective global radiation (EGR), monthly precipitation totals, and monthly relative available water (AWR) for the period 1961–2011.

not stable over time (Fig. 8b), decreasing at the beginning of the 1980s. Sulfur and nitrogen depositions reached their peak across Central Europe during this period (Fig. 8e, f). Starting from the late 1980s, decreasing sulfur deposition resulted again in the effect of temperature on TRW. The March precipitation effect started to decline in the early 1990s. The temperature effect started to weaken at the turn of the millennium in spite of the controlled low deposition values (Fig. 8e, f) as well as the increasing mean temperature (Fig. 8c) and precipitation totals (Fig. 8d).

4. Discussion and conclusion

4.1. Climate-growth relationship

Our results suggest that TRW is mainly driven by changes in growing season temperature, and additionally by fluctuating EGR (e.g., Sander et al., 1995; Bednarz et al., 1999; Büntgen et al., 2007; Rybníček et al., 2009), (Fig. 6b). The temperatures and EGR in May and July have the strongest effects on TRW. Compared with the cold April temperatures, the average May temperatures reached 9 °C, which corresponds to average spring temperatures for the onset of xylogenesis (Rossi et al., 2008). At the same time, EGR increased considerably along with the length of the photoperiod. The EGR values continued to increase until July, when they reached a peak. Higher values of the EGR during the growing season favor photosynthesis. This finding indicates that spruce TRW is



Fig. 7. Differences of the mean TRW indices and standard deviation before, during, and after the highest acid deposition on the Krkonoše Mountains.

supported by a higher rate of carbon fixation due to sufficient photosynthetically active radiation. Spruce, a species requiring light, responds sensitively to higher radiation. It is likely that ongoing temperature increases stimulate a prolonged season of vegetative growth (early thaw), therefore increasing carbon allocation to woody biomass.



Fig. 8. (*A*) Mean TRW indexed chronology (gray) smoothed by the Lowess curve (black); (*B*) 13-year moving correlations of TRW indices with temperature means (May–July; black line), precipitation sums (March; light gray line), S deposition values (dark gray line) and N deposition values (gray dashed line) of the given year. Values of individual parameters (gray) smoothed by the Lowess curves (black) of May–July temperature (*C*), March precipitation (*D*), S deposition (*F*).

In the last year, TRW was positively related to AWR (Fig. 6b). A rainy and humid season during the preceding summer can facilitate the storage of nutrients, which are then used for growth at the initial stage of tree-ring formation in the following year, as previously reported, (e.g., Feliksik, 1993; Rybníček et al., 2010b). Rainy months before the onset of cambial activity can also positively affect spruce growth at high altitudes (Fig. 6b). High February/March precipitation totals, or rather snow cover, in this period protect the ground from being frozen solid. Therefore, the root system cannot be damaged due to the physiological drought at the beginning of spring.

4.2. TRW reduction induced by acid deposition

Soil acidification in the Krkonoše Mountains did not progress significantly until the 1950s-1960s with respect to the soil parameters important for tree species (e.g., Bc/Al ratio and Al concentration; Hruška et al., 2001). The deposited base cations and those from the ionexchange soil complex were able to eliminate the acidic inputs. However, as soil acidification progressed, the base cations were leached from the soil-rooting zone (Hruška and Cienciala, 2003; Oulehle et al., 2007). In the second half of the 1950s, the sulfur and nitrogen depositions increased, exceeding 30 kg S ha^{-1} per year and 15 kg N ha^{-1} per year⁻¹, and increased further to 60 kg S ha⁻¹ per year and 30 kg N ha^{-1} per year during the 1970s and 1980s (Fig. 8). The increases in acid deposition led to a depletion of the ion-exchange soil complex and suppression of litter decomposition (Oulehle et al., 2012). The Al concentration in the soil increased and the base saturation of the soil and the pH of the soil solution decreased considerably; consequently, the Bc/Al molar ratio in the soil water was reduced (e.g., Krám et al., 1995). These changes were modeled in detail for one study site in the Krkonoše Mountains, namely, the 1L site (Hruška and Cienciala, 2003). These soil parameters showed a significant decreasing trend between 1955 and 1960. In spite of the soil acidification, the TRW decreased relatively slowly beginning in the 1950s. Concurrently, the frequency of the negative pointer years gradually increased in comparison with the period before World War II (Fig. 5).

Aerial liming was used to mitigate soil acidity and reduced Al toxicity in several areas of the Czech Republic (e.g., Hruška and Cienciala, 2003; Krám et al., 1995). Only the 1L site was limed in the first half of the 1980s with 5 tons ha⁻¹ of dolomitic limestone (pers. comm., Krkonoše National Park Authority), without any apparent effect on TRW (Fig. 5).

Worse soil conditions at the beginning of the 1960s led to a weakening of the stands in the 1970s, when summer temperatures were no longer the driving factor (Fig. 8b). Negative extremes in 1965 (45% of the trees responded negatively) and mainly in 1974 (78%) were caused by a lower mean May-July temperature and EGR as well as lower March precipitation totals compared with the long-term means. As shown by the results in Fig. 6b, these factors play a key role in determining spruce TRW in mountain areas. Moreover, significant TRW reductions in 1923, 1942 (Treml et al., 2012), and 1956 (Kroupová, 2002) were related to anomalies in temperature. By contrast, the most extreme droughts in the Czech Republic in 1868 (summer), 1904 (summer), 1911 (summer), 1946 (spring), 1947, 1976, or 2003 (Brázdil et al., 2009, 2015) were not reflected in our TRW chronology data. Therefore, we suppose that subalpine spruce stands primarily reflect a temperature signal because the precipitation totals in this region are sufficient for spruce. The combined temperature and precipitation effect drive TRW growth in the lower mountain forests (e.g., Rybníček et al., 2009).

After the extremely cold and harsh winter of 1978/1979, the TRW reached minimum values (until approximately 1985) throughout the tree growth period. An intensive cold front hit the Czech Republic on 31 December 1978 and 1 January 1979. The meteorological stations registered the sudden drop in temperature (approximately 25 °C in 24 h) from approximately + 10 °C at the New Year's Eve night of 1978/1979 (Rein and Štekl, 1981). The coincidence of freezing temperatures and high values of SO₂ usually leads to forest decline (Sheppard and Pfanz,

2001). This coincidence had already led to extensive forest dieback in the Ore Mountains (Krušné hory) in the winter of 1977/1978. Chlorophyll damage and withering of needles were observed when high concentrations of SO₂ came in contact with the assimilatory organs of spruce. Such damage leading to forest dieback can occur after just tens of minutes of contact under suitable weather conditions (Hruška et al., 2009). Moreover, the trees very likely lost their frost resistance over Christmas of 1978, when the temperature was approximately 10 °C. An extreme temperature drop can also cause damage to buds, which can lead to a change in the phytohormone content. Such changes can play a crucial role in the sequence of stress-induced processes (Itai, 1999). The marked reduction of TRW, noted by Kroupová (2002) and Dittmar and Elling (2004), can be attributed not only to the disrupted assimilation apparatus (primary shoots) but also to a reduced adaptability of trees to emissions, that is, the ability to regenerate (limitation of the secondary shoot growth).

During this time, the health of the spruce stands in the Krkonoše Mountains declined. Many missing rings were observed in this period. Defoliation increased by approximately 1% annually until 1980 and by 3-16% over the period 1981-1989 (Vacek and Matějka, 2010). The damaged assimilation apparatus was repeatedly replaced with secondary shoots (Polák et al., 2007). In addition, acid deposition influenced the soil carbon cycle by reducing dissolved organic carbon leaching (Evans et al., 2005) and fine root and mycorrhizal biomass (Clemensson-Lindell and Persson, 1995; Peter et al., 2008) and by changing the fungi to bacteria ratio (Kopáček et al., 2013), which might result in suppressed organic matter turnover and litter decomposition (Meiwes et al., 2009; Oulehle et al., 2011; Lawrence et al., 2012). Reduced forest biomass productivity and an impaired soil environment resulted in high N losses from mountain forest ecosystems in the Krkonoše Mountains (Oulehle et al., 2008; Kopáček et al., 2013). The largest acid deposition was observed in the 1980s, as the stands were exposed to extreme airborne SO₂ concentrations.

4.3. TRW recovery

At the end of the 1980s, a reduction in sulfur and nitrogen deposition was accompanied by TRW restoration. Despite the forest decline and dieback, the surviving spruce stands exhibited an increased TRW growth. In the first half of the 1990s, the sulfur deposition effect became insignificant, although the 1996 TRW reduction was linked to acidic rime. The May–July temperature and March precipitation effects intensified once again. Therefore, the TRW increase was most likely triggered by the combined effect of efficient pollution control and a warmer but not drier climate (Fig. 8). The widespread forest decline could lead also to growth rebound of surviving trees because of increased availability of light or a nutrient pool caused by decomposition of the biomass of dead trees. However, the main tree dieback occurred at the end of the 1970s, whereas the TRW restoration started ca. 10 years later. Moreover, most of the dead or dying trees were cut down and removed from the forest (Hruška and Cienciala, 2003).

The marked temperature–growth relationship weakened again at the turn of the millennium for various, as yet unclear reasons. Despite these changes, the TRW recovery was uprecedented (Figs. 5a and 8a), and our observations are in line with those of Treml et al. (2012), who observed similar TRW increases in the same area over the last two decades. In warmer regions at lower elevations, such changes can be explained by warmer temperatures over the traditional dormant period (e.g., Kosiba et al., 2013 or Ögren et al., 1997).

Sulfur and nitrogen deposition are important drivers of terrestrial carbon and N cycling. A majority of temperate ecosystems are N limited (Vitousek and Howarth, 1991); consequently, fertilization by atmospheric N deposition is thought to have increased C storage in the biomass and soils of terrestrial ecosystems and thus stimulate C sequestration. The increased N availability may increase net primary productivity (NPP) and/or decrease the decomposition of N-rich litter

(Janssens et al., 2010; Liu and Greaver, 2010). Acidification (from either S or N deposition) may also affect the suppression of litter decomposition. This effect may occur by reduced availability of C for microbes rather than a direct effect on the microorganisms themselves (Pennanen et al., 1998; Persson et al., 1989).

However, despite the high N deposition in the 1980s, forests in this period were markedly affected by acid deposition, resulting in suppressed TRW and weak N retention with high nitrate losses to surface waters. Subsequently, the temporal correlation between acid deposition reduction and TRW recovery led to higher nitrogen immobilization within the forest ecosystem, which explained the decreased nitrate leaching beginning in the 1990s (Oulehle et al., 2008). We hypothesize that the cessation of acid deposition has led to the recovery of fine root biomass and mycorrhizae and has enhanced soil organic matter decomposition and the release of nutrients (N, Ca, and Mg) that had accumulated in soil organic matter. As a consequence, forest ecosystems have benefited from enhanced nutrient availability that stimulated forest productivity, observed since the 1990s.

4.4. Implications for dendroclimatological reconstructions

The reduced sensitivity of recent TRW to temperature, known as the "divergence problem," has been investigated in many studies (e.g., Briffa et al., 1998; Büntgen et al., 2008; D'Arrigo et al., 2008). The described TRW fluctuations and the weakening of the climate signal related to the atmospheric pollution effect should be considered as another potential factor in this problem. Considering that large parts of mountain forests have been historically subjected to acid deposition, the effects of acid deposition on TRW have to be included in dendrochronology studies, particularly with respect to dendroclimatological reconstructions. Furthermore, the chemical recovery of forest soils following the reduction of acid deposition, with wider implications for soil organic matter cycling and nutrient availability, may have altered recent TRW development. Forest ecosystems are subject to several environmental changes, including climate change. Our research highlights the importance of the legacy of acid deposition on dendroclimatological reconstructions.

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