Water Air Soil Pollut (2012) 223:5727–5744
DOI 10.1007/s11270-012-1310-6

The Impact of Industrial SO_2 Pollution on North Bohemia Conifers

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Received: 21 March 2012 / Accepted: 29 August 2012 / Published online: 29 September 2012

Abstract Conifer forests in the Jizerské Mountains, Czech Republic have experienced widespread and long-lasting effects related to industrial SO_2 pollution. To explore the spatial and temporal impact of this phenomenon on Norway spruce stands, a transect of sites was sampled to the southeast of the Polish coal-fired power station Turów. Tree growth at all sites displayed a significant reduction around 1980, which could not be explained by climate alone. However, by incorporating both climate and SO_2 variables in multiple regression models, the chronology trends could be explained well. The lowest growth rates were found to coincide with the period of greatest atmospheric SO_2 concentrations and the degree of suppression decreased with increasing distance from the power station. The period of growth suppression in a Silver fir site appeared to be more severe and longer in duration than for the spruce, although differing site conditions prevented a direct comparison. Fir trees also appeared to be affected by SO_2 pollution earlier in the twentieth century compared to spruce. Growth of both species, however, did not return to predicted levels following the reduction of pollution levels in the 1990s. A comparison with spruce and fir data from the Bavarian Forest, a region also affected by pollution in the past, revealed a temporal difference in growth suppression, likely related to different timings and loadings of SO_2 emissions between both regions. This study highlights pollution as another potential causal factor for the 'divergence problem' and dendroclimatic reconstructions in polluted regions should be developed with caution.

Keywords Dendrochronology · Ring width · Atmospheric pollution · SO_2 emissions · Tree growth modelling · Divergence

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1 Introduction

Atmospheric pollution from industrial and other sources has profoundly influenced the function of natural ecosystems in many regions, with dramatic consequences in some cases. During the second half of the twentieth century, levels of atmospheric pollution in parts of central Europe were among the highest on the continent. Forest stands in previously severely polluted areas such as the 'Black Triangle' (Fig. 1), including the northwest of the Czech Republic, have faced decline and problems remain to this day (Neuhöferová 2005). Although previous dendroecological studies in the region have addressed forest reaction to atmospheric pollution (e.g. Kroupová 2002), most do not consider the impact of a single point source and instead generally focus on the temporal context of tree response, although there are some exceptions (e.g.

Danek 2007). Additional dendrochronological information from the area is therefore required to assess the spatial influence of point source pollution on tree growth.

Atmospheric pollution can impact tree growth both directly and indirectly. Direct impact is related to stomatal uptake of high concentrations of gaseous pollutants. For example, Slovik et al. (1996) found a significant correlation between crown damage and stomatal SO_2 uptake in Norway spruce in central Germany. Tree vitality can also be impacted indirectly when SO_2 and NO_x react with water in the atmosphere, forming acidic solutions which are then deposited in the environment (Bäck et al. 1995). One of the consequences of soil acidification from acid deposition is leaching of Ca^{2+} and Mg^{2+} cations and toxicity from increased concentrations of Al, as well as increased fluxes of nitrate and H^+ through the soil profile. Such disruption of biogeochemical cycling may result in nutrient deficiency and imbalances, leading to a decline of tree vitality and growth (Sherman and Fahey 1994). In a study investigating the effects of SO_2 pollution on Silver fir in southern Germany, Elling et al. (2009) suggested that the rapid negative (i.e. decreased growth or mortality) response to increased SO_2 concentrations and quick recovery following a decline in concentration of the pollutant indicates a direct impact on the organism as described above. At least with Silver fir, this may be indicative of the greater significance of the first process and could also be the case with other conifers.

The detrimental effects of atmospheric pollution (and more specifically SO_2) on plant and tree growth were initially identified in the first half of the twentieth century particularly in and around large cities and industrial areas (e.g. Benner and O'Connor 1913; Metcalfe 1941; Ruston 1921; Swain 1923). In the 1980s and 1990s, studies on 'forest decline' in Europe, mainly centred around the Fichtelgebige, Erzgebirge and the Bavarian Forest in Germany, addressed whether forest stand decline, expressed by tree ring width reduction and mortality, was natural or anthropogenic in origin. For example, multiple studies identified atmospheric pollution as the primary cause (e.g. Elling 1993; Elling 2001; Visser and Molenaar 1992a, b), while some researchers proposed pathogenic factors (e.g. Kandler 1992, 1993). See also review by Pitelka and Raynal (1989). Along with this debate on the cause of forest decline, the spatial extent of a potential decline was also discussed (Kandler and Innes 1995).

More recent investigations have shed further light on this issue. Wilson and Elling (2004) expanded upon earlier work by Eckstein and Sass (1989) and Kandler (1993) focussing on an anomalous period of suppressed growth in Silver fir in the 1970s and early 1980s in the Bavarian Forest. They explained that 'Silver fir is known to be sensitive to SO_2 emissions and is considered a bio-indicator for SO_2' (Wilson and Elling 2004: p. 25) and suggested that although fir trees are less sensitive to drought, they appear to be more sensitive to non-climatic factors such as pollution than spruce. The negative impact of increased atmospheric SO_2 pollution on Silver fir was further highlighted in Filipiak and Ufnalski (2004) and Elling et al. (2009).

Although not as sensitive as Silver fir, other conifers such as Scots pine and Norway spruce have also been used in atmospheric pollution studies (e.g. Danek 2007; Wilczyński 2006; Wilczyński and Feliksik 2005). Interestingly, Vacek and Lepš (1996) noted the increased susceptibility of Norway spruce to pollution at higher elevations in the Krkonoše Mountains of the Czech Republic (Fig. 1), particularly near the upper tree line, where the growing season is significantly shorter and where the trees are near their temperature physiological limit compared to lower elevation stands. Wilczyński (2006) suggested that this is the reason for the differential impact of atmospheric pollution on Norway spruce and Scots pine in the Sudetes in Poland. Although the reaction of both species to SO_2 is similar (Mooi 1976), Scots pine trees are predominantly located at lower elevations, resulting in decreased exposure to atmospheric pollution and therefore a greater capacity to withstand its effects compared to stands at more temperature-limited higher elevation sites.

Most of the above studies, investigating the effects of SO_2 on coniferous tree species, suggest a similar response to pollution, regardless of species, geographical location and site-specific conditions, although these factors may influence the degree to which the trees are affected. In general, an increase in SO_2 concentrations in the atmosphere leads to a suppression of growth rates. When SO_2 concentrations decrease, a period of growth recovery follows and it has been suggested that there is a connection between the suppression severity and recovery strength (Filipiak and Ufnalski 2004).

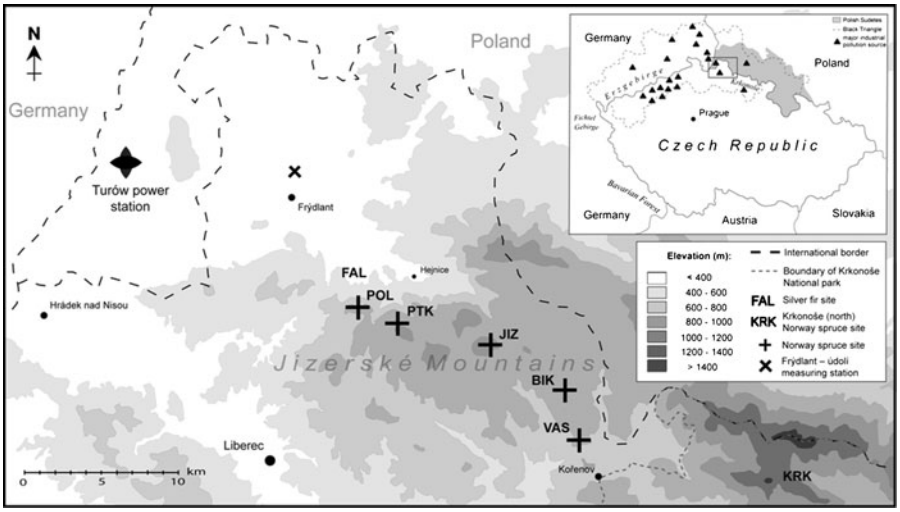

Fig. 1 Study region: Jizerské Mountains and surrounding area. The 'Black Triangle' is shown in the *inset map with a dashed line*

In this paper, we detail dendrochronological analyses to investigate the spatiotemporal impact of SO_2 pollution in the Jizerské Mountains of the Czech Republic on the growth of Norway spruce in the period since the Turów power station commenced service in 1962 (Kasztelewicz and Ptak 2009). Identifying how forests react to an environmental stress factor both spatially and temporally has the potential to enhance the understanding of forest dynamics at polluted sites. Such information will not only aid the development of forest conservation and renewal strategies for forests affected by pollution but also addresses one of the possible causal factors of the 'divergence problem' in dendroclimatology, expressed as a weakening or loss of tree growth response to temperature in the latter half of the twentieth century (D'Arrigo et al. 2008).

2 Methodology

2.1 Study Region

The study region is centrally placed within the 'Black Triangle' (Fig. 1). This region was considered to have the highest levels of atmospheric pollution in Europe in the 1970s and 1980s (Wilczyński 2006) and was therefore a suitable choice of location to carry out an investigation into the effects of atmospheric pollution on tree growth as well as assessing growth recovery as emissions decreased. The high levels of pollution were the result of heavy industrial development, particularly the operation of several power stations burning locally mined lignite with a high sulphur content, which were constructed along the border area of the Czech Republic, Germany and Poland (Filipiak and Ufnalski 2004; Wilczyński 2006). The sampling sites were located in the Jizerské Mountains in northern Bohemia in the Czech Republic (Fig. 1), which is classified as a protected natural region (Chráněná Krajinná Oblast—CHKO). Climatically, the Jizerské Mountains can be characterised as mild cold with considerable amounts of precipitation owing to the orographic nature of the area and supply of moist oceanic air by westerly winds. The majority of the upper plateau is situated between 800 and 1,000 m asl. (Ulbrichová and Šimková 2005).

The coal-fired Turów power station was located to the northwest of the investigated area with several smaller coal power stations and other less significant industrial pollution sources in the 'Black Triangle' farther west (Fig. 1). The targeted sites were the closest conifer woodlands at high elevation and were well situated to study the spatial influence of industrial emission output on tree growth for this region. Although some influence on local pollution levels from other industrial sources, particularly upwind (SW) of the sampled area, may be expected, sulphur deposition models in the region suggest that the influence of emissions is greatest in close proximity to each emission source and rapidly declines with increasing distance (Ardö et al. 2000; Zemek et al. 2006). As the largest regional SO_2 source, Turów's influence on the sampled sites is undoubtedly dominant, particularly considering its close proximity.

Turów commenced operation in 1962 and currently operates at an output capacity of just over

2,100 MW using brown coal as the primary energy source (Kasztelewicz and Ptak 2009). Local (and regional) atmospheric SO_2 concentrations in the study area from 1971 to 2007 (1969 to 2007), along with the historical, country-wide context of sulphur emissions during the 1850–1990 period illustrate the varying levels of this pollutant over time (Fig. 2). During the 1990s, particularly after 1994, steps were taken to modernise Turów with the introduction of new technology, including the replacement of old coal boilers, adoption of equipment for desulphurising flue gases and electrostatic filters to minimise dust release into the atmosphere which resulted in an 82 % decrease of SO_2 emissions, reduction of NO_x by 45 % and a 96 % reduction of dust pollution (Nordic Investment Bank 2005).

2.2 Site Selection and Sample Collection

Although the sensitivity of Silver fir to environmental pollution would make it the best candidate species for this study (Elling et al. 2009), its presence in the Jizerské Mountains today is rare mainly due to past forest management practices (Macel and Sakai 2003). Sampling was therefore primarily focussed on Norway spruce—the prevailing tree species in the region (Mládková et al. 2006). In order to assess the influence of SO_2 emissions on tree growth in the Jizerské Mountains, six sites were sampled along a SE directional transect from the Turów power station (Fig. 1) in August of 2008, ensuring that the primary modulating variable was distance from the emission source and other variables such as elevation, aspect, etc. were kept relatively constant. Sampling downwind along a

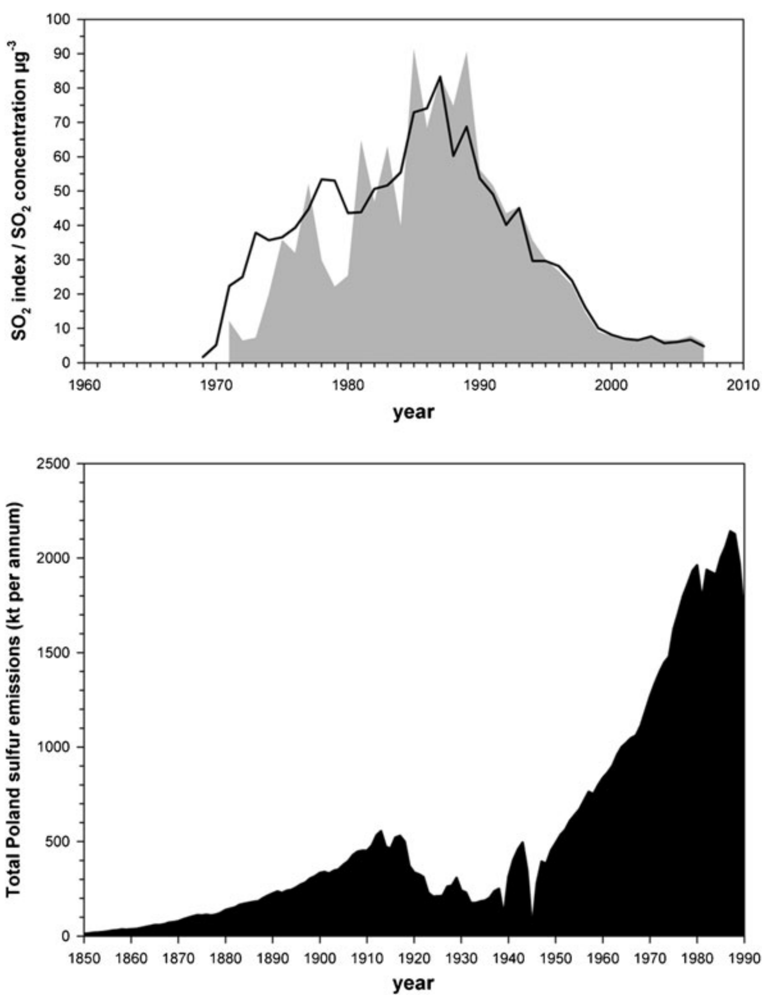

Fig. 2 *Top* Atmospheric concentrations of SO_2 from the Frýdlant measuring station (*shaded area*; see Fig. 1 for location) and regional SO_2 concentration index including data from 41 atmospheric SO_2 concentration datasets from the studied region (*black line*). *Bottom* Total country-wide historical sulphur emissions data for Poland. Note that the Frýdlant data are composed of two separate datasets covering an earlier (1971–1995) and later (1993–2007) period with a 2-year period of overlap. Both datasets were combined due to very close geographical proximity and good agreement in the overlapping period

NE transect from Turów was not possible due to the low elevation of the land and sparse forest cover. Samples of Norway spruce were collected at five sites and Silver fir sampled at one. Podzol soils were present at the Norway spruce sites, while the Silver fir trees were situated on a Cambisol (Němeček et al. 2001). Two cores were collected per tree using a Swedish increment corer (Grissino-Mayer 2003), with a minimum of 15 trees sampled per site (Table 1).

Although every effort was made to select sites with similar conditions, this was hindered by local topography, site-specific features and the absence of suitable stands. Finding stands of adequate age in severely damaged areas proved particularly problematic. The importance of selecting sites of a similar elevation was highlighted by Wilson and Hopfmueller (2001), who identified that the response of Norway spruce to climatic factors differed with elevation. Additionally, differences in elevation and aspect would undoubtedly influence the amount of exposure to pollutants in the atmosphere. Ultimately, a compromise between the above factors was necessary during site selection due to varying local conditions. Site summary information is detailed in Table 1.

Mean monthly SO_2 measurements from local measuring stations in the study area were provided by CHMÚ (Czech Hydrometeorological Institute). To address the temporally patchy nature of SO_2 data from a network of measuring stations around the Jizerské Mountains region, annual means from 44 station datasets were used to create an index of atmospheric SO_2 levels. The mean and standard deviation of the datasets from each station were adjusted to the most complete SO_2 measurement datasets from the Frýdlant measuring stations in close proximity to Turów (Fig. 1). The means of all data were calculated for each year and the variance adjusted to account for the varying number of values in a particular year (Osborn et al. 1997) (Fig. 2—top, black line). This composite mean was strongly correlated (r=0.86) with data from Frýdlant.

Climate data, consisting of mean monthly precipitation and mean monthly temperatures at a resolution of 0.5° by 0.5°, were used for tree growth modelling (New et al. 2002). The additional 'Krkonose north' (KRK) (Fig. 1) Norway spruce dataset was acquired from the International Tree Ring Data Bank (Sander et al. 1995) and historical sulphur data for Poland were obtained from Lefohn et al. (1999).

2.3 Sample Preparation and Statistical Analysis

The samples were prepared, cross-dated and measured according to standard dendrochronological practice (Stokes and Smiley 1968). The samples were initially inspected and visually cross-dated. Ring width was measured on a Velmex traversing measuring stage using the measuring programme MeasureJ2X. Additionally, programmes COFECHA (Grissino-Mayer 2001) and Cdendro (Larsson 2008) were used to statistically verify the dating of the series.

Prior to detrending, an adaptive power transformation was applied to the raw ring width in order to

Table 1 Parameters of sampled sites

Site symbol	Estimated elevation (m asl.)	Horizontal distance from Turów (km)	Estimated average slope (°)	Aspect	Mean series length (years)	Number of trees sampled	Number of series	First year with EPS>0.85
KRK[a]	1,000+[b]	46[b]	Unknown	N	137	45	45	1822
VAS	830	35.5	0–5	S	88.2	15	30	1913
BIK	950	32.5	5	NW–W	71.3	15	30	1934
JIZ	1,060–1,080	26.6	10–15	N–NW	69.2	15	30	1925
PTK	960	20.6	5–20	NW	63.2	17	33	1937
POL	820–850	17.9	25–35	NNW	82.7	17	34	1836
FAL[c]	380	17.3	0	–	116.5	15	30	1883

[a] Data acquired from ITRDB (2008)

[b] Rough estimation (exact location of site is unknown)

[c] Silver fir site

stabilise variance in the data and reduce potential 'end-effect' index inflation in the recent period (Cook and Peters 1997). In order to remove the age-related growth trend, measured values were detrended using either a negative exponential curve or regression function of negative or zero slope (Cook et al. 1990). The product of the detrending process was a standardised chronology where the raw ring width measurements were converted to a standard ring width index (RWI). Index values were calculated by subtracting the idealised least squares fitted function from the power transformed ring width data. Detrending of the raw chronologies was performed using ARSTAN (Cook 1985).

The Expressed Population Signal (EPS) was used as a measure of signal strength. Because the EPS incorporates sample depth (or replication) in the form of the number of tree series in determining the signal strength, the 'expressed' signal will inevitably diminish towards the earlier portions of a chronology as replication decreases (Briffa 1995). It is therefore possible to determine a break-off point when the EPS falls below an arbitrary threshold value in the chronology and the data are no longer considered sufficiently robust for analysis. A value of 0.85 is often cited as an acceptable threshold (e.g. Wigley 1984) and was used herein. For the full period of analysis, EPS remained above this threshold value in all chronologies (Table 1).

2.4 Correlation Response Function Analysis (CRFA) and Growth Modelling

CRFA uses the Pearson's correlation coefficient to identify significant empirical relationships between RWI and monthly and seasonal climate (temperature and precipitation) parameters. Estimates of climate/growth relationships were devised by developing predictive models for all chronologies. It should be noted that the common length of the period for model development was limited by sample replication in the BIK and PTK chronologies where the EPS fell below 0.85 around the 1930s (Table 1). The models were subsequently used to predict theoretical growth as a function of climate through the period of greatest SO_2 influence. This climate/growth model (CGM) was developed using linear regression. According to Cook et al. (1990), if a linear relationship exists between the predictor and predictand, then a statistical function can be applied in order to estimate the predictand (in this case ring width) as a function of the predictor (climate data) for each year. Calibration of the model was carried out over the 1940–1974 period by estimating the growth/climate relationship using the optimal season identified in the CRFA for a given period (the calibration period), when both the predictor and predictand are known. Thereafter, extrapolation into the prediction (verification) period (1975–2005) was performed by inputting the climate data from that period into the model, yielding the predicted RWI values for that period. In the case when growth was significantly affected by both temperature and precipitation, both parameters entered the CGM using multiple regression.

The analysis was extended by developing a model that also included mean annual values of atmospheric SO_2 concentrations as an additional predictor (in addition to the climate parameters) in a similar way to that of the CGM described above (hereafter referred to as climate/sulphur/growth model—CSGM). In this case, however, calibration was performed over the 1969–2005 period using the same climate parameter(s) from the CGM, but also including SO_2. The results from this regression modelling could then be compared to the prediction results of the CGM to address whether the addition of the SO_2 data helped to better model the trends noted in the RWI data.

3 Results

A total of 268 missing rings were identified from all of the analysed samples (Fig. 3). Site chronologies VAS, BIK and JIZ exhibited no missing rings prior to 1980, PTK showed only sporadic missing rings and KRK contained only a couple of missing rings immediately prior to 1980. However, more missing rings occurred in POL during the pre-1980 period than in the other Norway spruce sites, particularly during the 1940s and 1950s. The fir site FAL showed sporadic missing rings in the early parts of the chronology, which increased markedly after 1965. A significant change was observed from 1980 onwards when the numbers of missing rings increased substantially at all sites and even began to appear at VAS, BIK and JIZ, where no missing rings were observed in any of the earlier sections of the chronologies. The number of missing rings started to

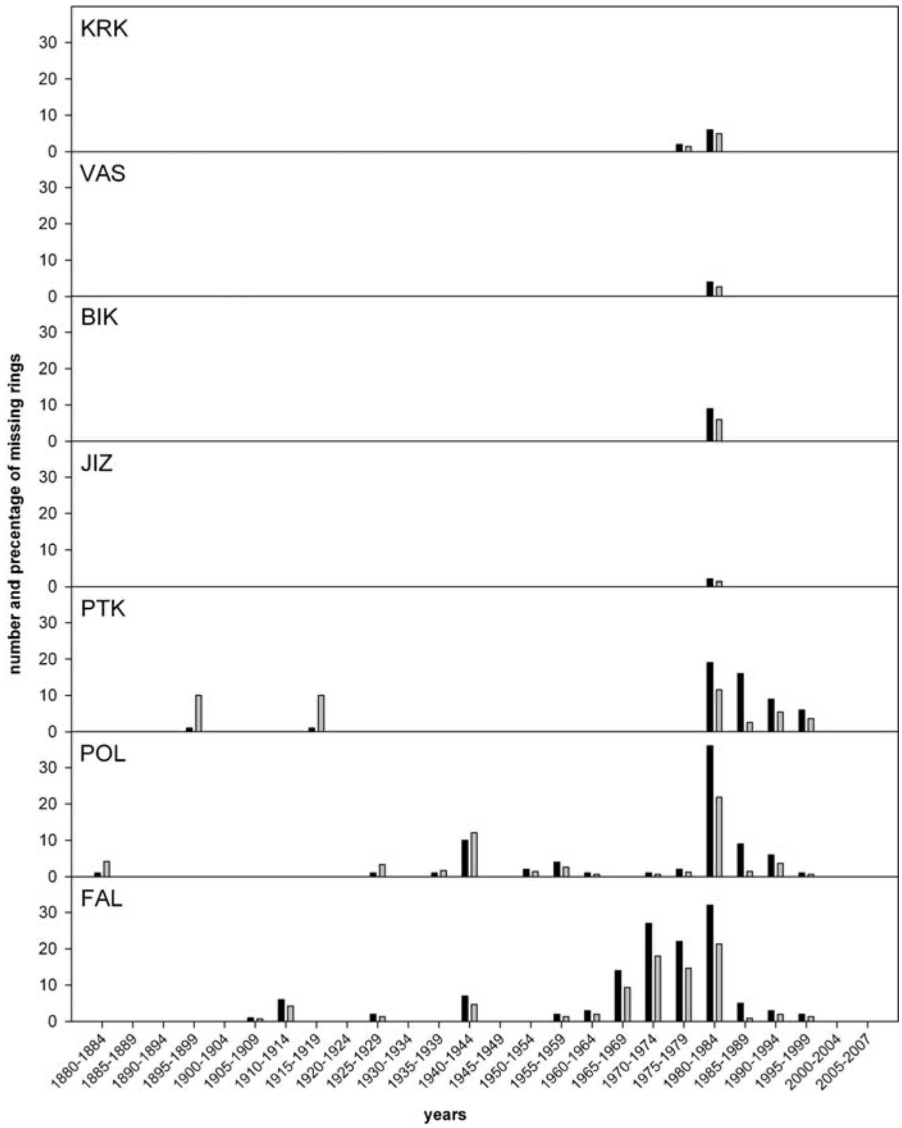

Fig. 3 Chronological list of missing rings from all seven sites. Missing rings are grouped together in 5-year intervals (*black bars*—absolute number of missing rings, *grey bars*—percentage of missing rings)

reduce after 1990 and none of the trees studied had any missing rings in the recent 2000–2007 period.

3.1 RWI and Distance

An analysis of the spatial variation of the chronologies identified a significant relationship between distance and RWI (Fig. 4). A very strong positive linear relationship (r_{adj}^2=90 %) was noted between distance from the Turów power station and the RWI for 1980 (the first and most severe year of growth suppression in the spruce chronologies). The existence of a similar relationship (r_{adj}^2=77.5 %) was observed between distance from Turów and average RWI for the 11-year period (1975–1985) around the year of lowest RWI values (Fig. 4b). Regression analyses were also carried out in order to determine whether any relationship existed between RWI in 1980 and site elevation, mean RWI for the 1975–1985 period and site elevation, or site elevation and distance from Turów. None of these regression analyses produced any significant relationship.

3.2 Correlation Response Function Analysis and Growth Modelling

The correlation response function analysis (1940–1974; Fig. 5 and Table 2) showed that for all sites response to temperature was greatest in the summer months, during

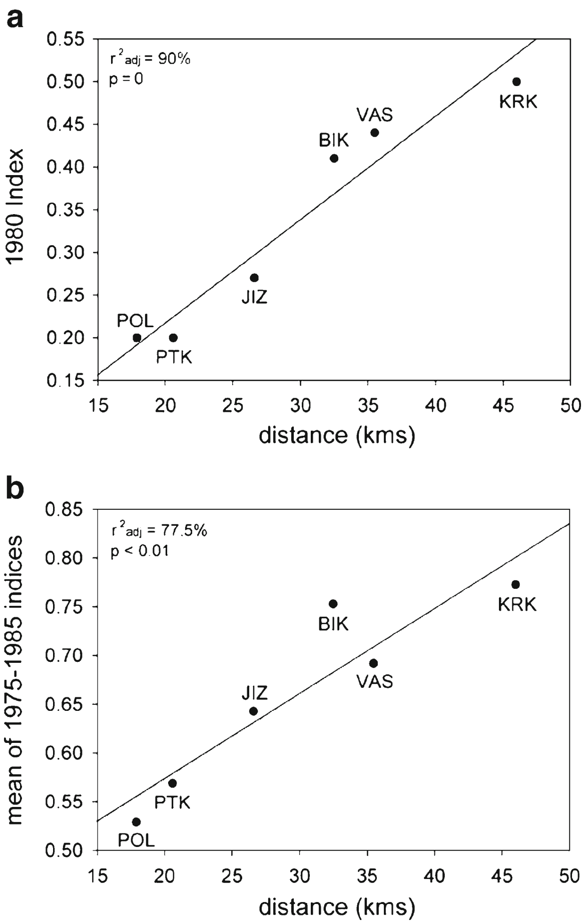

Fig. 4 Regression of RWI and distance of sampling sites from Turów power station. **a** 1980 growth index value and distance; **b** 1975–1985 average growth index and distance

the main growing season. The response, however, with regards to individual months was variable between sites. Results for KRK showed the strongest response for April–July (Table 2), while VAS and FAL correlated best with the April–August period. The BIK and JIZ sites both exhibited the highest correlations from May to July and the best results for PTK and POL were achieved with only May and June. Although the response at BIK was much weaker compared to the other sites, the May–July season was used for CGM as it was only just beneath the 95 % confidence level and still proved useful for the purposes of this study. In comparison to temperature, the influence of precipitation on growth was minimal. Only four correlations between monthly precipitation and RWI showed a significant (95 % CL) relationship. These included correlations between July and PTK, July and POL, February and KRK, and a negative correlation between January precipitation and BIK.

Although the time-series expressed site-specific influences, the detrended chronologies displayed several common features (Fig. 6). The most prominent feature was the sudden RWI suppression in 1980 visible in all Norway spruce chronologies. Although not as prominent, the 1980 dip was also present in the Silver fir (FAL) chronology. In general, however, FAL was substantially different to the other series. The FAL chronology showed distinctly low RWI values (e.g. 1940, 1956 and 1974), which were not as distinctive in the Norway spruce chronologies. In contrast to the spruce sites, FAL also showed a steady dip centred around the late 1970s, with 1974 being the lowest index value in the chronology and the 1980 minimum appearing on average as the fourth thinnest ring.

In the decade prior to 1980, decreasing RWI values were also present in PTK and POL, the two Norway spruce sites closest to Turów. Also, the dip during the period after 1979 at KRK did not appear to be as severe and protracted compared to the other sites. All sites had relatively inflated RWI values to a varying extent in the late 1990s and post-2000 period. There was also a period of unusually high RWI values around 1958–1979 in the BIK site, which was not apparent in any of the other chronologies.

Calibration of the CGM was performed for the 1940–1974 period (Fig. 6a) and RWI predictions over the 1975–2005 period. In addition to seasonal temperature, monthly precipitation was also included in the development of CGM models for sites where it was identified as significant (Fig. 5, Table 2). The competence of the CGM calibration models (Fig. 6a) ranged from an r_{adj}^2 of 13.7 % at BIK to 41.3 % at KRK, although all models were significant at the 95 % confidence level (Table 3). Over the prediction (verification) period, r_{adj}^2 results ranged from 0 % at KRK (note, however, that the KRK chronology ends in 1989) to 32.9 % at FAL. The model r_{adj}^2 values decreased for nearly all sites in the prediction period compared to the calibration period, and there were obvious misfits in trends between the actual and predicted data. Interestingly, the only exception was FAL, which exhibited marginal improvement

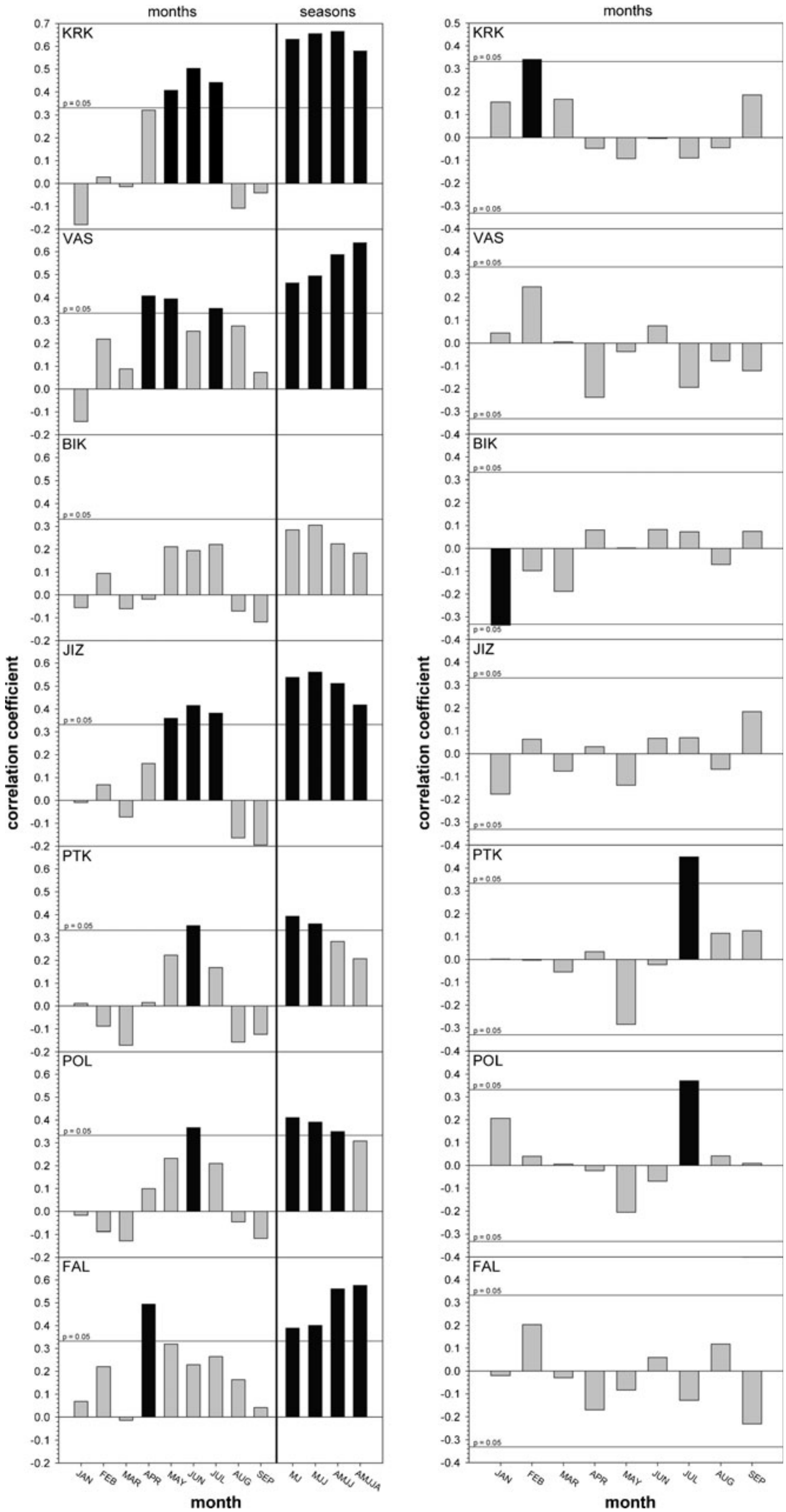

Fig. 5 Correlation response function analysis results with temperature (*left column*) and precipitation (*right column*) for all sites for the 1940–1974 period. Correlation coefficients are represented as *bars* with correlations exceeding the 95 % confidence level shown in *black* and correlations lower than the 95 % confidence level in *grey*. Optimal season correlations for temperature are listed in Table 2. Note that since tree growth in a given year cannot be influenced by any factor after the growing season, October–December are excluded from the analysis

Table 2 Summary of the optimal season and correlation results for each site

Site	KRK	VAS	BIK	JIZ	PTK	POL	FAL
Season (temperature)	Apr–Jul	Apr–Aug	May–Jul	May–Jul	May, Jun	May, Jun	Apr–Aug
r	0.67	0.64	0.28	0.56	0.39	0.41	0.58
Significant precipitation	Feb	–	Jan	–	Jul	Jul	–
r	0.34	–	−0.34	–	0.45	0.37	–

Correlations are between mean monthly temperature data averaged for the months included and RWI. Months with significant precipitation which are included in a multiple regression in the CGM and CSGM are also displayed for each site along with the respective correlations

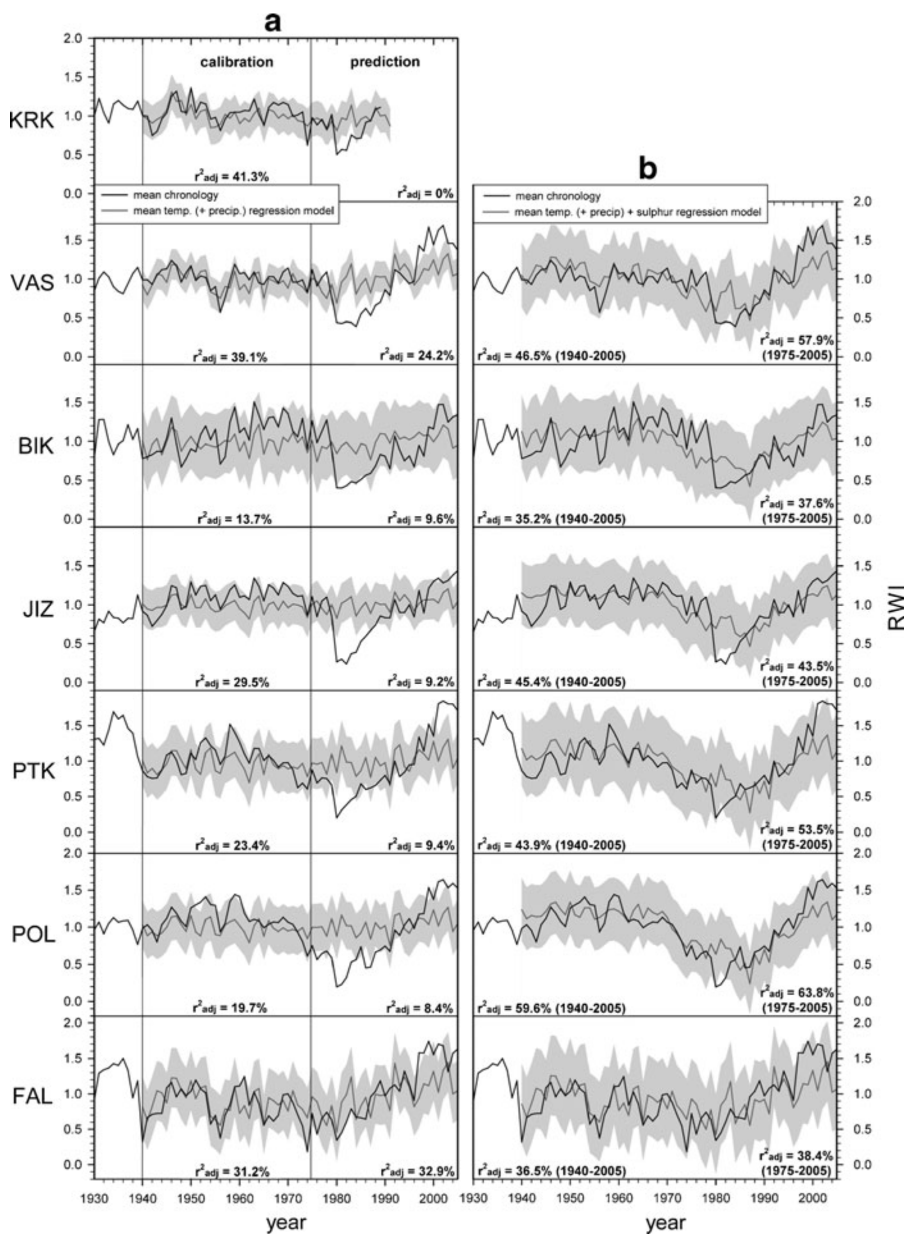

Fig. 6 RWI chronologies of all sites showing **a** CGM from 1940 until 2005, calibration period for the model between 1940 and 1974, prediction period between 1975 and 2005 and explained variance between the CGM model and RWI during each period. **b** CSGM in the 1940 to 2005 period and explained variance with RWI during the 1940 to 2005 and 1975 to 2005 periods. Note that the KRK chronology only extends until 1989 and was excluded from **b**. Two SE (standard error) *error bars are included as shaded area* with the model output

Table 3 Statistics of CGM and CSGM modelling

Site	CGM (1940–1974)	CGM (1975–2005[a])	CSGM (1975–2005[a])
KRK	$KRK=-0.265+0.0946\times T_{AMJJ}+0.0335\times P_{FEB}$		–
	$r=0.669$; $r_{adj}^2=0.413$	$r=0.212$; $r_{adj}^2=0$	
	$p<0.001$	$p=0.413$	
VAS	$VAS=-1.23+0.165\times T_{AMJJA}$		$VAS=-0.831+0.144\times T_{AMJJA}-0.00596\times S$
	$r=0.639$; $r_{adj}^2=0.391$	$r=0.517$; $r_{adj}^2=0.242$	$r=0.779$; $r_{adj}^2=0.579$
	$p<0.001$	$p=0.003$	$p<0.001$
BIK	$BIK=-0.091+0.0854\times T_{MJJ}-0.0667\times P_{JAN}$		$BIK=0.276+0.0668\times T_{MJJ}-0.0564\times P_{JAN}-0.00609\times S$
	$r=0.433$; $r_{adj}^2=0.137$	$r=0.396$; $r_{adj}^2=0.096$	$r=0.662$; $r_{adj}^2=0.376$
	$p=0.009$	$p=0.027$	$p<0.001$
JIZ	$JIZ=-0.282+0.0882\times T_{MJJ}$		$JIZ=0.155+0.0680\times T_{MJJ}-0.00714\times S$
	$r=0.562$; $r_{adj}^2=0.295$	$r=0.349$; $r_{adj}^2=0.092$	$r=0.688$; $r_{adj}^2=0.435$
	$p<0.001$	$p=0.054$	$p<0.001$
PTK	$PTK=-0.628+0.112\times T_{MJ}+0.0281\times P_{JUL}$		$PTK=-0.228+0.0979\times T_{MJ}+0.0171\times P_{JUL}-0.00831\times S$
	$r=0.528$; $r_{adj}^2=0.234$	$r=0.393$; $r_{adj}^2=0.094$	$r=0.763$; $r_{adj}^2=0.535$
	$p=0.001$	$p=0.029$	$p<0.001$
POL	$POL=-0.252+0.0895\times T_{MJ}+0.0164\times P_{JUL}$		$POL=0.235+0.0719\times T_{MJ}+0.0029\times P_{JUL}-0.0101\times S$
	$r=0.494$; $r_{adj}^2=0.197$	$r=0.380$; $r_{adj}^2=0.084$	$r=0.821$; $r_{adj}^2=0.638$
	$p=0.003$	$p=0.035$	$p<0.001$
FAL	$FAL=-2.49+0.254\times T_{AMJJA}$		$FAL=-2.37+0.248\times T_{AMJJA}-0.00185\times S$
	$r=0.576$; $r_{adj}^2=0.312$	$r=0.593$; $r_{adj}^2=0.329$	$r=0.652$; $r_{adj}^2=0.384$
	$p<0.001$	$p<0.001$	$p<0.001$

In the regression equation, T, P and S represent temperature, precipitation and SO_2, respectively. Correlations and r^2 are between the CGM vs. RWI and CSGM vs. RWI

[a] The KRK chronology only extends until 1989 which is therefore the final year included in the CGM calculations. The KRK data were also excluded from CSGM analysis

in the predicted period. With the inclusion of SO_2 concentration data in the CSGM (Fig. 6b), the modelling for all sites (significant at the 95 % confidence limit, Table 3) showed a marked improvement in agreement between the model predicted values and actual RWI values. For example, the r_{adj}^2 values in the 1975–2005 period were >37 % at all sites and >53 % at VAS, PTK and POL. This improvement is further confirmed by RMSE results for the two model types during the same period (Table 4).

4 Discussion

The results presented above give an indication of the spatial influence of SO_2 pollution upon forest stands in the Jizerské Mountains during recent decades. All of the chronologies (Fig. 6a) showed a set of years with distinctly low growth from the late 1970s to the 1990s. Significant divergence of RWI from the CGM was not apparent in the Norway spruce sites until 1980 when an exceptionally pronounced and rapid decrease in growth was observed at all sites followed by a series

Table 4 Root mean squared error values for CGM and CSGM models over the 1975–2005 period

	VAS	BIK	JIZ	PTK	POL	FAL
RMSE $CGM_{(1975–2005)}$	0.349	0.318	0.343	0.436	0.419	0.343
RMSE $CSGM_{(1975–2005)}$	0.272	0.246	0.250	0.331	0.262	0.333

The KRK chronology is excluded due to its limited temporal extent of years of suppressed growth. Perhaps the only exception to the above pattern were the CGM results for the PTK and POL chronologies (Fig. 6a), which were closest to Turów and appeared to begin experiencing decreased growth during the 1970s before the 1980 decline. From a spatial perspective, because a gradual decline in RWI was observed only in the two Norway spruce chronologies from sites in close proximity to the power station, while sites farther away showed little or no growth suppression before 1980, it is hypothesised that the differing reaction was the consequence of lower exposure to SO_2 with increasing distance. The results (Fig. 4a, b) support such a conclusion and demonstrate that lower RWIs were observed at sites closest to the Turów power station, becoming less suppressed with increasing distance from it. A decreasing influence of pollution with distance would also explain the less pronounced RWI suppression at the KRK site compared to the other Norway spruce sites (Figs. 6a and 4a, b). The substantial improvement in the models' predictive capability at all Norway spruce sites observed with the inclusion of SO_2 concentration data in the CSGM models (Fig. 6b) provides further evidence that SO_2 pollution was an important factor affecting growth post-1974.

Arguably, the influence of climate on growth alone could not result in such pronounced and long-term suppression of growth observed in the chronologies, since even exceptionally unfavourable conditions, as in 1956, led only to low growth rates lasting no more than 1 year. Concentrations of SO_2 exceeding 50 $\mu g/m^3$ over longer periods, however, have been associated with a reduction in ring width (Wilczyński 2006), and SO_2 concentrations from the Frýdlant measuring station (Fig. 1) indicated that SO_2 concentrations remained above 50 $\mu g/m^3$ from 1977 until 1991 with the exception of 1978–1980, 1982 and 1984 (Fig. 2b). However, with rising SO_2 (and NO_x) concentrations, the increasingly stressed stands may have become more susceptible to disturbances such as pathogen and insect attacks or climatic extremes (Price et al. 2011; Žid and Čermák 2007) and such disturbances may have acted as triggers initiating decline. Herein, we hypothesise a final 'trigger' of climatic origin which pushed the trees into the severe 1980s decline due to the large spatial extent of the suppression and short period in which the onset of growth suppression occurred.

Previous research in the region also identified the sharp decline in forest vitality, along with a sudden defoliation event in the Giant Mountains near the KRK site (Fig. 1) (Vacek and Lepš 1996). The study suggested that defoliation and death of trees in the 1980/1981 winter were, in addition to high pollution levels, triggered by a sudden temperature decrease from 10 to −20 °C over a 15-h period during the 1978/1979 winter followed by a steady rate of defoliation until 1988 with greater impact with increasing elevation. Kroupová (2002) suggested that high pollution levels, several unfavourable growth years and an exceptionally cold growing season in 1980 initiated the observed growth decline and temperature variability during the 1980/1981 winter and heavy frost in January of 1982 protracted the suppressed period.

The CGM and CSGM models provided insight into the relative significance of climate and SO_2 and allowed for a clearer understanding of the environmental changes which have affected tree growth in the Jizerské Mountains. Qualitatively, there was a clear breakdown of RWI response to climate after ca. 1980 (Fig. 6a) as divergence between CGM predictions and actual RWI becomes apparent. Additionally, there was generally a decrease in explained variance between CGM and RWI at most sites during the 1975–2005 period in comparison to 1940–1974. The sudden onset of growth suppression at all Norway spruce sites was also evidenced by the pattern of missing rings (Fig. 3), which showed a sharp transition in 1980 when the number of missing rings increases markedly compared to earlier decades.

An unexpected deviation from the general trend of increasing average RWI with distance from Turów between 1975 and 1985 (Fig. 4b) was the lower average RWI in the VAS chronology compared to BIK. Although site conditions may have differed in the past, BIK was considerably more open, while VAS was located in the middle of a forest, where stand dynamics may play a greater role. This factor could have affected how Norway spruce trees reacted to increased SO_2 pollution. The interplay of positive 'sheltering' effects of closed canopies and negative effects of competition with neighbouring trees is important in denser stands compared to more open and less competitive conditions of sites with lower tree densities (Vacek and Lepš 1996). Vacek and Lepš (1996) demonstrated that impact of atmospheric pollution was greater at stands with a greater tree density. This may be due to the additional stress of competition with neighbouring trees. Although a 'sheltering' effect may be considerable, gradual defoliation of the canopy would diminish the effect. This observation agrees with the findings of Slodičák and Novák (2005), who suggested that thinning is beneficial in stands under stress from atmospheric pollution. Although greater competition at VAS is a possible explanation, slightly higher average growth rates at BIK (mean RW= 2.36 mm) compared to VAS (mean RW=2.20 mm) may also explain the anomaly.

The period of above-average RWI between the end of the 1950s and end of the 1970s observed in the BIK chronology (Fig. 6a) was not present in any other chronology. Although abnormally increased growth has been cited as the reaction of Norway spruce to soil damage by excess nitrogen deposits (Fürst et al. 2005), a similar reaction would be expected at the other sites and the anomalous event begins before the construction of the Turów power station. However, there is a possibility that it was a reaction to regional pollution and increased susceptibility of the trees at BIK related to local conditions. Alternatively, some other site-specific factor may have influenced growth rates.

The results (Fig. 6) suggest that contrary to the reaction of Norway spruce, the Silver fir chronology (FAL) retained its climate signal in the 1975–2005 period and that the inclusion of SO_2 in the model enhanced the model's competence only modestly. Interestingly, the pattern of strong 1980s growth suppression seen in Norway spruce chronologies was absent from the Silver fir chronology, despite the species frequently described as being more sensitive to pollution than Norway spruce (Elling 2001; Wilson and Elling 2004). While the influence of pollution may have been less severe at this less exposed, lower elevation site despite its close proximity to the power station, perhaps a more likely explanation is that calibration of the CGM was partly undertaken during the period when the regional SO_2 concentration was already elevated and Turów was already emitting SO_2 pollution. Arguably, because growth-climate modelling was carried out in the interval when SO_2 pollution was already affecting and perhaps suppressing growth, the longer term suppression would not be obvious since the model was only operating in a relatively short temporal window without fully expressing lower frequency trends.

Elling et al. (2009) attributed increased susceptibility of Silver fir to winter frost and summer drought in southern Germany to elevated SO_2 pollution levels and found a highly significant correlation between missing rings and SO_2 loads. The results of this study are consistent with their findings. The pattern of missing rings (Fig. 3) mirrors atmospheric SO_2 concentrations in the area (Fig. 2). In fact, a significant correlation (r=0.392, p=0.013) between the regional SO_2 index (Fig. 2—top) and the FAL missing rings data was identified during the 1969–2007 period. Additionally, a very strong correlation (r=0.916; p< 0.001) was observed between the country-wide historical sulphur data for Poland and the regional index in the 1969–1990 period of overlap. This high level of agreement enabled the comparison of missing rings at FAL with the longer Poland sulphur series as an extension of the regional series. The high correlation (r= 0.720, p<0.001) detected in the 1914–1990 period suggested that sulphur emissions had already been affecting growth at FAL before the construction of Turów in 1962.

A further consideration is that the RWI variance for FAL was greater compared to the two Norway spruce and the other Silver fir chronologies from the Bavarian Forest (Fig. 7), particularly considering the extremely low index values (e.g. 1940, 1956, 1974 and 1980). Arguably, although the influence of SO_2 pollution was not manifested in the same way as it was at the Norway spruce sites and the response to temperature was retained, the above results indicate the significant influence of pollution on fir growth at FAL in the pattern of missing rings, their agreement with sulphur levels since the beginning of the twentieth century and exceptionally low RWI values in climatically stressful years, resulting in a long-term impact on growth and increased sensitivity to climatic extremes. The above discussion highlights fundamental differences in the way Norway spruce and Silver fir respond to increased atmospheric SO_2 concentrations. Whether these relationships are only true under particular environmental conditions or a specific degree of SO_2 loading, or applies more generally, remains unclear.

The study's regional composite Norway spruce and Silver fir chronologies were compared to similar composite series from the Bavarian Forest in southeast Germany (Fig. 7), another area affected by atmospheric pollution in the past. Focussing only on chronologies exhibiting a marked period of growth

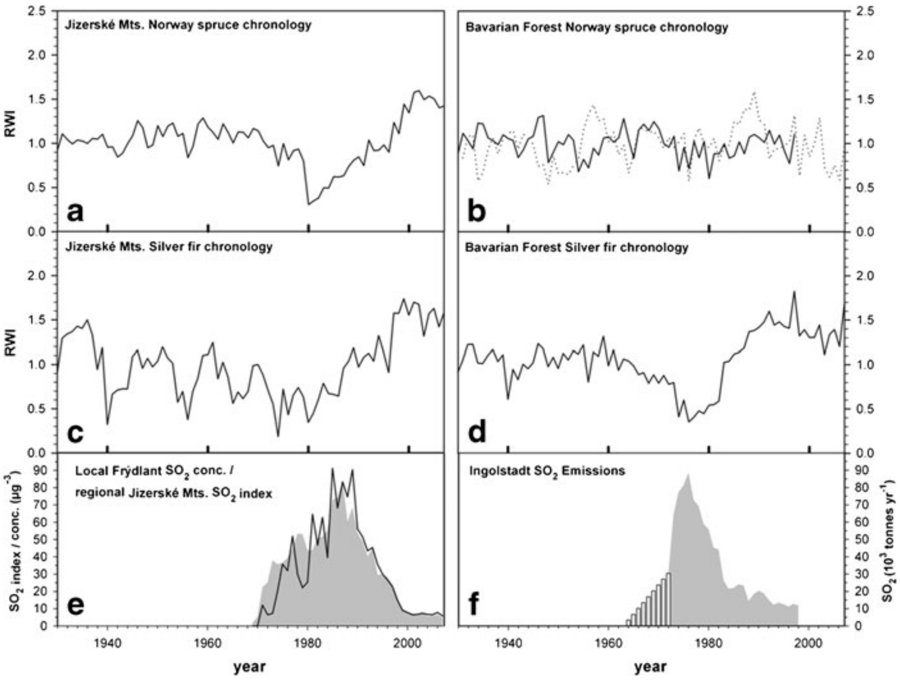

Fig. 7 Comparison of Norway spruce and Silver fir chronologies and SO_2 data. The Czech spruce chronology (**a**) is composed of all Norway spruce data from sites VAS, BIK, JIZ, PTK and POL, while the Czech fir chronology (**c**) includes data from FAL. Note that two separate chronologies in **b** represent low (*dotted* <700 m) and high (*line* >1,100 m) elevation Norway spruce from the Bavarian Forest (see Wilson and Hopfmüller 2001); **d** composite Silver fir chronology (Wilson and Elling 2004) from the Bavarian Forest in Germany; **e** atmospheric SO_2 concentrations from the Frýdlant measuring station (*shaded area*—same as in Fig. 2—*top*) and regional Jizerské Mountains atmospheric SO_2 concentration index (*black line*); **f** emissions of SO_2 from oil refineries near Ingolstadt west of the Bavarian Forest (*white bars* represent linear interpolation of emissions between commencement of industrial production and the first measured value)

suppression (Fig. 7a, d) and SO_2 emission/atmospheric concentration data (Fig. 7e, f), one of the most intriguing observations is the asynchronous nature of the responses in the two regions (i.e. peak pollution and reduction occurred earlier in SE Germany). Although both areas are approximately 250 km apart and exposed to similar climatic conditions, the difference in timing of growth reduction onset and duration suggest that differing regimes in pollution emissions were the primary cause of the growth reductions.

The difference in reaction of both tree species at the two locations is clear. Although SO_2 emission and concentration data offered some explanation for the observed patterns, the influence of additional factors, not addressed here, undoubtedly results in a far from clear-cut story. We offer some possible explanations for the differences in response. Wilson and Elling (2004) identified that the Bavarian Forest Silver fir trees (Fig. 7d) responded most strongly to spring/summer precipitation, whereas the Silver fir in this study responded to temperature. The Bavarian Forest Silver fir sites extended from 420 to 885 m (Wilson and Elling 2004: Fig. 1), while the FAL site was located 380 m asl. (Table 1) suggesting that the response of Silver fir to climate is complex and may not follow the 'classic' picture of moisture stress at low elevations and temperature limitation at higher elevations (Wilson and Hopfmueller 2001). Differences also exist between the response of Jizerské Mountains and Bavarian Forest Norway spruce. Differences in elevation may also have been a factor. The Bavarian Forest Norway spruce were split into high (>1,100 m) and low (<700 m) elevation chronologies (Fig. 7b), while the Jizerské Mountains sites were located at elevations from 820 to 1,080 m (Table 1). The Bavarian high elevation spruce chronologies responded most strongly to summer temperatures, while the low elevation sites responded to spring/summer precipitation (Wilson and Hopfmueller 2001), and the influence of SO_2 emissions on growth was only noted as a weakening in response to climate for the low elevation chronologies (Wilson and Elling 2004).

While a direct comparison of the respective degree of atmospheric pollution in the two regions was not possible (Fig. 7e, f), we argue that the impact on forest stands was greater in the Jizerské Mountains for two reasons. Firstly, the observed degree of ring width suppression in Norway spruce from our study region (Fig. 7a) indicates the greater severity of the environmental impact compared to the Bavarian Forest data (Fig. 7b) (Wilson and Hopfmueller 2001; Wilson and Elling 2004). Secondly, the distance of spruce and fir sites from the emission source in the Bavarian Forest was substantially greater than in the Jizerské Mountains (Wilson and Elling 2004: Fig. 1 and Fig. 1 herein).

In our study, the rapid recovery of tree growth following the reduction of SO_2 emissions and decline of atmospheric SO_2 concentrations during the 1990s indicated the link between growth suppression and the direct impact of atmospheric pollution (see also Wilson and Elling 2004; Elling et al. 2009). Although growth had recovered, RWI and temperature did not correlate in the 1992–2005 period (results not shown), despite falling SO_2 levels, indicating that the tree growth response had not yet returned to the state prior to suppression. Instead, a period of abnormally high growth rates was observed, which may be attributed to several factors including (1) detrending artefacts despite minimising index inflation effects by utilising the adaptive power transformation prior to detrending (Cook and Peters 1997), (2) a response to increased nitrogen deposition and higher atmospheric CO_2 concentrations and (3) a more open canopy and decreased competition resulting from mortality of trees nearby (Dittmar and Elling 2004; Elling et al. 2009).

Wilczyński (2006) stated that recovery following a period of growth suppression is stronger in trees which experienced a greater degree of stress. Assuming that the number and temporal extent of missing rings is an indication of stress and growth suppression, this is in line with the observation that enhanced growth during the years following the reduction of SO_2 concentrations was greatest at FAL, where the influence of SO_2 on growth lasted for a longer period than at the Norway spruce sites. Wilson and Elling (2004) suggested opening of the canopy due to tree mortality as one of the possible causes of enhanced radial increment growth in Silver fir following the reduction of atmospheric SO_2 concentrations in the Bavarian Forest. In our study, the enhanced growth at FAL was unlikely to have been caused by decreased competitive stress since the site was located in the middle of a dense, closed canopy forest, where little change in stand dynamics was likely to have occurred because the Silver fir trees were growing in the middle of a forest of less pollution-sensitive Norway spruce. However, the possibility of a difference in physiological response to increased levels of SO_2 pollution and the subsequent decrease of pollution levels between Norway spruce and Silver fir must also be considered.

Lastly, it should be noted that this investigation focussed on a single point source of SO_2 pollution. We acknowledge that it was not possible to attribute the influence of SO_2 pollution on tree growth solely to emissions from the Turów power station since several other power stations and industrial emission sources were in operation farther west of the studied region during the second half of the twentieth century.

5 Conclusion

The evidence presented in this study clearly demonstrates that the effect of SO_2 pollution on tree growth declines with distance from a point source of pollution. Although a sudden and protracted growth reduction was identified from the tree ring records of Norway spruce, it was not possible to attribute such an unprecedented growth reduction to the influence of pollution or climate alone. Instead, a combination of damagingly high pollution levels and a series of unfavourable climatic conditions interacted to produce abnormally low growth rates.

The inclusion of atmospheric SO_2 concentration data in the CGM during the 1975–2005 period resulted in a significant improvement of RWI prediction. The same period coincided with the period of highest atmospheric pollution concentrations followed by a recovery in growth after the decrease in pollution levels, indicating that atmospheric SO_2 had a profound influence on growth. The reaction of Silver fir to the effects of pollution differed significantly from that of Norway spruce. Despite retaining sensitivity to temperature variability, the pattern of missing rings, their relationship to atmospheric SO_2 concentrations and increased susceptibility to climatic extremes revealed a significant influence of SO_2 pollution showing a longer lasting reaction to pollution with an earlier onset compared to Norway spruce.

One significant observation was the lack of correlation between RWI and climate during the recent 1992–2005 period, which suggested that growth conditions have not yet returned to a state where temperature influences were the dominant determinant of growth rates, as was the case before the collapse of growth rates in the early 1980s. Regarding the results of this study more generally, although the influence of atmospheric SO_2 concentrations certainly helped explain the growth reaction of stands in the region, it must be acknowledged that the severity of the impact of pollution on the conifer woodlands along with a range of other additional factors resulted in a complex story. Longer term effects of high SO_2 and NO_x concentrations, such as soil acidification and nutrient imbalances, may mean that the health of forests in the Jizerské Mountains and the region as a whole is still in question.

On a final note, in dendroclimatology, the issue of 'divergence' has widely been discussed, resulting in speculation about possible causes (D'Arrigo et al. 2008). Our results pointed to the weakening of the climate signal in the study region and suggested that this was related to the impact of high atmospheric pollution levels on growth and subsequent environmental alterations following the recent reduction of pollution emissions. Regarding dendroclimatological reconstruction efforts, and particularly considering dendroclimatic calibration, we caution that climate reconstruction attempts in affected parts of central Europe (e.g. Büntgen et al. 2011) and other regions that are experiencing or have experienced high atmospheric pollution levels in the past (Wilson et al. 2012) may be hindered by this influence. Additionally, as was documented by Lloyd and Bunn (2007) and Savva and Berninger (2010), the issue may be of relevance to a wider range of locations than previously believed, including those away from major sources of pollution that have in the past been considered unaffected.

Acknowledgments The authors wish to thank Dr. Jana Nováková and Dr. Josef Křeček for their cooperation and provision of useful information related to the study area and fieldwork logistics. We also thank Jana Rydvalová and Ing. Miloš Rydval Sr. for their assistance during fieldwork. Finally, we also wish to acknowledge the help of Oldřich Kober, who proved to be a valuable source of local knowledge.

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