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The influence of O_3, NO_2 and SO_2 on growth of *Picea abies* and *Fagus sylvatica* in the Carpathian Mountains

R.M. Muzika[a,*], R.P. Guyette[a], T. Zielonka[b], A.M. Liebhold[c]

[a]*Department of Forestry, University of Missouri, Columbia, MO 65211, USA*
[b]*Institute of Botany, Polish Academy of Sciences, Krakow, Poland*
[c]*USDA Forest Service Northeastern Research Station, Morgantown, WV 26505, USA*

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"Capsule": *Trends in tree growth rates can be explained by long-term effects of air pollution.*

Abstract

At 17 long-term pollution monitoring sites throughout the Carpathian Mountains, tree growth patterns and variation in growth rate were examined to determine relationship of tree growth to specific pollutants. Canopy dominant *Picea abies* and *Fagus sylvatica* were selected at each site. Basal area increment (BAI) values were calculated from raw ring widths and used as an estimate of tree growth. Across all sites, BAI chronologies were highly variable, therefore local conditions and forest structure accounted for considerable variation. Several significant relationships, however, implicated a role of pollutants on tree growth. Average levels (1997–1999) of NO_2 and SO_2 were inversely related to BAI means (1989–1999). Although average O_3 alone was not related to growth, the maximum O_3 value reported at the sites was negatively correlated with overall growth. A variable representing the combined effect of O_3, NO_2 and SO_2 was negatively correlated with both *P. abies* and *F. sylvatica* growth. Pollution data were used to categorize all sites into 'high' or 'low' pollution sites. Difference chronologies based on these categories indicated trends of decline in the 'high' pollution sites relative to 'low' pollution site. In the more heavily polluted sites, the BAI of *Fagus sylvatica* has declined approximately 50% and *Picea abies* has declined 20% over the past 45 years.

Keywords: Dendrochronology; Basal area increment; Carpathian Mountains; Spruce; Beech; Air pollution

1. Introduction

The overall effect of atmospheric pollutants on individual tree growth and vigor as well as forest-wide patterns of growth has been evaluated in many forest types across the globe. Dendrochronology has been used to examine specific effects of pollutants, to identify particular episodes of pollution events, and to examine the correspondence between ring width and pollution. Research has examined both the effects of point source pollutants as well as identifying causes of regional growth declines. In Eastern North America, consistent relationships between atmospheric pollutants and tree growth have not been documented extensively, but results suggest that atmospheric pollutants represent one of many causal factors related to forest decline (Cogbill, 1977; LeBlanc, 1992). Acid deposition and ozone have been implicated as having an adverse effect on the growth of *Picea rubens* (McLaughlin and Kohut, 1992). In western North America, high levels of ozone have been related to prolonged growth declines in conifers (Peterson et al., 1991).

The effects of air pollution on tree growth and as a contributor to forest decline in east-central Europe has been documented over recent decades (Schulze, 1989; Grodzinska et al., 1990; Petráš et al., 1993). There is growing evidence to suggest that the production of airborne pollutants from many parts of Europe may be deposited in the Carpathian Mountains and that these materials may be adversely affecting vegetation in the area (Dovland, 1987; Materna, 1989; Rzychon and Worsztynowicz, 1995). Ozone and other pollutants have been recognized as a contributor, if not a suggested

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* Corresponding author.
E-mail address: muzika@missouri.edu (R.M. Muzika).

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cause of forest declines in several areas (Krupa and Manning, 1988), including the Carpathian Mountains (Bytnerowicz et al., 1993). Recent research suggests that phytotoxic levels of O_3 are evident throughout the Carpathian Mountain Region (Bytnerowicz et al., 2002). The effects of pollutants on forest declines are often subtle and difficult to detect (Cogbill, 1977; LeBlanc, 1992); but documenting these effects will increase an understanding of causal agents and mechanisms.

Recent summaries, e.g., the Environmental Monitoring European Programme (EMEP) data indicate that levels of pollutants in Slovakia, Poland and the Czech Republic are still among the highest in Europe. Critical levels of gaseous pollutants (O_3, SO_2, NO_x) and total critical load by sulfur and nitrogen have been largely exceeded in major portions of those countries (Posch et al., 1997). Pollution reduction efforts are in place, but with great variation in the results. Romania, Poland, Slovakia and the Czech Republic have reduced sulfur emissions by 14, 58, 78 and 88%, respectively, as a percentage of 1980 levels (UNECE, 1999). Despite overall reductions, transboundary pollution will likely remain significant as long as there are point sources, since emissions can persist in the air chemically unchanged for 1–3 days, move with the air masses and affect areas relatively far from the place of origin (Dovland, 1987).

Reduction in tree growth, insect and pathogen attacks, and tree mortality may all play important roles in the underlying long-term dynamic patterns of forest succession and regeneration; consequently separation of forest declines due to anthropogenic causes as opposed to natural processes may be difficult, but critical. Long-term data are essential for differentiating changes in growth and mortality due to natural processes from the cumulative effects of pollution on trees and soil. Dendrochronological approaches represent one of the best ways to acquire long-term, high-resolution data on individual and stand-wide tree growth.

This study focuses on the radial growth response of the dominant overstory trees to the measured levels of pollutants at 17 long-term monitoring sites. This research complements, and indeed is dependent on, research conducted at the air pollution monitoring sites in the Carpathian Mountains (Szaro et al., 2002). The primary objective is to ascertain if an association exists between air pollution levels and the growth of the two predominant overstory tree species, *P. abies* and *Fagus sylvatica*, throughout the region.

2. Methods

2.1. Study areas

This study represents a portion of a larger project in which 32 forested sites were used to monitor pollutants in the Carpathian Mountains. The region includes portions of the Czech Republic, Poland, Slovakia, Ukraine and Romania, and the 32 air pollution monitoring sites were distributed throughout the region (Bytnenowicz et al., 2002). Owing to the intensity of sampling required for dendrochronological analysis, a subset of 17 from the 32 sites was chosen for this project. At a minimum two sites per country were selected (Table 1). The selections were chosen to include an equivalent number of sites sampled for *F. sylvatica* and *P. abies*, so preliminary

Table 1
Study locations for air pollution monitoring and dendrochronological sampling

Country	Site name	Species sampled	Altitude (m)
Czech Republic	Bílý Kříž	*Picea*	890
	Javorina	*Fagus*	900
Poland	Brenna	*Fagus*	600
	Tatra National Park	*Picea*	900
	Magura National Park	*Fagus*	540
	Bieszczady National Park	*Picea*	920
Slovakia	Male Karpaty	*Fagus*	686
	Malá Fatra	*Fagus*	690
	Polana	*Fagus* and *Picea*	987
	Novaveská Huta	*Picea*	954
Ukraine	Uzhoksky Pass	*Picea*	900
	Synevir	*Picea*	850
	Kuzij	*Fagus*	750
	Vizhnitsa	*Fagus*	705
Romania	Rarau	*Picea*	978
	Stana de Valle	*Fagus* and *Picea*	1270
	Fundata	*Fagus*	1321

vegetation analysis was necessary for site selection. At each site, at least 15 dominant or codominant trees were chosen for dendrochronological sampling and analysis. Sampled trees represented the species mix of the stand and were of fair or good crown condition. Within the above criteria, the trees were randomly chosen. The diameter (at 1.37 m) of each tree was measured; the vigor of the tree was noted, and two cores were extracted from each tree at 1.37 m height.

2.2. Dendrochronology measurements and data analysis

Increment cores were extracted, stored, dried and mounted. These were sanded and each ring measured to the nearest 0.01 mm using an electronic transducer and binocular scope fixed over a moving stage. Measurements were made at the University of Missouri's Tree Ring Laboratory. All resulting ring-width series from each sample's measurements were visually inspected, plotted and cross dated (Stokes and Smiley, 1968). Correlation analysis using the program COFECHA was used to ensure the accuracy of cross dating (Grissino-Mayer, 2001).

We converted ring-width series to basal area increment (BAI) as a means of adjusting for the effects of stem geometry on ring width. BAI also more closely approximates overall tree growth, and may be more appropriate than annual ring width to examine decline-related phenomena (LeBlanc, 1998) despite concerns about its applicability in all analyses (Biondi, 1999). We calculated BAI (the cross-sectional area of wood produced annually) from the raw ring-widths and measured diameters. A BAI chronology was calculated for each site by averaging the BAI for each year across all trees sampled at that site. Site chronologies included only the years with more than 10 individual tree ring series for all years included in the chronology. BAI values for *Fagus* were not determined from the two sites in Romania, Fundata and Stana de Valle. The diameters of these trees were not included during the initial data collection, and the condition of the cores precluded an accurate estimate of BAI.

2.3. Statistical treatment of data and time series development

We used correlation analysis to test and evaluate the relationship between recent radial growth, averaged for each site and species, and the pollution data from each site. Complete pollution data can be found in Bytnerowicz et al. (2002). Pollution data for the sites sampled were available for years 1997, 1998 and 1999 only. Growth variables included in the correlation analysis were derived from the BAI of the most recent 10 years growth (1990–1999). Although there is not a direct year-to-year correspondence of the pollution data and tree ring data, this study assumes that changes in pollution levels among sites have been relatively constant over the last ten years. Since 1989 air pollution levels within the region have declined relative to the previous decades, and evidence suggests that since 1989 variation has not been dramatic (Pavlinek, 2002; Carter and Kantowicz, 2002). In addition, the use of 10 years of BAI data minimizes variance due to the annual effects of climate. Ten-year BAI means were standardized by species by dividing by the average BAI for all sites to compensate for possible species differences in growth. These values were transformed using simple natural logarithm transformation. Studies have shown nonlinear relationships among pollution variables and tree growth (Alexeyev, 1989; Sverdrup and Warfvinge, 1993). Pollution variables used in the analysis included average O_3, maximum O_3, NO_2 and SO_2 average values for each site as well as transformations of the values. In addition, a combined pollution variable was created as the product of mean values of O_3, NO_2 and SO_2 for each site. Biochemical and physiological effects of each pollutant varies, as does the response of vegetation; however, synergistic effects of these three pollutants have been noted (Cowling and Heck, 1989). Specific and quantitative allocation of the effect of each pollutant is not possible; therefore a variable derived from the product of each will allow for equal weights of each pollutant and minimize biases from the raw data.

Based on the existing 1997–1999 site pollution data (Bytnerowicz et al., 2002), BAI chronologies were divided into two groups, high pollution and low pollution for each tree species. Each species/site combination BAI chronology was assigned to one of the two categories. In order to examine the annual values in the two groups, we created a difference chronology by dividing individual values (high pollution sites/low pollution sites) for each species. The period 1955–1999, common to all the chronologies, was used in the analysis. A third group of chronologies was created for all sites and species that included BAI chronologies derived from stands with trees that were older than 80 years. With this age criteria, a high pollution chronology was developed from four sites and a low pollution chronology from two sites. We compared chronologies using an 11-year moving average as a way to examine long-term trends and to reduce yearly variation from climate effects on ring width. A difference chronology was constructed from these extended BAI chronologies. Both *Fagus* and *Picea* were included in the high and low pollution chronologies.

3. Results

Results of the correlation analyses between the most recent 10-year BAI means from each site and pollution

Table 2
Correlations among growth variables and pollution variables

	Maximum O_3	Average O_3	SO_2	NO_2	Total pollution	Total pollution (*ln*)
BAI (both species)	−0.572(0.016)	−0.224(0.389)	−0.492(0.045)	−0.474(0.054)	−0.685(0.002)	−0.676(0.003)
Spruce BAI	−0.638(0.064)	−0.326(0.392)	−0.621(0.075)	−0.147(0.705)	−0.671(0.048)	−0.734(0.024)
Beech BAI	−0.544(0.163)	−0.172(0.684)	−0.551(0.157)	−0.621(0.100)	−0.752(0.032)	−0.678(0.064)
BAI (*ln*) (both species)	−0.547(0.023)	−0.163(0.531)	−0.474(0.054)	−0.587(0.013)	−0.676(0.003)	−0.705(0.001)
Spruce BAI (*ln*)	0.623(0.073)	−0.274(0.476)	−0.649(0.058)	−0.224(0.561)	−0.684(0.042)	−0.767(0.016)
Beech BAI (*ln*)	−0.524(0.182)	−0.137(0.746)	−0.582(0.129)	−0.659(0.075)	−0.771(0.025)	−0.712(0.048)

Correlation coefficients, *r*-values, are presented and *P* values are beneath coefficients. Significant r values ($\alpha = 0.05$) are in boldface type and represent individual tests of a priori hypotheses for known pollutants. See text for explanation of variables.

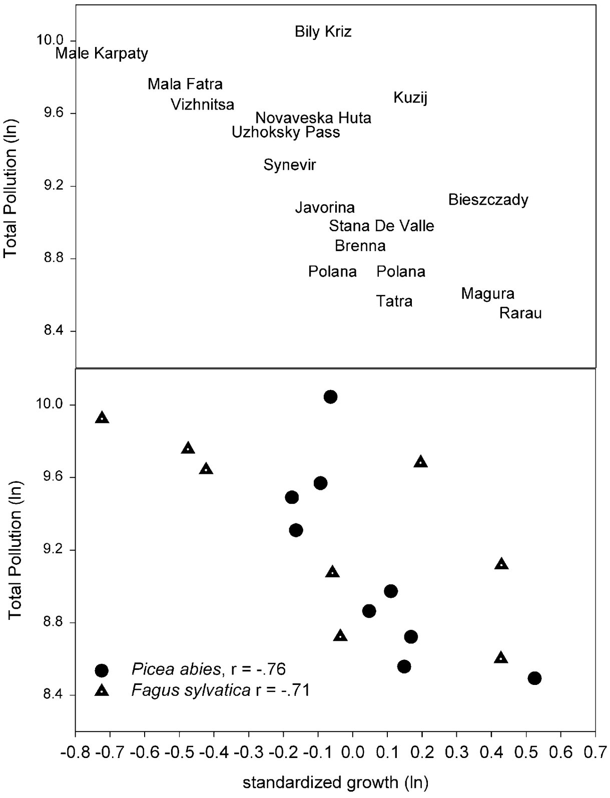

Fig. 1. (top) Study sites arrayed by the relationships between pollution and growth. See Table 1 for details of the site characteristics. (bottom) BAI (10 year mean) versus *ln* Pollution for all sites by species. Pollution variable represents the product of average O_3, average SO_2, and average NO_2, and was transformed using natural logarithm (*ln*).

variables reflect hypothesized relationships for the effects of pollution on tree growth, expressed as BAI (Table 2). Mean O_3 levels were not significantly correlated with growth. The maximum O_3 per site, however was negatively and significantly related to both raw BAI (growth) and *ln* BAI of the pooled species data. A strong negative, although not significant, relationship existed between maximum O_3 and spruce growth, both as BAI and *ln* BAI.

As anticipated, SO_2 was negatively related to overall BAI and negatively (although not significantly, $\alpha = 0.05$) to a transformed BAI value of *Picea*. NO_2 was significantly and negatively related only to *ln* BAI. The most complete relationship, however, became apparent by combining the pollution variables. All growth variables are significantly and strongly correlated with the combination pollution variable represented by the product of the mean levels of O_3, SO_2 and NO_2 (Table 2).

A visual examination of the associations between pollution and growth indicates strong site–level relationships (Fig. 1a). The correlation coefficient is high with either species (Fig. 1b), and indicates an inverse relationship between pollution and growth. With location names as identifiers (Fig. 1a), it is apparent that certain sites have high pollutant levels and correspondingly low growth, while others have low pollutants and high growth. For example, Magura (Poland) and Rarau (Romania) are clearly low pollution sites, where the tree growth is high, whereas the Slovakia sites of Male Kapaty and Malá Fatra had higher pollution values and lower growth. The geographic organization of the air pollution data corresponds with overall differences in forest health and defoliation (Badea et al., 2002). We divided the sites into low pollution sites and high pollution sites; the delineation was at the pollution index value of 9.2 (Fig. 1b). All sites in the Ukraine fell within the high pollution category as well as Bílý Kříž, Novoveská Huta, and Malá Fatra. In the low pollution category, all sites from Poland and Romania were included as well as both Polana sites (Slovakia) and Javorina (Czech Republic).

Using chronologies combined by pollution categories, and plotting overall growth by calendar year, growth patterns seem to have diverged in recent years (Fig. 2). These relatively long chronologies show similar trends before the 1980s, although absolute values are very

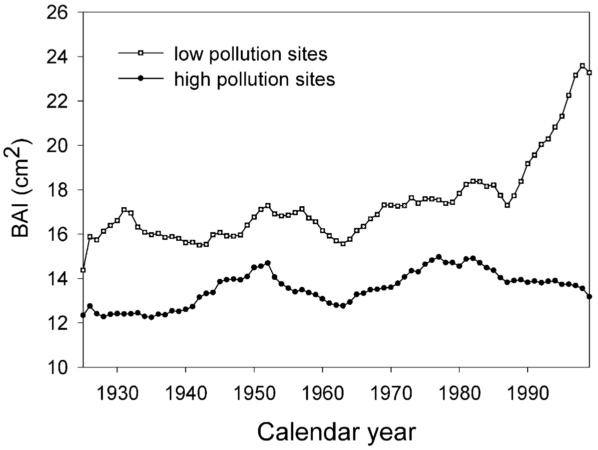

Fig. 2. Trends (11-year moving average) in basal area increment (BAI) for the longest available chronologies from sites with high and low levels of SO_2, NO_2, and O_3. Both *Fagus* and *Picea* chronologies are combined by high or low pollution.

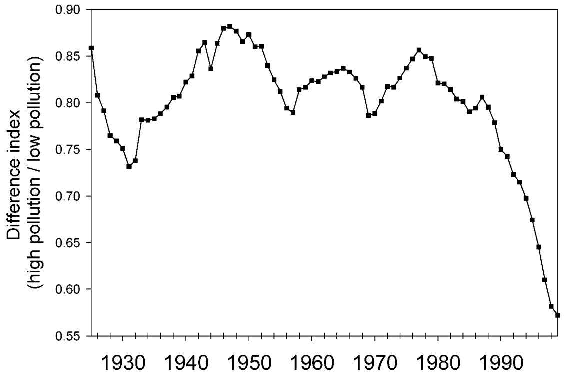

Fig. 3. A BAI difference index chronologies for the time period 1925–1999. The difference chronology represents a ratio of high pollution BAI:low pollution BAI. Pollution designation was based on data conducted in a related study (Bytnerowicz et al., 2002). Values lower than one represent low BAI values at high pollution sites relative to low pollution sites.

different. After 1980, the BAI value differed dramatically between the pollution types. At high pollution sites BAI remains unchanged or declines slightly while at low pollution sites BAI increased (Fig. 2). A difference index composed of both species confirms this by indication of a strong relative decline in growth beginning approximately 1980 (Fig. 3). Since the difference index represents a ratio of growth, the closer the difference index is to 1, the more similar growth is at the sites. With both species combined, it is apparent that growth was always lower at the high pollution sites throughout this approximately 70-year period. Clearly there was a very steady difference in the growth between high and low pollution types since approximately 1980.

The strong differences apparent in Fig. 3 can be further distinguished by species. Using an abbreviated, more recent chronology (Fig. 4), difference indices each of *Picea* and *Fagus* indicate that tree BAI at high pollution sites has declined over the past 45 years relative to those at low pollution sites. The BAI of *Fagus* has declined approximately 50% over the last 45 years while that of *Picea* has declined 20%.

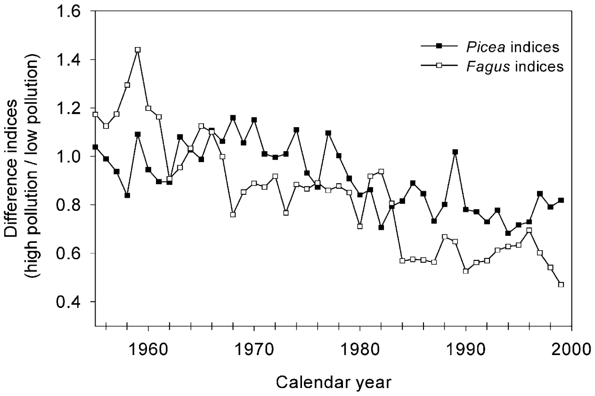

Fig. 4. BAI difference index from 1955 to 1999 for all sites combined, for *Fagus* and *Picea*, with trends illustrating the decline of BAI at sites with high levels of SO_2, NO_2, and O_3 versus those with low levels.

4. Discussion

The development and analysis of BAI difference indices by species documents trends and patterns that are consistent with spatial associations of pollution and growth data. Lower BAI over the past several decades, and relatively low 10-year BAI means were found at sites with high pollution. Thus, both BAI trend and absolute values are associated with pollution exposure. These results also indicate that although only three recent years of site level pollution data were used in this analysis, these data may represent spatial patterns in pollution that have occurred over the past several decades and represent the cumulative impacts of pollution to tree growth and soils over much longer durations.

Similar trends between high and low pollution with extended, combined species chronologies indicate that before the 1980s (Fig. 2) these sites were well matched and should serve well for a time series comparison. The break down of this similarity after the 1980s demonstrates that pollution or other factors that influence tree growth have changed at these sites. The pronounced difference in chronologies seems to be related to increased growth at the low pollution sites since the mid 1980s, rather than a decrease per se, at the high pollution sites. Although some pollutants have remained unchanged, there have been steady reductions in sulfur oxides emissions in Europe since 1980 (UNECE, 1999). These reductions, however, may have very little effect on the current overstory trees. Tree vigor and soils may have been adversely affected for many years and may likely not recover soon. As demonstrated in other studies, the effects of pollutants persist, even when active contributions of pollutants may have ceased (Cook and Innes, 1989; Carter and Kantowicz, 2002).

The use of a difference index representing a BAI ratio of high/low pollution sites indicated a notable decline. The greater the difference in polluted versus non-polluted sites, the more pronounced the decline. Across the Carpathian region, then, and when combining both species sampled, it is apparent that the difference has increased greater in the past two decades (Fig. 3). The dramatic decline is attributed to both species, but probably more strongly reflective of the decline in *Fagus* BAI (Fig. 4). Pre-1960 *Fagus* growth at what we have identified as high pollution sites, had exceeded the growth of *Fagus* at the low pollution sites. *Picea* growth appeared to be greater at the high pollution sites in the 1970s and the 1980s. There is minimal correspondence between BAI of these two species with the exception of similar direction of growth in the late 1950s. These species-specific trends likely reflect the species related tolerances to environmental conditions. Furthermore, the assigned high and low pollution labels to these sites represent the values of the recent monitoring activities. It is possible the pollution levels differed during the course of the chronology, particularly point-source pollutants, as well as the relative contributions of O_3, SO_2 and NO_2.

Different airborne pollutants may interact in a synergistic, multiplicative, or interactive way on vegetation. Synergistic effects of ozone, sulfur dioxide and nitrogen oxides have been previously documented, most clearly on agricultural crops (Cowling and Heck, 1989). The interaction of pollutants with each other as well as with abiotic factors has been documented, however, it is challenging to quantify (Andersen and Grulke, 2001; Takemoto et al., 2001). Recent research has shown that ozone predisposes trees to drought or cold stress (Maier-Maekcker, 1999), and that site related factors might exacerbate ozone effects (Bartholomay et al., 1997). The data in this analysis show a potential multiplicative effect in that the strongest correlations were with the products of the means of O_3, NO_2 and SO_2. However, it is important to note that there is likely colinearity among O_3, NO_2 and SO_2, therefore distinguishing unique relationships becomes difficult with such data.

5. Concluding remarks

The scientific consensus recognizes pollutants as predisposing or triggering factors to forest decline. Strong evidence exists from the deteriorating crown conditions on forest plots throughout east central Europe. Throughout much of this region of Europe, rates of SO_2 and NO_2 emission have decreased substantially since 1980 (UNECE, 1999), but there have been reported increases in O_3 (Klemme and Lange, 1999). Bytnerowicz et al. (2002) have shown that high concentrations of O_3 can be expected in the Carpathian Mountains, particularly with suitable local conditions. Concentrations set by the UN ECE convention as critical levels are often exceeded throughout the regions, e.g. (Godzik, 1998; Tschimala Mbuyi, 2001). Considerable effort has been made to identify targets for further reduction of pollutants as well. Nonetheless, the effect of air pollutants tend to be long-lasting and the reduction of emissions may have little immediate effect on tree growth patterns that may have been adversely affected by pollution decades previous. Furthermore, tree growth represents only one facet of 'vigorous' ecosystems. Long-term pollution damage includes disruptions of nutrient cycling as well as alteration of habitats for flora and fauna. Although in Polish National Parks, SO_2, NO_2 and O_3 has not had a long-term demonstrated influence on *Picea* growth, there have been noted effects on soil chemistry and nutrient cycling (Grodzinska et al., 1990).

Strong evidence that relates regional air pollution or point source pollutants to tree growth is rare and the causal relationships are difficult to demonstrate despite sophisticated approaches (Cook and Innes, 1989). Our findings support the possibility of air pollution related growth reductions and declines in the Carpathian Mountains, but the limited temporal air pollution data do not allow for extrapolation to long-term trends. The correlations presented here do not provide sufficient evidence to formulate an exact cause-and-effect relationship; however they do support consistent trends in tree growth related to air quality. Future studies and continued monitoring of air pollutants will provide opportunities to more closely observe the patterns described in this study.

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