Historical trends in tree-ring growth and chemistry across an air-quality gradient in Wisconsin

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Basal-area increment and chemical composition of xylem wood were measured in three old-growth (ca. 75–100 years) white pine (Pinus strobus L.) and three sugar maple (Acer saccharum Marsh.) stands across a pH and SO$_4$ gradient in precipitation in Wisconsin. In 1986 the volume-weighted mean pH and SO$_4$ content of precipitation ranged from 4.5 to 5.0 and from 21 to 11 kg · ha$^{-1}$, respectively, from southeastern to northwestern Wisconsin. With one exception (a white pine site at Point Beach in eastern Wisconsin), basal-area increment increased from the 1890s until the 1950s (sugar maple) or 1970s (white pine), then leveled off. Growth efficiency, estimated as the ratio of basal area to exposed crown area or crown volume for the 1980–1985 period, was similar for sugar maple across the gradient; however, growth efficiency of white pine was lower at Point Beach than at the two northern Wisconsin sites. Leach concentrations in xylem wood of both species have increased with time, except at Crotte Creek in northwestern Wisconsin, and Pb concentrations in xylem wood of both species were significantly greater in southeastern than in northwestern Wisconsin. Sulfur concentrations in xylem wood of white pine have increased since the 1960s at Point Beach and at one site in north central Wisconsin; S concentrations are significantly greater for both species in southeastern than in northwestern Wisconsin. Concentrations of Ca, Mg, and K in xylem wood of sugar maple have decreased over the past century. Whereas xylem wood concentrations of Mn and Zn generally show no age-related trends, Fe and P concentrations have increased markedly at all sites, particularly during the past decade. Although additional research is needed to determine the potential of dendrochemistry in evaluating the consequences of environmental pollution, the age- and site-related trends in chemical composition of xylem wood of white pine and sugar maple appear to be related to vehicular emissions (Pb), air pollution (S), migration along ray paths during conversion of sapwood into heartwood (P, Fe, Ca, K, Mg), and possibly re-allocation of nutrients from the labile soil pool to perennial tree tissues during stand development (Ca, K, Mg).


L’accroissement en surface terrière ainsi que la composition chimique du bois de xylème ont été mesurés dans trois peuplements de première venue (env. 75 à 100 ans) de Pin blanc (Pinus strobus L.) et trois peuplements d’Érable à sucré (Acer saccharum Marsh.) localisés le long d’un gradient de pH et SO$_4$ de la précipitation au Wisconsin. En 1986, le pH et le contenu en SO$_4$ moyens pondérés du volume de la précipitation ont varié de 4.5 à 5.0 et de 21 à 11 kg · ha$^{-1}$, respectivement, du sud-est au nord-ouest du Wisconsin. À une exception près (la station de Pin blanc de Point Beach dans l’est du Wisconsin), l’accroissement en surface terrière a augmenté depuis les années 1890 jusqu’aux années 1950 (Érable à sucré) ou 1970 (Pin blanc), puis a culminé. La perte de croissance, estimée comme le ratio de la surface terrière à la surface ou au volume de la cime pour la période allant de 1980 à 1985, était semblable pour l’Érable à sucré le long du gradient; cependant, l’efficacité de la croissance du Pin blanc était moindre à Point Beach qu’aux deux stations situées plus au nord. Les concentrations de plomb dans le bois de xylème des deux essences ont augmenté avec le temps, sauf à Crotte Creek au nord-ouest du Wisconsin, et les concentrations de Pb étaient significativement plus élevées dans le bois de xylème des deux essences au sud-est qu’au nord-ouest du Wisconsin. Les concentrations de soufre dans le bois de xylème du Pin blanc ont augmenté depuis les années 1960 à Point Beach et à une station du centre-nord du Wisconsin; les concentrations de S sont significativement plus élevées pour les deux essences au sud-est qu’au nord du Wisconsin. Les concentrations de Ca, Mg et K dans le bois de xylème de l’Érable à sucré ont diminué durant le dernier siècle. Alors que les concentrations de soufre du bois de xylème en Mn et Zn ne montrent en général aucune tendance liée à l’âge, celles de Fe et P augmentent de façon sensible à toutes les stations, surtout durant la dernière décennie. Bien qu’il soit souhaitable de poursuivre ces recherches pour déterminer le potentiel de la dendrochimie dans l’évaluation des effets de la pollution environnementale, les tendances liées à l’âge et à la station dans la composition chimique du bois de xylème de Pin blanc et d’Érable à sucré semblent liées aux émissions des véhicules (Pb), à la pollution de l’air (S), à la migration le long des rayons durant la transformation de l’auviers en bois de cœur (P, Fe, Ca, K, Mg) et possiblement à la réallocation des éléments nutritifs du sol vers les tissus terminaux de l’arbre durant le développement du peuplement (Ca, K, Mg).

[Intraduit par la revue]

Introduction

Tree-ring analysis has been used in recent years to assess the degree to which acidic deposition and other sources of air pollution have affected tree growth in the eastern United States. Acidic deposition has been implicated in growth reduction of several tree species over the past 25 years (Johnson et al. 1981; Puckett 1982; Johnson and Sicamana 1983; Hornbeck and Smith 1985; McClanahan and Dochinger 1985; McLaughlin 1985). Chronological sequences of elemental concentrations in wood (dendrochemistry) have been used to assess historical changes in anthropogenic emissions. Symeonides (1979) showed that

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a reduction in growth of *Pinus sylvestris* L. downwind from a smelter in northern Sweden was positively correlated with concentrations of Cu and Pb in tree cores. Vogelmann (1982) reported higher Al concentrations in tree rings from 1950 to the present than in those from the early 1900s. Baes and McLaughlin (1984) detected growth suppression and increased concentrations of Fe and other metals during periods of smelting activity in Tennessee. Rolfe (1974) and Kardell and Larsson (1978) reported increased Pb in rings of trees near major traffic routes. More recently, Arp and Manasc (1988) attributed variations in wood concentrations of 11 elements in *Picea rubens* Sarg. to (i) affinity of the wood for each element during wood formation, (ii) removal through flow of xylem sap, (iii) changed ion availabilities in the soil during stand development, and (iv) air pollution.

Precipitation records collected over the 1980-1986 period at 10 stations in Wisconsin show that sulfate loading is greatest and the volume-weighted pH of precipitation is lowest in the more heavily industrialized southeastern portion of the state (Wisconsin Department of Natural Resources 1985; National Atmospheric Deposition Program 1987). In 1986, for example, sulfate loadings diminished along a gradient from southeast to northwest of 21–11 kg·ha⁻¹·year⁻¹; pH increased from 4.5 to 5.0 along the same gradient (National Atmospheric Deposition Program 1987). There are differences in levels of SO₂ and ozone along this gradient, with the highest levels being recorded in the southeastern part of the state (Rezabec et al. 1986).

Emissions of sulfur and nitrogen oxides have increased dramatically since World War II in the eastern United States (Geschwandner et al. 1986). The pH of precipitation has declined in many areas of the country, including Wisconsin, since the 1950s (Cobgill and Likens 1974). Therefore, it is reasonable to hypothesize that if air pollution has affected tree growth and tree-ring chemistry, these effects should be evident across a spatial gradient from southeastern to north-eastern Wisconsin and a temporal gradient spanning the last 75–100 years. Implicit in this hypothesis are the assumptions that trees respond to increases in emissions by taking up increased amounts of chemical elements and that these elements are preserved in xylem wood. As dendrochemistry is an emerging field of research, these assumptions may or may not be correct.

To test the aforementioned hypothesis, old-growth stands of sugar maple (*Acer saccharum* Marsh.) and white pine (*Pinus strobus* L.) situated along the regional air-quality gradient were selected for study. These species were chosen because of their economic importance and because their range extends across much of the air-pollution gradient of the state. The two species differ in their site requirements, with white pine growing on sandy soils with relatively low buffering and sugar maple occupying loamy soils with greater buffering potential. White pine is known to be sensitive to SO₂ and ozone (Benoit et al. 1982; Rezabec et al. 1986). Recent studies in eastern Canada suggest that acidic deposition may be involved in sugar maple decline (Gagnon 1986).

**Study areas**

The three white pine study sites, Point Beach State Forest, American Legion State Forest, and Crotic Creek, are distributed across the regional air-quality gradient (Fig. 1). Each site features deep loamy sands with level to gently rolling topography. The stands on these sites are defined as northern dry mesic (Curtis 1959) and are dominated by white pine with smaller amounts of red pine (*Pinus resinosa* Ait.), red maple (*Acer rubrum* L.), and eastern hemlock (*Tsuga canadensis* L.). Beech (*Fagus grandifolia* Ehrh.) shares dominance with white pine at Point Beach. The three stands are well stocked and multi-aged, with at least three widely spaced age-classes represented. Trees in the older age-classes were generally not sound and those in the younger age-classes were not large enough for this study. Therefore, the intermediate (ca. 75–100 years) age-class was selected for the analysis. The canopy dominants sampled at each site originated during 20-year episodes of recruitment, beginning in 1890, 1895, and 1910, respectively.

Two of the three sugar maple study sites, Renak–Polock Woods and Kewaskum Woods, are part of the southern mesic forest (Curtis 1959) and are dominated by sugar maple and beech with scattered northern red oak (*Quercus borealis* Michx.). The Plum Lake site is an example of northern mesic forest in which sugar maple shares dominance with hemlock and yellow birch (*Betula alleghaniensis* L.). The three sugar maple stands are broadly multi-aged. Sample trees were released from suppression between 1824 and 1904, with an age range of at least 50 years at each of the three sites.

The climate at all six study sites is cold-temperate, humid, and continental. January temperatures range from −13°C at Crotic Lake to −6°C at Renak–Polock Woods (National Oceanic and Atmospheric Administration 1986). July temperatures average from 19°C at Plum Lake to 21°C at Renak–Polock Woods. Average annual precipitation varies only slightly from 740 to 810 mm among the six sites.

**Methods**

**Field**

At each of the six study sites, 10 (8 at Renak–Polock Woods) dominant-codominant trees were sampled for analysis of tree-ring growth and chemistry. To insure independence among sample trees, a stratified, randomized sampling procedure was used in which the nearest suitable tree was sampled at intervals of about 20 m along one or two transects. A suitable tree was considered to be any sound dominant-codominant representative of the species being studied.
Table 1. A comparison of growth characteristics of the six study sites

<table>
<thead>
<tr>
<th></th>
<th>Sugar maple</th>
<th>White pine</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Renak-Polak</td>
<td>Kewaskum</td>
</tr>
<tr>
<td>Mean age (yr)</td>
<td>103</td>
<td>103</td>
</tr>
<tr>
<td>Mean DBH (cm)</td>
<td>51.2a</td>
<td>49.8ab</td>
</tr>
<tr>
<td>Mean height (m)</td>
<td>27.1a</td>
<td>25.5b</td>
</tr>
<tr>
<td>Height:DBH (∗10^3)</td>
<td>53.1a</td>
<td>51.7a</td>
</tr>
<tr>
<td>Mean exposed crown area (m²)</td>
<td>74.4a</td>
<td>69.9a</td>
</tr>
<tr>
<td>Mean crown volume (m³)</td>
<td>546a</td>
<td>477a</td>
</tr>
<tr>
<td>Mean annual basal-area increment, 1980–1985 (cm²/tree)</td>
<td>23.6a</td>
<td>21.8a</td>
</tr>
<tr>
<td>Growth efficiency (∗ cm².m⁻²)</td>
<td>0.32a</td>
<td>0.32a</td>
</tr>
<tr>
<td>Growth efficiency (∗ cm⁻³)</td>
<td>0.044a</td>
<td>0.046a</td>
</tr>
</tbody>
</table>

Note: Values followed by the same letter for a species are not significantly different at P = 0.05, based on Fisher’s PLSD.

∗Mean annual basal-area increment (cm²), 1980–1985; per cubic metre of exposed crown area.

Most of the sample trees were large (≥46.0 cm DBH), although a few smaller trees (35.0–45.9 cm) were sampled in cases where no large tree was nearby. Each sample tree was cored twice on a north–south axis at 1.37 m above ground level with a Teflon-coated increment borer. Two sizes of increment borer were used: 4.3 mm diameter for sugar maple and 5.6 mm diameter for white pine. To minimize surface contamination, the increment borer was washed with Aliquat 336-S in 2-heptone (Moore 1972) and rinsed with distilled water before insertion. The cores were stored in acid-washed sealed plastic tubes.

The Point Beach and American Legion white pine sites were revisited in late summer to obtain foliar samples, after preliminary data analysis showed major growth differences between these two sites. Small sun-exposed branches were shot down with a rifle or removed with a pole pruner from the mid-crown position of 10 trees at each site.

The following measurements were taken to estimate maximum crown area and crown volume. Total height, height to the widest portion of the crown, and height to the base of the live crown were measured with a clinometer. The exposed crown radius (portion not overlapping adjacent crowns) was measured in the four cardinal compass directions. Crown volume was estimated using the ellipsoid summation technique of Guldin and Lorimer (1985).

Soil samples were collected from each horizon to a depth of 100 cm from a representative profile at each site. In addition, bulk density samples were collected from each horizon, using a coring device.

Laboratory

The increment cores were prepared by removing a very small slice of wood with a Teflon-coated razor blade. This was done as uniformly as possible to avoid altering the relative mass of the rings. The ring widths were read to the nearest 0.01 mm, using a digitizing moving-stage micrometer. The cores were sectioned into the following growth intervals: 1886–1895, 1906–1925, 1926–1945, 1946–1960, 1961–1975, and 1976–1986. These intervals were selected (i) to represent major episodes of industrial activity in the region, (ii) to span major inflections in periodic basal-area increment, and (iii) to yield a sufficient amount of core (0.5 g) for elemental analysis. The corresponding north–south parts from each tree were pooled, rinsed in 0.1 M HNO₃ for 20 s to remove surface contaminants, dried at 70°C, and weighed to the nearest 0.1 mg.

Current and 1-year needles were separated and dried at 65°C. Foliar and tree-ring samples were milled with Teflon-coated razor blades. A subsample of the milled tissue was combusted in a muffle furnace at 500°C for 16 h and dissolved in concentrated HNO₃ and HClO₄. The solutions were analyzed for P, K, Ca, Mg, S, Zn, B, Mn, Fe, Cu, Na, and Al by the Wisconsin Soil and Plant Analysis Laboratory, using an inductively coupled plasma emission spectrometer (ICP) (Schulte et al. 1987). Boron, Cu, Na and Al concentrations in xylem wood were below the detection limit for the ICP, therefore these values are not reported. Concentrations of Pb in tree rings likewise were too low for the ICP to detect reliably. Therefore, Pb concentrations were determined using standard additions on a Perkin-Elmer 603 atomic absorption spectrophotometer equipped with an HGA-2100 graphite furnace. For foliar samples, N was determined on a second subsample by means of a semi-micro Kjeldahl apparatus.

The following analyses were performed on soil samples by the Wisconsin Soil and Plant Analysis Laboratory: pH in 1:1 soil:water extracts; organic C by the Walkley–Black titration procedure; total organic N by a semi-micro Kjeldahl procedure; extractable P by the Bray P1 procedure; and exchangeable K, Ca, and Mg following extraction with 1 M NH₄OAc, pH 7.0 (Schulte et al. 1987). The proportions of sand, silt, and clay were measured using the Bouyoucos hydrometer method, following dispersion in Na₂(SO₄)₆.

Results and discussion

Tree growth

Sugar maple

The following growth parameters were greater at Renak-Polak Woods than at the other two sites: mean DBH, mean height, mean exposed crown area, mean crown volume, and mean annual basal-area increment (BAI) for the 1980–1985 period (Table 1). Site index is not a good indicator of site quality for tolerant species, such as sugar maple, where periodic suppression has occurred. The ratio of height to DBH was proposed as a site-class index for tolerant species such as red spruce (Picea rubens Sarg.) (McLintock and Bickford 1957), sugar maple, and hemlock (Lorimer et al. 1988). There were no significant differences in height:DBH for sugar maple among the three sites (Table 1). Similarly, there were no significant differences among the three sites in growth efficiency of sugar maple, estimated as the ratio of mean annual BAI for the 1980–1985 period to exposed crown area or crown volume.

Mean basal-area increment of sugar maple at the three sites increased from the 1890s to the mid-1950s (Fig. 2A). Whereas sugar maple growth increased between 1976 and 1986 at Renak-Polak and Kewaskum Woods, BAI at Plum Lake has declined since 1955. These growth trends can be explained by physiological and site factors. The recent decrease in BAI is not unexpected for sugar maple trees over 110 years old and in DBH classes ≥45 cm (Lorimer and
## Table 2. Properties of soils from the six sampling sites

<table>
<thead>
<tr>
<th>Horizon</th>
<th>Depth (cm)</th>
<th>Bulk density (g·cm⁻³)</th>
<th>pH</th>
<th>Organic C (%)</th>
<th>Total N (%)</th>
<th>C/N</th>
<th>Extractable P (mg·kg⁻¹)</th>
<th>Exchangeable ions (cmol·kg⁻¹)</th>
<th>% by mass</th>
</tr>
</thead>
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<td>A</td>
<td>0–17</td>
<td>0.91</td>
<td>5.4</td>
<td>3.64</td>
<td>0.18</td>
<td>20.2</td>
<td>8.5</td>
<td>0.211 7.38 1.63</td>
<td>64 26 10</td>
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<tr>
<td>Bw</td>
<td>17–39</td>
<td>1.15</td>
<td>5.8</td>
<td>0.59</td>
<td>0.04</td>
<td>14.7</td>
<td>5.5</td>
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<td>14.3</td>
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<td>0.256 9.88 2.75</td>
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</tbody>
</table>

**Fig. 2.** Mean annual basal-area increment of (A) sugar maple and (B) white pine, from 1895 to 1985.

Frellich (1984). The soil at Plum Lake is a sandy Entic Haplorthod containing lower amounts of extractable N, K, and Mg than the fine-loamy Typic Hapludalfs and Typic Argiudolls present at the Renak–Polak and Kewaskum sites (Table 2).

**White pine**

Although there were no significant differences in mean DBH among the sites, the following growth parameters were significantly lower at Point Beach in eastern Wisconsin than at the two northern Wisconsin sites: mean height, height:DBH, mean BAI during the 1980–1985 period, and growth efficiency (Table 1).

Mean BAI of white pine at the two northern Wisconsin sites (Crotte Creek and American Legion State Forest) increased from the 1890s until about 1970, at which time the growth rate levelled off, then increased again in 1975–1985 (Fig. 2B). In contrast, BAI at Point Beach in eastern Wisconsin increased rapidly until 1910, but gradually declined thereafter until 1981. The declines were steepest during the 1910–1936 period and during the 1960s. The greatest differences in growth rates between Point Beach and the other two sites were from 1895 to 1925 and after 1955. The striking differences in growth of white pine between Point Beach and the other two sites cannot be
explained solely by physiological factors. White pine yield tables clearly show that BAI of individual trees increases until they are approximately 100 years of age (Marty 1965).

Differences in site quality may explain some of the differences in white pine growth among the three sites. The soil at Point Beach, a sandy Typic Udipsamment derived from beach ridge deposits, contains less K (0.22 mol · cm⁻²) in the upper 100 cm than either the Entic Haplorthod (0.61 mol · cm⁻²) or the Typic Udifluvent (0.83 mol · cm⁻²) at the two northern Wisconsin sites. In addition, the uniform sandy texture of the soil at Point Beach (Table 2) may result in less moisture in the upper soil horizons and consequently limit the availability of nutrients whose pool sizes generally are similar among the three sites. To test the hypothesis of lower nutrient availability at Point Beach, we collected current and 1-year-old foliage from white pine at this site and at the American Legion site. Concentrations of N, P, K, Ca, and Mn were significantly lower (p < 0.05) in current and 1-year-old foliage at Point Beach than at the American Legion site (Table 3). The rapid early growth of white pine at Point Beach may reflect utilization of the available nutrient pool; the subsequent growth reduction may indicate a gradual decrease in nutrient availability accompanying the depletion of this pool as nutrients are incorporated into perennial tissues.

**Elemental composition of xylem wood**

**Sugar maple**

Lead concentrations in xylem wood of sugar maple have increased at all three sites over the past century (Fig. 3A). Lead concentrations for a given sampling interval were greatest at Renak-Polak Woods in southeastern Wisconsin and least at Plum Lake in northern Wisconsin (Table 4). We ascribe these age- and site-related differences to vehicular emissions of Pb. Other investigators (Szopa et al. 1973; Rolfe 1974; Kardell and Larsson 1978) recorded historical increases in Pb concentrations in tree rings in heavily trafficked areas. The decrease in Pb over the past 10–25 years at Renak-Polak Woods (Fig. 3A) may represent a reduction in the use of leaded fuels, as was suggested to explain a similar reduction in Pb content in bulk precipitation collected during the 1971–1978 period at Hubbard Brook (Smith and Sicama 1981).

Sulfur concentrations in xylem wood of sugar maple have not changed dramatically over the past 100 years (Table 4). However, at the two southeastern Wisconsin sites, the greatest S levels were recorded in the past decade (1976–1986). In addition, S concentrations in xylem wood for a given sampling period are significantly greater at Renak-Polak Woods, where SO₄ loadings in precipitation are greatest, than at the other two sites.

The concentrations of Ca, Mg, and Mn in xylem wood of sugar maple have decreased substantially over the past 100 years (Table 4). Similar findings were reported by Arp and Manasc (1988) in xylem wood of red spruce. Likens and Bormann (1970) reported greater Ca and Mg concentrations in heartwood than in sapwood of sugar maple at Hubbard Brook. It is possible that Ca, Mg, and Mn are retained in heartwood as immobile elements while mobile elements are exported to sapwood. Alternatively, the decrease in Ca, Mg, and Mn over time may represent reduced nutrient availability due to a gradual accumulation in perennial tissues at the expense of labile soil pools (e.g., Arp and Manasc 1988).

Zinc and K concentrations in xylem wood of sugar maple were fairly uniform with time (Table 4). In contrast, P and Fe increased from the 1890s to the present, particularly during the last decade (1975–1986). Sharp increases in P and Fe concentrations were likewise identified in recent xylem wood of red spruce by Arp and Manasc (1988). Woodwell et al. (1975) reported greater concentrations of P and Fe in sapwood than in heartwood of temperate deciduous species at the Brookhaven Forest. According to Hillis (1987), P may be transferred to sapwood during heartwood formation.

**White pine**

There were fewer age-related trends in elemental concen-
### TABLE 4. Elemental concentrations (µg·g⁻¹) in xylem wood of sugar maple at three sites along an air-quality gradient in Wisconsin

<table>
<thead>
<tr>
<th>Site and growth interval</th>
<th>Pb</th>
<th>P</th>
<th>K</th>
<th>Ca</th>
<th>Mg</th>
<th>S</th>
<th>Zn</th>
<th>Mn</th>
<th>Fe</th>
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<tr>
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<td>898</td>
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<td>122</td>
<td>4.2</td>
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<td>817</td>
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Note: Mean values followed by the same letter are not significantly different at $P = 0.05$, based on Fisher’s PLSD.

### TABLE 5. Elemental concentrations (µg·g⁻¹) in xylem wood of white pine at three sites along an air-quality gradient in Wisconsin

<table>
<thead>
<tr>
<th>Site and growth interval</th>
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<th>K</th>
<th>Ca</th>
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Note: Mean values followed by the same letter are not significantly different at $P = 0.05$, based on Fisher’s PLSD.
trations of xylem wood in white pine than in sugar maple (Table 5). Lead concentrations have increased over the past 10–25 years at Point Beach in eastern Wisconsin and at the American Legion site in northern Wisconsin (Fig. 3B). In addition, Pb concentrations for a given sampling interval are significantly greater at Point Beach than at the two sites in northern Wisconsin (Table 5). As in the case of sugar maple at Renak–Polak Woods, these differences were attributed to greater vehicular traffic in eastern than in northern Wisconsin.

Sulfur concentrations in xylem wood of white pine have increased since 1961–1975 at Point Beach and the American Legion site and in the past decade (1976–1986) at Crotte Creek (Table 5). In addition, S concentrations are significantly greater at Point Beach in eastern Wisconsin than at the sites in northern Wisconsin. These trends are in accordance with SO₄ loadings in precipitation, which decline from southeastern to northwestern Wisconsin. However, there were no significant differences in S concentrations in foliage from white pine at Point Beach and the American Legion State Forest (Table 3).

There were no apparent age-related trends in xylem wood concentrations of Ca, Mg, K, Zn, and Mn in white pine at Point Beach and the American Legion State Forest (Table 5). However, concentrations of Ca, Mg, K, and Zn declined somewhat with time at Crotte Creek. As in the case of sugar maple, P and Fe concentrations in xylem wood of white pine increased at all three sites, particularly during the past decade (1976–1986). The increases in P and Fe may be due to migration along ray paths during conversion of sapwood into heartwood (Hillis 1987).

**Estimation of elemental accumulation in xylem wood**

Elemental composition of xylem wood traditionally is expressed as concentration per unit mass of dry matter (e.g., μg · g⁻¹). Concentrations of heavy metals in xylem wood often increase over time in remote areas (e.g., Berish and Ragsdale 1985) as well as in areas subject to air pollution (Baes and McLaughlin 1984). Berish and Ragsdale (1985) argued that elemental “burdens,” estimated as the product of core mass per growth interval and elemental concentration, are important because concentration increases can result from constant elemental deposition into smaller annual rings. Their estimates of elemental burden are equivalent to the “xylem accumulation rates” calculated by Baes and McLaughlin (1984). The study by Berish and Ragsdale (1985) showed that whereas the concentrations of Pb, Al, and Zn increased over the past century, the burdens of these elements remained constant during the 1900s.

Elemental burdens do not reflect the absolute amount of an element accumulated in the tree, because volume increment and vertical differences in elemental composition within the bole are not taken into consideration. Because ring widths become progressively narrower with time, even in stands presumably unaffected by atmospheric pollutants (Zedaker et al. 1987), elemental burdens may actually mask any increases in elemental accumulation in xylem wood with time. Because we were unable to do stem analyses in these natural and scientific areas for estimating volume increment, we calculated the amounts of elements incorporated into basal-area increment by taking the product of (i) mean periodic basal-area increment (cm²/tree), (ii) specific gravity of xylem wood over the growth interval (estimated by dividing ring mass by the product of cross-sectional area of the core and core length), and (iii) elemental concentration. The data are expressed in micrograms of element per vertical millimetre of stem at breast height. These data were compared with trends in elemental concentration and burden for S in white pine at Point Beach.

The concentrations of S in xylem wood of white pine at Point Beach remain relatively constant until 1946–1961, followed by a marked increase (Fig. 4B). These data would appear to suggest that the trees were responding to increases
in SO$_x$ emissions over time by incorporating greater amounts of S in xylem wood in the last 25-40 years. Although xylem wood concentrations of S are significantly greater at Point Beach than at the two northern Wisconsin sites (Table 5), there were no significant differences in foliar concentrations of S at Point Beach and the American Legion site (Table 3). Sulfur burdens in xylem wood decline sharply from 1886-1902 to 1926-1945, remaining relatively constant thereafter (Fig. 4B). The levelling off in S burden after 1926-1945 is due to a concurrent decline in ring width (Fig. 4A) and an increase in concentration. When the data were expressed in terms of S content in basal-area increment, no change over the past century was apparent (Fig. 4B). These data suggest that historical trends in both elemental concentrations and in xylem wood burdens should be interpreted with caution. A major limitation to interpreting historical trends on the basis of elemental composition of basal-area increment is the vertical variation of elements within bolewood (e.g., Messina et al. 1983).

At the present time very few data are available regarding preservation of chemical characteristics of annual growth rings. Ziegler (1968) suggested that centripetal assimilate-shifting and centrifugal ion-shifting processes occur during heartwood formation. According to Legge et al. (1984), coniferous species have greater potential than deciduous species for constructing heavy metal pollution histories from wood because of better preservation of chemical characteristics. Conifers have fewer and shorter ray cells and have tracheids rather than vessels. Although accumulation of heavy metals in tree rings downwind from smelters has been documented (Baes and McLaughlin 1984; Legge et al. 1984; Arp and Manasc 1988), additional work should focus on the behavior of plant nutrients and trace metals within xylem tissue.

**Conclusions**

In Wisconsin, in contrast to eastern Canada, growth of sugar maple appears to have been relatively unaffected by anthropogenic activities over the past century. Likewise, white pine in northern Wisconsin has not shown the growth reduction reported in other areas subjected to industrial pollution. At Point Beach, white pine growth has decreased since ca. 1910. This decrease may be related to early exploitation of available nutrients from the soil. Although high levels of ozone have been recorded in eastern Wisconsin (Rezabec et al. 1986), the timing of the reduction in white pine growth at Point Beach does not conform with recent increases in ozone. The temporal and spatial differences in xylem wood concentrations of elements in both species may be related to vehicular emissions (Pb), SO$_x$ emissions (S), migration along ray paths between sapwood and heartwood (P, Fe, Ca, Mg, K, Mn), and possibly to retention in biomass at the expense of the labile soil pool (Ca, Mg, K, Mn), particularly in sugar maple. We recommend that interpretations from xylem wood accumulation take into consideration not only elemental concentration and core mass per growth interval, but also basal-area and volume increments.

**Acknowledgments**

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