RADIAL PATTERNS OF TREE-RING CHEMICAL ELEMENT CONCENTRATION IN TWO APPALACHIAN HARDWOOD STANDS

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Abstract: Radial patterns in tree-ring chemical element concentration in red oak (*Quercus rubra* L.) and black cherry (*Prunus serotina* Ehrh.) were analyzed to infer past environmental changes at two mature Appalachian forest sites. Multiple cores extracted from five trees of each species at each site were divided into 5-yr segments, measured for length, composited, and analyzed for 19 elements using plasma emission spectroscopy. Radial patterns of tree-ring chemical elements were categorized into three types: elements showing relatively constant radial concentration in an inner wood zone with increased concentrations in an outer wood zone near the cambium, elements showing relatively constant radial concentration distributions with no rise near the cambium and elements showing gradually increasing concentrations from pith to cambium. Piece-wise multiple regression with correction for serial correlation was used to develop models of these patterns. Significant effects on chemical element concentrations by basal area growth rates and the outer 15 years of growth were detected. With the possible exception of Ca and Sr, trends in chemical element concentrations in inner zone wood with tree-ring age did not indicate a response to the effects of timber cutting, chestnut blight or atmospheric deposition.

INTRODUCTION

Tree-ring chemistry has frequently been used to record changes in levels of air pollutants and, more recently, changes in the soil chemical environment. Long and Davis (1989) recently found that Sr concentrations in white oak (*Quercus alba* L.) in Pennsylvania showed a consistent pattern of higher Sr accumulation from fly-ash in xylem for periods when coal-fired emission stacks were lowest and at locations near power plants. Guyette et al. (1989) showed a decrease in heartwood molybdenum concentration in eastern redcedar (*Juniperus virginiana* L.) which was believed to be caused by increased atmospheric sulfur deposition. McClenahen et al. (1989) showed significantly increased chemical element content of tree-rings in yellow-poplar (*Liriodendron tulipifera* L.) in response to applications of N-P-K fertilizer and dolomitic limestone to the soil. Bondietti et al. (1989) used radial trends in the ratios of Al to base cations in red spruce (*Picea rubens* Sarg.) and eastern hemlock (*Tsuga canadensis* (L.) Carr.) wood to infer the impact of atmospheric deposition on

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soil in Tennessee and North Carolina. Sayre (1987) and DeWalle et al. (1990) both showed that the chemical element content of sapwood xylem in several Appalachian tree species varied with soil fertility. All of these papers indicate that tree-ring chemistry has potential to record time trends in environmental pollution.

Despite the successes in the application of tree-ring chemical analysis to assess environmental pollution, interpretation of results can be ambiguous. Lepp (1975) pointed to many uncertainties with regard to pathways of metal uptake, complexation, and lateral transport in trees which limits use of tree-ring chemical chronologies in studies of environmental pollution. For example, McClenahen et al. (1989) found evidence that P, S, K, Ca, and Zn were mobile in yellow-poplar wood. Thus, application of tree-ring chemistry is not an exact science.

This study was initiated to determine if the radial variation of tree-ring chemistry in two mature Northern Appalachian hardwood stands could be used to determine if changes have occurred in the forest chemical environment. Environmental stresses in these stands can be documented and have included early forest cutting, chestnut blight, and relatively high levels of acidic atmospheric deposition. The sites, Fork Mountain and Pea Vine Hill, have differing soil fertility levels (DeWalle et al. 1988) and any trends related to soil changes were emphasized in this paper. Tree species studied were red oak (*Quercus rubra* L.) and black cherry (*Prunus serotina* Ehrh.), both important commercial species.

STUDY AREAS

Trees studied were located on Fork Mountain at Fernow Experimental Forest in northcentral West Virginia (39°, 03'N; 79°, 41', 30" W) and Pea Vine Hill on Laurel Mountain in southeastern Pennsylvania (40°, 10', 45" N; 79°, 09' W). Fernow Experimental Forest is located near Parsons, WV and is managed by the U. S. Forest Service, Northeastern Forest Experiment Station. The Pea Vine Hill site is part of Rolling Rock Farms near Ligonier, PA. Both sites were gently sloping (<10% slope) ridge-top sites.

The Fork Mountain site was dominated by red oak, black cherry, and sugar maple (*Acer saccharum* Marsh.) trees with minor amounts of other hardwoods occurring in the overstory. Soils were derived from sandstone and siltstone from the Catskill and Chemung formations. Soils were Dekalb and Berks series loamy-skeletal, mixed, mesic, Typic Dystrochrepts. Elevation of this site was about 850 m. Mean annual precipitation at the Fernow Experimental Forest is 142 cm and mean annual air temperature is 8.9 degrees C.

At Fork Mountain heavy high-grade cutting occurred in 1910-15. In addition, residual American chestnut (*Castanea dentata* (Marsh.) Borkh.) trees began to die from chestnut blight in 1910 and by 1925 all mature trees were killed. Salvage logging of dead American chestnut occurred in this stand at about this time.

The Pea Vine Hill site consisted of red oak and black cherry trees almost exclusively. Sandstone and shale from the Pottsville and Mauch Chunk Formations, respectively, were found in soil pits at the site. Soils were Hazleton loamy-skeletal, mixed, mesic Typic Dystrochrepts and Leck Kill fine-loamy, mixed, mesic Typic Hapludults. Elevation of this site was about 880 m. Mean annual precipitation on top of Laurel Mountain near Pea Vine Hill is about 135 cm with 278 cm of annual snowfall depth. Mean annual air temperature is 7.2 degrees C. Forest cutting on Pea Vine Hill has been prohibited since the tract was purchased for Rolling Rock Farms on November 18, 1916. Some dead American chestnut was removed in the 1940-50 period and thus it is assumed that chestnut died in this stand beginning around 1910-15.

Soil at Fork Mountain showed lower acidity and higher exchangeable base cations than soil at Pea Vine Hill. Exchangeable calcium at Fork Mountain averaged 0.82 meq/100 g compared to 0.59 meq/100 g at Pea Vine Hill (DeWalle et al. 1988). Soil water at Pea Vine Hill exhibited significantly higher H, Mn, and Al and lower Ca and Mg concentrations than at Fork Mountain. More complete soil and soil water chemistry at these two sites is available elsewhere (DeWalle et al., 1988).

Atmospheric deposition is relatively acidic at both sites. Out of 82 wet deposition monitoring stations in the United States, the Leading Ridge, Pennsylvania station which is located 130 km northeast of the Pea Vine Hill site ranks 1st in deposition of H^+ and 6th in sulfate deposition (Knapp et al. 1988). The Parsons, West Virginia station within 20 km of Fork Mtn. ranks 9th in H^+ and 10th in sulfate deposition.

METHODS

Five red oak and five black cherry trees were sampled at both the Pea Vine Hill and Fork Mountain sites in 1985. Trees selected for coring were dominant and co-dominant overstory trees with relatively straight boles and healthy crowns within about 500 m of ridge-top soil leachate monitoring sites used in other research. No hollow trees were sampled. Some of the red oak trees sampled at both sites had pith ages over 100 years; these trees apparently survived the heavy cutting in 1910-15 at Fork Mtn. Other red oak and all black cherry trees sampled had pith ages of 85 years or less. Diameters at breast height of sampled trees at Fork Mountain averaged 57 cm for red oak and 48 cm for black cherry. Diameters at Pea Vine Hill averaged 44 cm for red oak and 41 cm for black cherry.

Wood cores were extracted from tree boles at breast height using a 4-mm diameter Teflon-coated increment borer. Borers and extractors were cleaned with acetone and deionized water prior to each field visit and rinsed with deionized water before each extraction. Four cores from bark to pith were extracted from each tree. Cores were extracted at approximately 90-degree angles from each other around the circumference of the trees. Samples included any reaction wood present. No cores with diseased tissue were analyzed. Plastic gloves were used whenever cores were handled. Cores were rinsed with deionized water after extraction in the field and transported from the field and frozen in plastic straws.

In the laboratory, cores were divided into five-year growth increments and the segments of the four cores from each 5 yr segment were composited for each tree. Cores were not cross-dated before compositing. Errors could exist due to missing or indistinct rings when comparing the ages of 5 yr segments among trees and sites. Any such errors are more likely in black cherry with diffuse-porous wood and relatively indistinct rings than in red oak with ring-porous wood and more distinct rings. Composite samples were used to reduce sample variability and provide sufficient mass for chemical analysis. Length of each core segment was measured with a calipers and averaged for each composite sample. Composite sample dry weights averaged 0.7 g with a range from 0.11 to 1.91 g. The division between sapwood and heartwood in the cores was not recorded.

Wood cores were analyzed for phosphorus, potassium, calcium, magnesium, iron, manganese, molybdenum, copper, zinc, sodium, aluminum, silica, cobalt, chromium, nickel, lead, cadmium, strontium, and barium at the University of Georgia, Institute of Ecology using a Jarrel-Ash plasma emission spectrometer. Cores were oven dried, weighed, ashed and digested in 20% nitric acid prior to analysis (Jones 1977).

Multiple regression analysis was employed to analyze trends in tree-ring chemistry data for each element, species, and site. Tree-ring chemical concentrations were used as dependent variables and independent variables were ring age in years, basal area increment in mm^2 per five years, and a piecewise regression term which is described later. Basal area increments were computed from the mean lengths of five-year tree core segments. Serial correlation of residuals from these models, as determined using the Durbin-Watson statistic, was eliminated using an iterative procedure involving differencing all variables over time and using a fitted autocorrelation coefficient (Neter et al. 1985). All statistical tests were conducted at a 95% confidence level.

RESULTS AND DISCUSSION

Basal Area Growth of Trees

Radial patterns of basal area growth for the trees showed peak basal area increments occurred at different times among sites and species (Figure 1). Older red oak trees at both sites experienced rapid diameter growth beginning in about 1910-20 with greatest basal area increment in the 1930-40 period. Release from competition by cutting and chestnut blight probably explains this growth pattern. Younger red oaks and black cherry showed peak basal area increment around 1960 or a gradual increase in basal area increment up to the time of sampling. These younger trees reached breast height after 1910-20 and many are still growing vigorously. As indicated in Figure 1 black cherry trees were generally younger than the red oaks and the youngest black cherry were found at Pea Vine Hill.

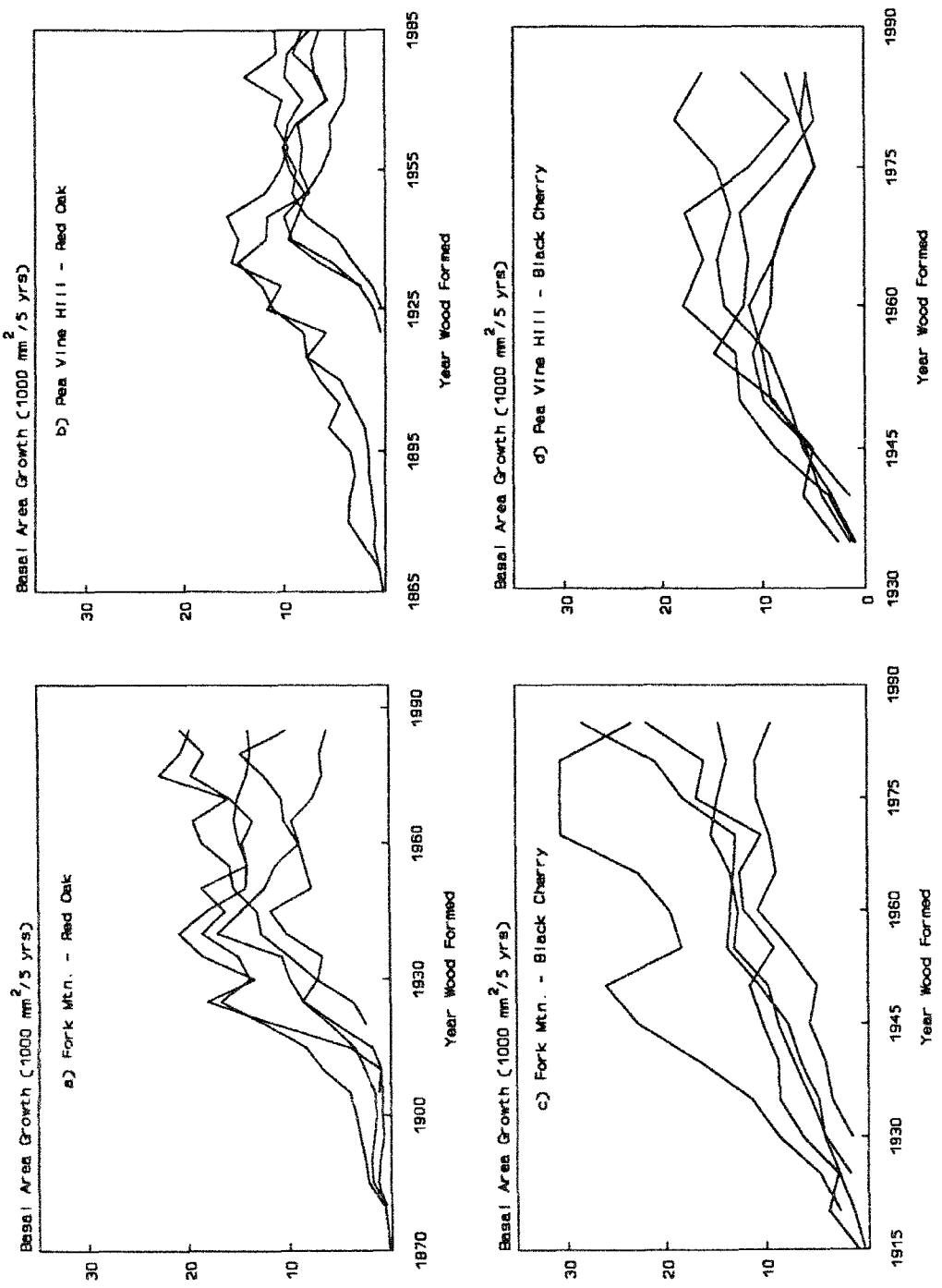

Figure 1. Basal area growth rates of (a) Fork Mtn. - red oak, (b) Pea Vine Hill - red oak, (c) Fork Mtn. - black cherry and (d) Pea Vine Hill - black cherry trees.

Distinct patterns of radial variations in the concentrations of chemical elements were found in tree-rings for both sites and species. Patterns observed varied with the element in question.

A pattern of radial variation in tree-ring element concentration was identified in which concentrations varied between an inner and outer zone. In the inner zone concentrations were relatively constant. In the outer zone concentrations increased sharply as the cambium was approached. This pattern of variation was designated Type 1. Designation of inner and outer zones does not necessarily represent heartwood and sapwood zones in these trees. Elements showing Type 1 variation were Ca, Mg, Sr, Ba, Mn, K, and P. Examples of this type of variation are given in Figure 2 for Ca in red oak and black cherry at both sites. Higher concentrations of Ca occurred near both the pith and the cambium in the older red oaks; the absolute maximum occurred near the cambium in the last increment in almost every case. Minimum concentrations in the older red oaks tended to occur during the 1910-50 period suggesting a growth rate effect on concentration for all alkaline earth metals including Ca. Potassium and phosphorus were also classed as Type 1 elements even though they are not alkaline earth metals.

Several features of the Type 1 variation suggest a growth rate effect on chemical element concentrations in tree-rings. Minimum concentrations occur at about the time of maximum basal area increments at least for the older oak trees. Concentrations also increase and basal area increments generally decrease as the cambium is approached. Both of these trends suggest an inverse relationship between concentration and basal area increment. The importance of this relationship was tested with regression analysis since growth rate effects must be removed to detect any trends in chemical element content over time.

The other extreme in radial pattern in tree-ring elements was designated Type 2 with extreme variability and general lack of any radial trend within trees. Elements which followed this Type 2 pattern include Cd, Pb, Cu, Co, Cr, Ni, and Al. Figure 3 illustrates this pattern for Pb in red oak and black cherry. Although trends in radial Pb are indicated for individual trees, no consistent trends are shown for all trees. This was typical of all the above trace metals. The concentrations of these trace metals did not appear to vary with growth rate or increase in the outer, near-cambium region.

Type 3 radial variation in tree-ring elements was intermediate between the other two types with variable, but gradually increasing, concentrations from pith to bark. No consistent steep increases occurred in the outer zone. Elements falling in this category include Na, Fe, Zn, Mo and Si. Examples of Type 3 radial pattern for Mo is given in Figure 4 for both species and sites. Again trends for some trees may look like Type 1 or 2 elements, but the trend was not consistent among trees or sites. Placement of the elements into one of the three patterns of radial variation was somewhat subjective and regression analysis was used to help quantify relationships.

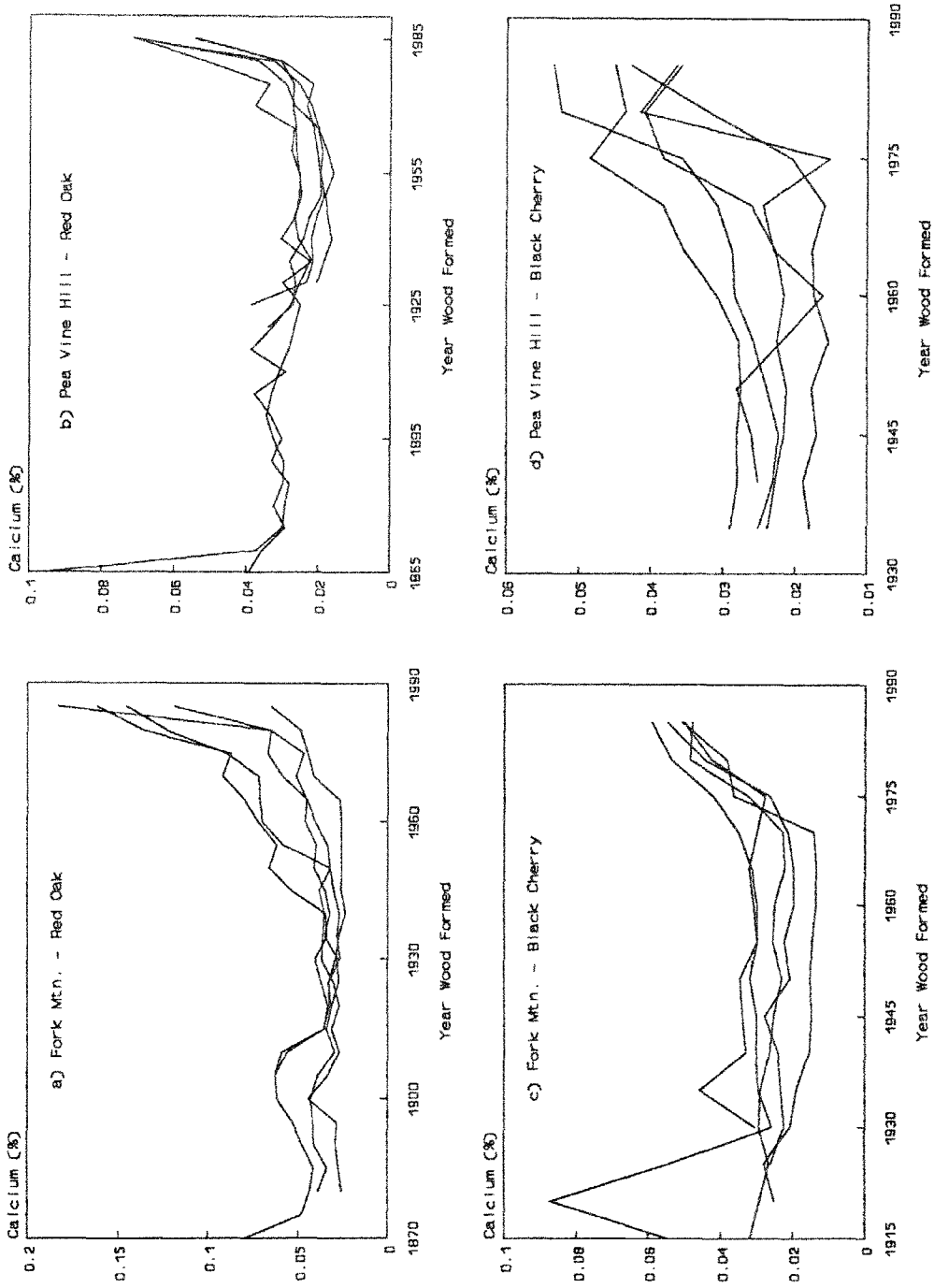

Figure 2. Radial variation of tree ring calcium content for (a) Fork Mtn - red oak, (b) Pea Vine Hill- red oak, (c) Fork Mtn.- black cherry and (d) Pea Vine Hill - black cherry. This pattern of variation was designated Type 1 with a marked increase in the sapwood.

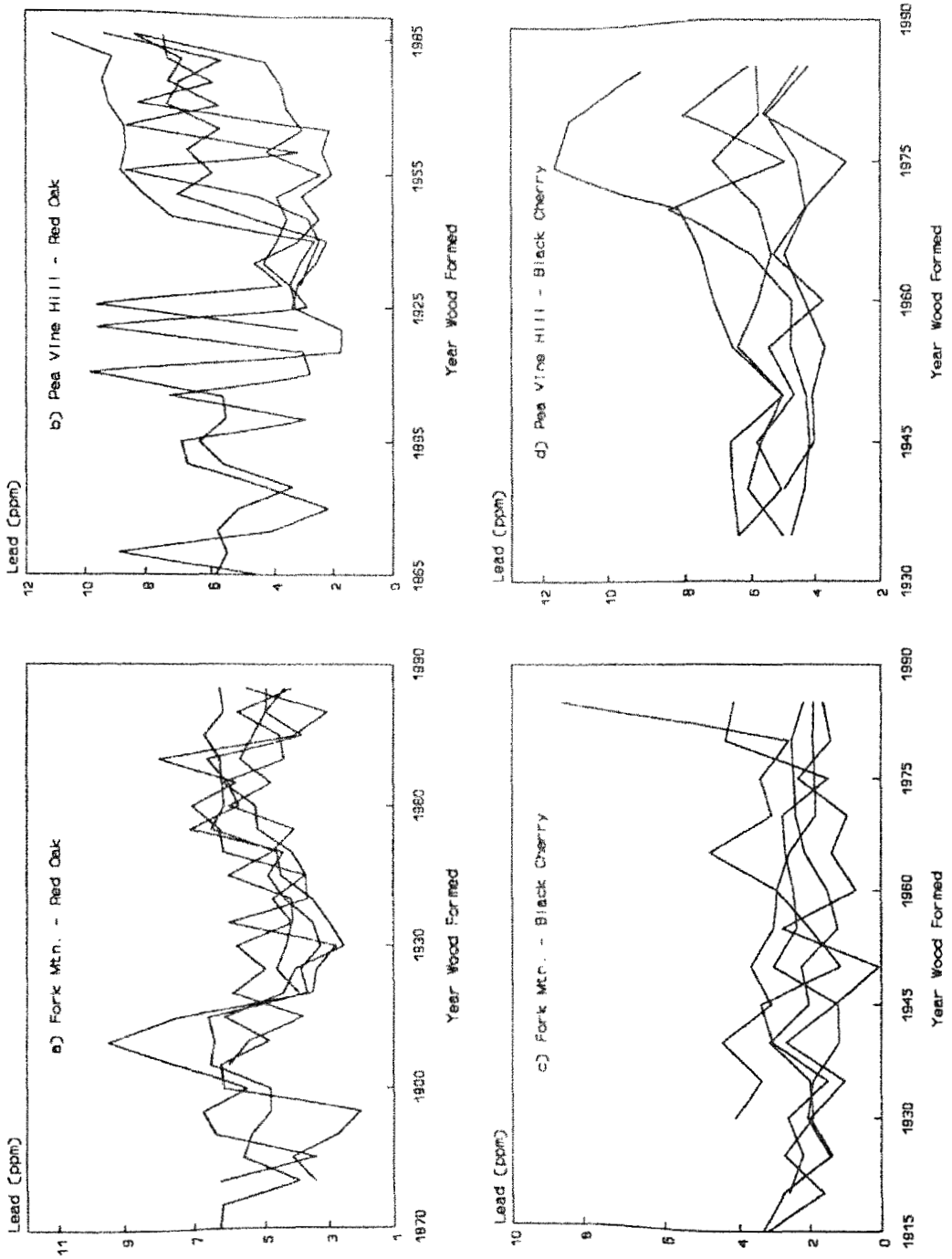

Figure 3. Radial variation of tree ring lead content for (a) Fork Mtn. - red oak, (b) Pea Vine Hill - red oak, (c) Fork Mtn. - black cherry and (d) Pea Vine Hill - black cherry. This pattern with great variation and little radial trend was designated Type 2.

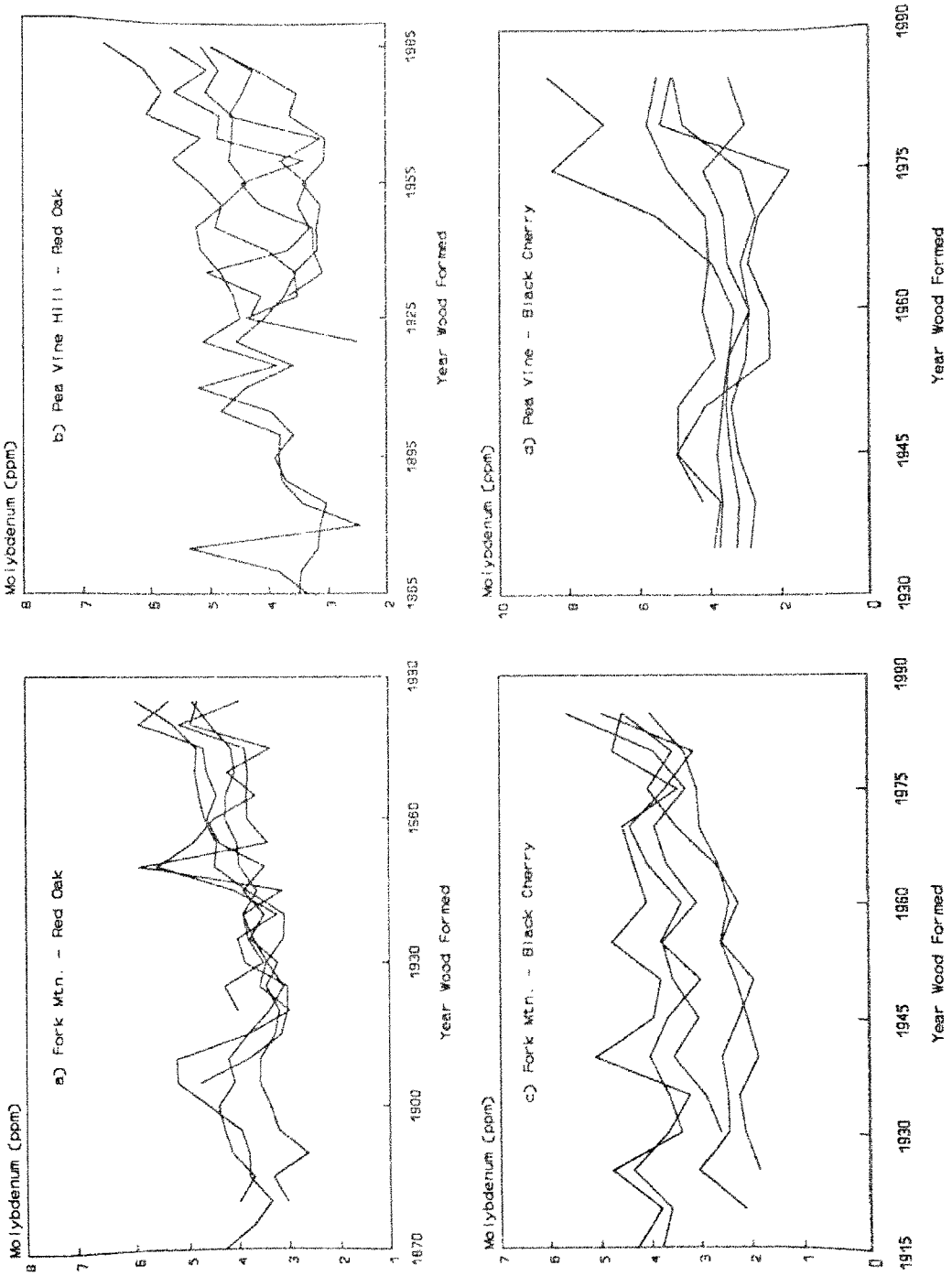

Figure 4. Radial variation of tree ring molybdenum content for (a) Fork Mtn. - red oak, (b) Pea Vine Hill - red oak, (c) Fork Mtn. - black cherry and (d) Pea Vine Hill - black cherry. This intermediate pattern of radial variation was designated Type 3.

The cause of the varying radial patterns among elements can not be determined with certainty at present, but some observations are appropriate. The fact that the same type of radial variation occurred for each element in both species indicates the pattern is closely linked to the nature of the element and not to the considerable difference in wood structure between the ring-porous red oak and diffuse-porous black cherry. Groupings of the elements into the three types of radial variation suggest the cause may be linked to the origin of the elements, e.g. soil vs. atmosphere, or the nature of the element. Type 1 elements were largely bivalent alkaline earth metals which are quite abundant in the soil. Type 2 and 3 elements are primarily trace metals found in the soil and atmosphere. The role radial translocation may have played in shaping the patterns of element variation observed is unknown. Thus, more research is needed to ascertain the true cause of the observed radial variation patterns.

Regression Models of Element Radial Variation

Piece-wise regression models were developed to describe the radial variation in concentration of elements from pith to cambium. Two line segments were used in the piece-wise regression: one for the near-cambium outer region needed for at least Type 1 elements and the second segment for the inner region. The dependence of concentration on growth rate was also included in the models by using basal area increment as an independent variable. The model form used was:

$$Y=b_0+b_1X_1+b_2X_2+b_3(X_2-c)X_3$$

where:

Y = concentration of element in segment, ppm
X_1 = basal area increment for tree, mm^2/5 yr
X_2 = age of growth segment, yr
c = age at inner zone-outer zone boundary, yr
X_3 = indicator variable, X_3=1 if outer, X_3=0 if inner zone.

Through trial and error analysis, the best value for the variable c was determined to be 15 years.

Serial correlation existed in the residuals from application of the above model in most cases. Since serial correlation invalidates hypothesis testing relative to the significance of regression coefficients, a method was needed to remove serial correlation. An adaptation of the transformed variable-iterative approach described by Neter et al. (1985) which involves running the regressions using all variables differenced over time with a serial correlation coefficient was used to remove serial correlation in the residuals. Using this procedure regressions are run on transformed variables as:

$$Y_t'=Y_t-rY_{t-1} \text{ and } X_t'=X_t-rX_{t-1}$$

where r is the serial correlation coefficient. The maximum R^2 equation with a non-significant Durbin-Watson coefficient was adopted after iteration on the serial correlation coefficient in increments of 0.2 from r = 1.0 to r = -1.0. Serial correlation coefficients ranging between -0.2 and 1.0 were needed to eliminate serial correlation in the model residuals based upon a two-tailed Durbin-Watson test statistic. Results of these regressions run with differenced data are summarized in Tables 1 and 2.

Trends of chemical element concentration with ring age or time in the inner-zone, as indexed by the b_1 coefficient, were highly variable among elements, species and sites. In Tables 1 and 2, a positive b_1 coefficient indicates that concentration increased with time or decreased with ring age. Significant trends in the inner zone were indicated in 21 equations, 14 positive and 7 negative regression coefficients, out of the 76 total equations. Within a species the only consistent trend was an increase in Mo concentrations with ring age for red oak at both sites. This trend is opposite that found by Guyette et al. (1989) in heartwood of eastern redcedar after 1860. Within sites the only consistent trend was an increase in P content with time in both species at Pea Vine Hill.

Trends for Ca with ring age in the inner zone varied between species. At Pea Vine Hill, Ca decreased in red oak and increased in black cherry with time. Release from competition by cutting and chestnut blight would be expected to increase availability of Ca. Atmospheric deposition would probably initially increase Ca availability due to mobile sulfate anions in the soil solution and later deplete Ca if the site fertility were low. Pea Vine Hill soil was less fertile than that at Fork Mtn. Increases of Ca with time in black cherry at Pea Vine Hill would more likely be a response to increased atmospheric deposition, since effects of release would be minor by 1935 when these young black cherry reached breast height. Older red oak at Pea Vine Hill showed a general decline of Ca with time in this inner wood zone. Alone, this trend might be taken to indicate general depletion of Ca from the soil by atmospheric deposition. The opposite trend in black cherry for at least the latter part of this time interval confuses the issue and may indicate species differences in accumulation of Ca in bole wood.

Differences in time trends for Sr were also found between species in inner zone wood at Fork Mtn. Strontium increased in red oak and decreased in black cherry at Fork Mtn. These trends are opposite those found for Ca in each species at Pea Vine Hill and may be related to the competition between Sr and Ca in biological systems. However, no significant trends were indicated for either Sr at Pea Vine Hill or Ca at Fork Mtn. to substantiate this relationship.

Other significant changes of element concentration with ring age were not duplicated at either site or for either species. Type 3 elements which were identified with general increases in concentration with ring age based upon visual inspection, did exhibit the majority of positive inner-zone ring age trends.

Variation of element concentration with basal area increment was almost always negative (negative b_2). In 17 out of 20 equations in which the b_2 coefficient was significant, the sign of the coefficient was negative. Negative correlations between element concentration and basal area increment were found for all types of elements. Thus, element concentration

Table 1.--Red oak regression analysis summary for Fork Mtn. and Pea Vine Hill based upon differenced data and use of serial correlation coeffcients.

	Fork Mtn.				Pea Vine Hill			
Element	R^2	b_1	b_2	b_3	R^2	b_1	b_2	b_3
Type 1								
Ca	42	-	N[2]	P	35	N	-	P
Mg	61	-	-	P	85	-	P	P
Sr	36	P	N	P	24	-	N	-
Ba	ns[1]	-	-	-	32	N	N	-
Mn	61	-	-	P	34	-	-	P
K	35	-	N	P	13	P	-	P
P	77	-	-	P	70	P	-	P
Type 2								
Cd	ns	-	-	-	ns	-	-	-
Pb	ns	-	-	-	ns	-	-	-
Cu	19	P	N	-	ns	-	-	-
Co	13	-	N	-	ns	-	-	-
Cr	ns	-	-	-	ns	-	-	-
Ni	18	P	N	-	ns	-	-	-
Al	13	-	N	-	ns	-	-	-
Type 3								
Na	21	-	N	P	ns	-	-	-
Fe	20	-	-	P	30	N	-	-
Zn	ns	-	-	-	28	-	N	-
Mo	30	P	N	P	13	P	-	-
Si	ns	-	-	-	ns	-	-	-

[1]ns = non-significant model, see text.
[2]N = negative trend with time.
P = positive trend with time.
- = no significant trend.

Table 2.--Black cherry regression analysis summary for Fork Mtn. and Pea Vine Hill based upon differenced data and use of serial correlation coeffcients.

	Fork Mtn.				Pea Vine Hill			
Element	R^2	b_1	b_2	b_3	R^2	b_1	b_2	b_3
Type 1								
Ca	19	-	-	P	30	P	N	P
Mg	ns[1]	-	-	-	63	P	N	P
Sr	27	N[2]	-	-	24	-	-	P
Ba	24	N	-	-	24	-	N	P
Mn	30	-	-	P	12	-	-	P
K	47	-	-	P	ns	-	-	-
P	70	N	-	P	70	P	N	P
Type 2								
Cd	ns	-	-	-	20	-	-	N
Pb	ns	-	-	-	ns	-	-	-
Cu	ns	-	-	-	ns	-	-	-
Co	20	N	P	P	ns	-	-	-
Cr	ns	-	-	-	ns	-	-	-
Ni	ns	-	-	-	ns	-	-	-
Al	ns	-	-	-	ns	-	-	-
Type 3								
Na	15	P	N	-	ns	-	-	-
Fe	ns	-	-	-	19	P	-	-
Zn	21	-	P	P	40	P	-	P
Mo	ns	-	-	-	ns	-	-	-
Si	ns	-	-	-	24	P	-	-

[1]ns = non-significant model, see text.
[2]N = negative trend with time.
P = positive trend with time.
- = no significant trend.

generally decreased as basal area growth rate increased. Similar negative correlation between ring widths and element concentration were found by Ragsdale and Berish (1988) for hickory (*Carya* spp.).

Analyses were also conducted using chemical element loads or xylem accumulation rates, computed as the product of concentration and weight of the five year core increments (Baes and McLaughlin 1984), as the dependent variable. This analysis approach did not eliminate growth rate effects. Since use of loads did not simplify the regression models, the analysis based upon concentrations and basal area increment was employed. It is clear that whatever form of dependent variable is used, growth rate effects must be considered when analyzing tree-ring chemical element time series.

Increases in the concentration of elements with time in the outer zone (positive b3) were commonly significant for both Type 1 and 3 elements, as expected. The regression coefficient b3 was significant in 28 out of 76 total equations; with b3 exhibiting a positive sign in 27 equations. Thus, element concentration increases in the outer zone were significant even after the effects of reduced basal area growth rates in this zone were removed. It is hypothesized that sap flow and cation exchange within the near-cambium xylem elements (Bondietti et al. 1989) is primarily influencing concentrations in this outer zone.

CONCLUSIONS

Distinctly different radial patterns of chemical element concentrations occur from pith to bark among elements regardless of species tested or site sampled. These patterns appear related to the nature of the elements and perhaps pathways for elements to reach the bole wood. Piece-wise multiple regression models with corrections for serial correlation were developed to describe radial chemical element concentration patterns. High concentrations of elements were generally associated with periods of low basal area growth. Chemical element concentrations also increased linearly as the cambium was approached in an outer zone of wood of 15 years width. With the possible exception of Ca and Sr, trends in chemical element concentrations with tree-ring age in the inner wood zone did not appear to be related to impacts of timber cutting, chestnut blight and atmospheric deposition.

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