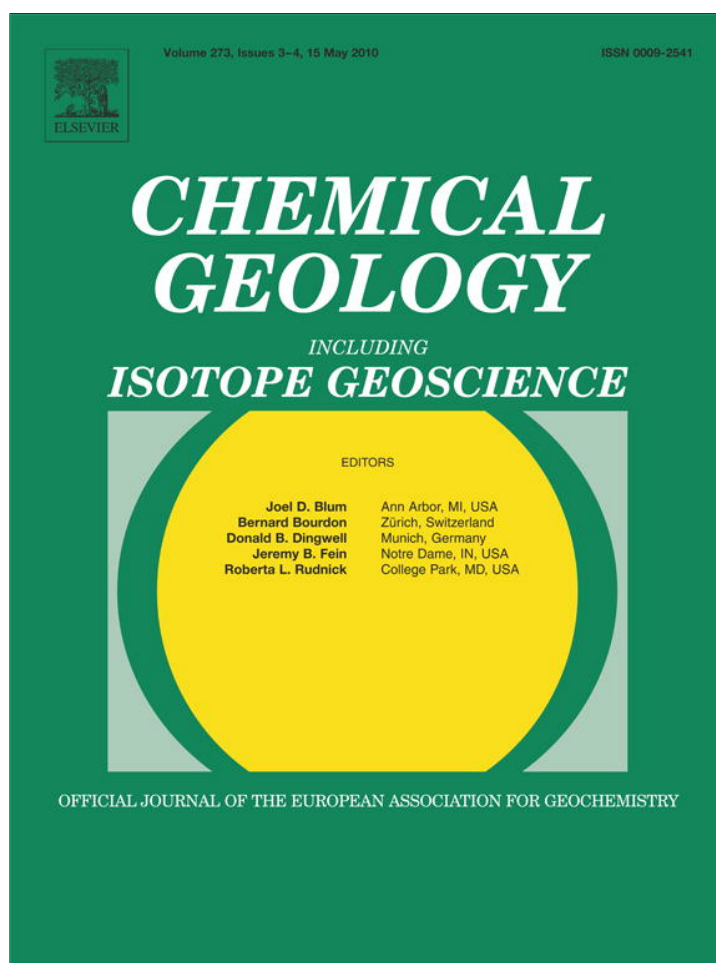


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Tree-ring width and $\delta^{13}\text{C}$ records of industrial stress and recovery in Pennsylvania and New Jersey forests: Implications for CO_2 uptake by temperate forests

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ABSTRACT

We present dendrochronological records of tree-ring width and $\delta^{13}\text{C}$ values for representative living trees (ages of 70 to 230 years) in Pennsylvania and New Jersey to investigate the ecological responses to industrial pollution and recent management and their effects on carbon storage in the temperate forests. Chestnut oak, red oak and cedar trees consistently show elevated $\delta^{13}\text{C}$ levels (1.3 to 4.1‰ higher than unpolluted normal values) during the period of intense pollution from ~1900 to ~1970, indicating significant industrially induced physiological stress on the trees. Since ~1970, oak and cedar trees have shown remarkable increase in growth rates and rapid decrease of 0.6 to 2.5‰ in $\delta^{13}\text{C}$ values, indicating the recovery of trees corresponding to the implementation of the Clean Air Act in the early 1970s. The growth enhancement since 1970 has resulted in an increase in biomass of ~26% in cedar trees and ~66% in oak trees, suggesting that air-cleaning efforts have made a significant contribution to CO_2 uptake by the temperate forests, at least in the northeastern USA.

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1. Introduction

Industrial emission in the past 150 years has caused tremendous increase in atmospheric CO_2 , NO_x , SO_2 and O_3 . In the USA, the levels of these pollutants have in general shown decrease after implementation of federal regulation of industrial emissions (e.g., in 1970 for SO_2 and in 1980 for NO_x), such as by the US Environmental Protection Agency (USEPA). Understanding of the responses of terrestrial ecosystems to elevated air pollutant levels, and later air-cleaning efforts, is crucial in modeling the terrestrial carbon (C) budget and plotting future management. Thus far, most of previous studies have focused on investigation of the effect of elevated pollutants (e.g., Darall, 1989; Strand, 1993). The effect of air-cleaning efforts on the recovery of terrestrial ecosystems has received far less attention.

A number of studies in the laboratory and field, including on natural forests, have revealed that the response of plants to elevated levels of individual industrial pollutants can be two-fold, involving (1) enhanced growth as a result of CO_2 fertilization (e.g., Ainsworth and Long, 2005; Curtis and Wang, 1998; DeLucia et al., 1999; Ellsworth, 1999; Norby et al., 1999; Nowak et al., 2004) or of elevated atmospheric N deposition in N-limited regions (e.g., LeBauer and Treseder, 2008; Magill et al., 1997; Magnani et al., 2007; Thomas et al.,

2010); and (2) reduced assimilation of CO_2 at high levels of O_3 (e.g., Chappelka and Samuelson, 1998; Darall, 1989; McLaughlin and Downing, 1995; Pye, 1988; Tjoelker et al., 1995) or SO_2 (e.g., Darall, 1989; Sakata and Suzuki, 2000; Savard et al., 2002, 2004; Strand, 1993) due to their phytotoxicity.

Fewer studies have examined the interactive effects of these pollutants. Experimental studies show that O_3 impacts can be ameliorated by elevated CO_2 levels (Volin et al., 1998; Karnosky et al., 2003). Modeling efforts consistently indicate that the enhanced CO_2 uptake in response to CO_2 fertilization and N deposition can be cancelled out by the impact of O_3 , apparently resulting in the growth estimate for the case of combined pollutant effects similar to that of no disturbance (Felzer et al., 2007; Ollinger et al., 2002). However, the effect of SO_2 (one of the major industrial pollutants) has not been integrated in experimental and modeling studies, impeding full understanding of industrial impacts on terrestrial ecosystems. A detailed dendrogeochemical study on the natural forests along an SO_2 gradient from a smelter in Québec (Canada) shows that SO_2 imposed great physiological stress on boreal trees, even on those trees more than 100 km away from the SO_2 point source (Savard et al., 2002, 2004). Because SO_2 is one of the major pollutants released by industrial activities, its impact may play an important role in the aggregate effect of all important pollutants on terrestrial ecological CO_2 uptake. Given that the positive effect by CO_2 and N fertilization and the negative effect by O_3 are roughly equivalent in magnitude, if no other factor is considered, the aggregate industrial effect would strongly depend

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on the SO₂ level. This has seemingly been demonstrated by the relationship between industrial SO₂ emission and the decline of fir forests in Japan (Sakata and Suzuki, 2000).

The aggregate industrial impact and the effect of air-cleaning efforts (if any) on forests can be probed by direct investigation of environment-sensitive proxies from old living trees that live through these periods. Tree-ring width and C isotope compositions are commonly used to detect the environmental stresses. Unlike tree-ring width that may be affected by age-related exponential decrease, obscuring environmental signals, C isotope compositions of tree wood are less affected by this age-related phenomenon and can provide a more sensitive record of environmental stresses, particularly anthropogenic environmental changes over the last 150 years (Freyer and Belacy, 1983). Trees can biologically discriminate C isotopes (preferentially using ¹²C) when they assimilate CO₂ from the atmosphere and yield ¹³C-depletion tree woods. The C isotope fractionation between tree wood and atmospheric CO₂ can be expressed as (Farquhar et al., 1982):

$$\Delta^{13}\text{C} = \delta^{13}\text{C}_{\text{tree}} - \delta^{13}\text{C}_{\text{air}} \approx a + (a-b)c_i/c_a, \quad (1)$$

where $\delta^{13}\text{C}$ is defined as:

$$\delta^{13}\text{C} = \left(\frac{^{13}\text{C}/^{12}\text{C}}{(^{13}\text{C}/^{12}\text{C})_{\text{standard}}} \right)_{\text{sample}} - 1 \quad (2)$$

and a represents the fractionation caused by CO₂ diffusion through leaf stomata, b represents the fractionation caused by the ribulose-biphosphate carboxylase-oxygenase (Rubisco), and c_i/c_a represents the ratio between leaf intercellular and ambient atmospheric CO₂ concentrations. Industrial emissions of toxins (e.g., SO₂ and O₃) can affect enzyme activity (Strand, 1993) and reduce a tree's ability to discriminate between isotopes, resulting in elevated $\delta^{13}\text{C}$ values.

In this study, we investigated tree-ring width and the $\delta^{13}\text{C}$ series of trees, of ages of up to >200 years, from natural forests in Pennsylvania (PA) and New Jersey (NJ), USA. This region, located downwind of the Ohio Valley (the industrial centre of the USA), has been heavily polluted. Industrial emissions (particularly of SO₂) increased significantly from ~1900 to 1970 (hereafter referred as the *pollution period*), due to the operation of smelters, and reduced rapidly after 1970 (hereafter as the *recovery period*) as a result of the regulation

mandated by the Clean Air Act. Investigation of ecological responses to these industry-related environmental changes may contribute to the understanding of ecological CO₂ uptake over the last 150 years and assist in projecting the terrestrial C sink in the future.

2. Study sites and analytical methods

2.1. Study sites and local industry history

Pennsylvania and New Jersey are among the most heavily industrialized areas in the northeastern USA (Fig. 1), where increasing emission of pollutants from industrial plants since ~1850 caused severe environmental problems. Since 1970, with the implementation of environmental regulation, the emission of SO₂ and heavy metals showed sharp decrease. Available data acquired since 1980 show that the ground-level O₃ has only slightly decreased (USEPA, 2004). Although there were more minor and transient industrial activities, two large industrial plants had the greatest environmental impact in this area over the last 100 years: the smelters of New Jersey Zinc Company located in Palmerton, Pennsylvania (1898–1980), and plants of Bethlehem Steel located in Bethlehem, Pennsylvania (1899–1995). Industrial activities for these localities are briefly described below.

2.1.1. Palmerton, Pennsylvania

The Palmerton valley is bounded by Blue Mountain to the south and Stony Ridge to the north, and is cut through by the Lehigh River to the west of the town of Palmerton. This topography strongly affects the local patterns of atmospheric circulation, temperature and precipitation. The atmospheric circulation pattern is highly inconsistent, mostly out of the north to the west and more frequently in the winter while intermittent episodes of low wind speed and stagnant air in the summer. The mean annual temperature is 10.4 °C with mean low of 4.6 °C and mean high of 16.1 °C. The mean annual precipitation is 1068.8 mm, which is mostly precipitated during the summer growing season.

The tree coring site is located in the Lehigh Gap Wildlife Refuge at the west bank of Lehigh River, within a few kilometers leeward of the west plant of the now-defunct zinc smelters of New Jersey Zinc Company, which is one of the few localities where trees are available in that area (also see summary by WEF, 2000). The windward side has been severely degraded and is often referred to as a barren wasteland. The greatest environmental damage to the mountain

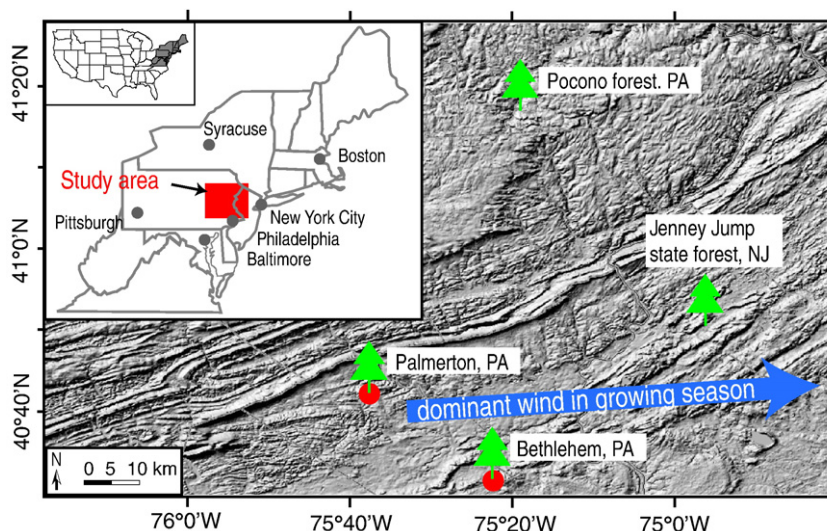


Fig. 1. Digital elevation model showing the study localities in Pennsylvania and New Jersey. Two major local industrial plants are marked by solid circles. Coring sites are marked by tree-like symbols. The dominant wind direction in the growing season is also shown by an arrow. Insets indicate the location of the study area as well as the major industrial centers in the northeastern United States.

forest at Palmerton can be traced back to the early 1800s, when the entire mountain slope was cut and the wood was used as furnace fuel for pig iron furnaces. In the 1840s, railroads were built and major lumbering began. Despite these activities, the mountain slope was still covered with a dense growth of oak and chestnut trees before 1880. In 1898, New Jersey Zinc Company, which was for many years the largest producer of zinc and zinc products in the USA, built the first (west) smelter to process a relatively pure zinc silicate ore. The other (east) smelter was constructed in 1911 and both were roasting zinc sulfide ore as of 1915. A contact acid plant was installed at the same time and SO₂ emission was kept at relatively low levels. Up until 1940, stream engines were used, an activity that produced annual forest fires resulting in drastic denudation of the mountain ecosystem between 1903 and the 1930s. In 1953, an electrostatic precipitator was installed, resulting in a significant decrease in heavy metal emission. However, significant amounts of SO₂ and heavy metals (e.g., Zn, Cd, Pb, and Cu) were released from the smelters, air-borne and deposited in surrounding areas, until the smelters were closed in 1980. As a result of this pollution, severe damage occurred on the landscape over the past century, and approximately 2000 ac of Blue Mountain at the east bank of Lehigh River still resemble barren wasteland unable to support substantial biological activity. Visual damage symptoms from pollutants were commonly observed on surviving trees in the surrounding area even in the late 1960s and early 1970s. For example, Jordan (1975) described that “The Lehigh Gap forest has an unusually open canopy. Trees are widely spaced, and top damage and branches are common. Some foliage is chlorotic; marginal necrosis often develops by mid-summer.”

2.1.2. Bethlehem, Pennsylvania

Located at the bank of Lehigh River in Bethlehem, the Bethlehem Steel Corporation, formed in 1899, was the third largest industrial company in America (see Garn, 1999, for more information). As of 1899, iron furnaces had already been widespread in the Lehigh Valley (23 in operation in 1860). The Saucona Iron Company, which became Bethlehem Steel Corp., was in operation as early as 1857. Production dramatically increased during World Wars I and II (see Fig. 3). The company decreased its production, beginning in the mid 1970s, and finally was closed in 1995. The intense manufacturing, which included a large coking plant, produced unforgettable air pollution in the Lehigh Valley which is still frequently mentioned among Bethlehem area inhabitants.

The coring site is located in a hill slope at the Mountaintop campus of Lehigh University. The mean annual temperature is 10.6 °C with mean annual high of 16.0 °C and mean annual low of 5.2 °C. The local mean annual precipitation is 1085 mm, which is relatively evenly distributed throughout the year.

2.2. Sampling strategy and analytical techniques

Pennsylvania and New Jersey are currently covered by hardwood forests dominated by oaks. Most of them are second-growth trees after the logging for timber and clearance for farmlands in the mid-late 19th century. To examine the local ecological response to industrial pollution and later air-cleaning efforts, we chose the forests on the campus at Lehigh University, Bethlehem (within 1 km of the Bethlehem Steel foundries; see Fig. 1) and in Palmerton (within a few km leeward of the smelters of the New Jersey Zinc Company, an active EPA remediation site). Two other sites farther from local industrial plants (>50 km) were also chosen to study regional patterns. These sites include the Jenny Jump State Forest (JJSF) in New Jersey and the Pocono forest in Pennsylvania. The JJSF is on the slope of a mountain and directly windward of the industrial plants in Palmerton and Bethlehem, whereas the Pocono forest is on a highland and relatively distal from these two pollutant sources (Fig. 1).

From 2003 to 2005, tree-ring samples were cored from trees, at chest height, using an increment borer. More than 20 relatively old trees were sampled at each site. Tree species investigated in this study include red oak (*Quercus rubra*), chestnut oak (*Quercus prinus*), red cedar (*Juniperus virginiana*), and hemlock (*Tsuga canadensis*), which are representative to local vegetations. The oak and cedar trees are mostly on hill slopes with relatively thin soil cover, whereas the hemlock trees are at the foot of small hills (in Palmerton) or on flat mountaintops (in Pocono plateau), in both cases with soil cover thicker than that for the oak and cedar trees.

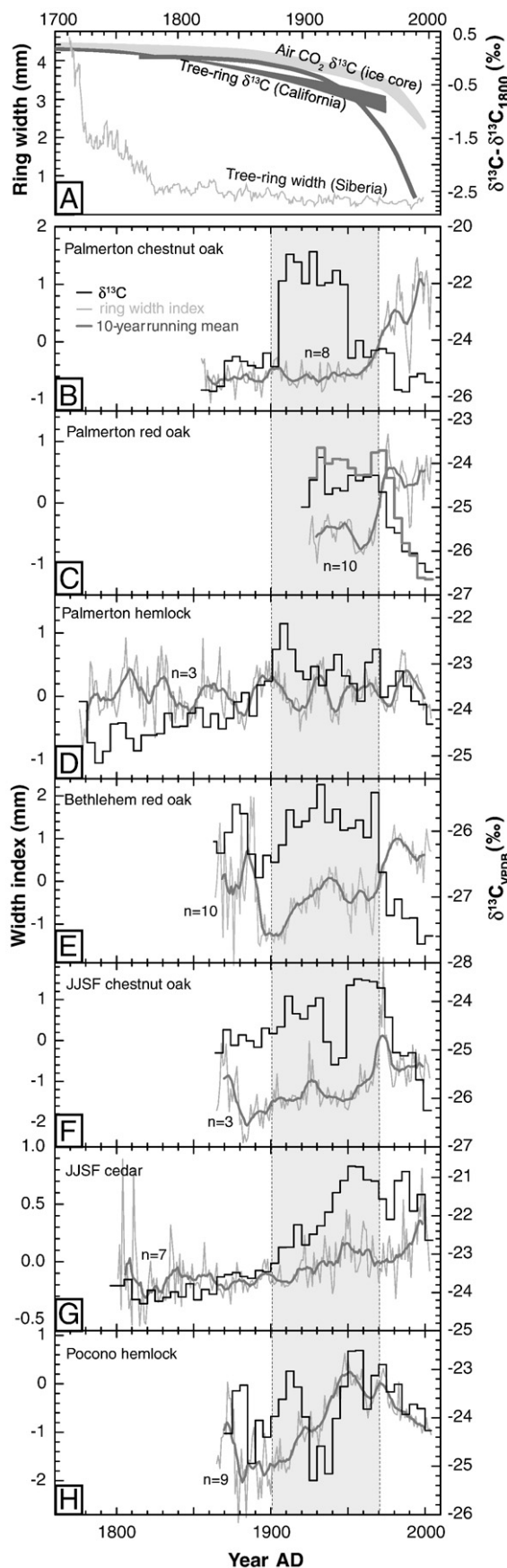
Dendrochronology and tree-rings width were determined for all the cores, using a Velmex tree-ring measurement system in the Lehigh Paleocology Laboratory. However, only trees greater than 70 years old (the age of the oldest tree reaches 230 years) were used to reconstruct the average growth series, as described below. First, individual tree-ring series were detrended using an exponential decay function to remove the ageing effect on tree-ring growth. Since most of the trees show fast growth during two periods (i.e., in their early lives and after 1970), and thus show a saddle pattern in their tree-ring width series, fitting an exponential decay function on all-year widths results in a line that is flat or slightly increasing, which is inappropriate for representation of the ageing effect. Therefore, only ring width data for ages before 1970 were used to fit the exponential decay curve, which is then extrapolated to ages after 1970. The detrended ring width series of individual tree were then standardized relative to the 10-year average value of the detrended width from 1966 to 1975 to deduce the relative growth of each tree. The period of 1966 to 1975 was chosen because the data for tree-ring widths are available for all of the trees and a significant environmental change occurred during this time. Finally, the average growth series was established by averaging all the detrended and standardized ring series for each tree species at each site.

One of the longest cores at each site was used for stable C isotope analysis. $\delta^{13}\text{C}$ values were measured from bulk tree wood, which has been demonstrated to yield as reliable a $\delta^{13}\text{C}$ record for a tree as cellulose and lignin (discussion by Loader et al., 2003). From each core, bulk tree wood representing five-year growth periods was pooled and combusted in quartz tubes with excess CuO_x-Cu reagents at 910 °C for 3 hours to produce CO₂. Carbon isotope compositions were determined in dual-inlet mode using a Finnigan MAT 252 mass spectrometer at Lehigh University. $\delta^{13}\text{C}$ values are reported relative to Vienna Pee Dee Belemite (VPDB), and the reproducibility of C isotope determination was better than 0.1‰ (2 σ) calculated from 12 replicate analyses of graphite standard IAEA-USGS 24 and 4 duplicate analyses of 2 individual tree-ring wood samples.

3. Results

The tree-ring $\delta^{13}\text{C}$ and width indices series are shown in Fig. 2. The $\delta^{13}\text{C}$ values of ring wood before 1900 in most of the trees range from -25 to -23‰. All trees except the hemlock from Pocono (Fig. 2H) show significant elevation in $\delta^{13}\text{C}$ values for the period between 1900 and 1970 (1.3 to 4.1‰ higher than those in pre-industry period). The largest increase in $\delta^{13}\text{C}$ values, for this period, occurs in the chestnut oak from Palmerton (Fig. 2B) whereas the smallest increase in $\delta^{13}\text{C}$ values occurs in the chestnut oak from JJSF. All trees show a rapid, steady decrease in $\delta^{13}\text{C}$ values since 1970 (0.6 to 2.5‰).

Tree-ring width indices do not show such similar patterns for all trees. The most distinct feature is a rapid increase in ring width indices after 1970, which is shown by oak and cedar trees (Fig. 2B, C, E-G) but this is not observed for hemlock trees (Fig. 2D, H). The growth pattern for pre-1970 period is more complex. Some trees (e.g., Bethlehem red oak; Fig. 2E) show reduced growth between 1900 and 1970, relative to growth before 1900, whereas others show no obvious reduction and even an increase in growth (e.g., the Pocono hemlock; Fig. 2H).



4. Discussion

4.1. Ecological response to industry-related environmental changes

In climatic studies, the best sampling strategy for isotope analyses is to pool >5 trees to average the inter-tree isotope variability and dilute the tree-specific, non-climate-related signals (e.g., McCarroll and Pawellek, 1998). In fact, due to the availability limitation of old trees in our study sites, only one or two cores of individual trees were analyzed for $\delta^{13}\text{C}$ values for each tree species. This may introduce more uncertainties in our tree-ring $\delta^{13}\text{C}$ series. However, as strengthened by McCarroll and Loader (2004), the similarity of isotope trends is more important than the absolute values in terms of reflecting environmental and climate changes. It has been demonstrated that individual trees should give similar isotope pattern in response to climate changes even the inter-tree isotope variation is larger than the intra-tree isotope variation (Gagen et al., 2004; McCarroll and Pawellek, 1998) and thus the $\delta^{13}\text{C}$ series of individual tree can be equivalently used to reveal the climate changes (Gagen et al., 2004). Our results also show great similarity of $\delta^{13}\text{C}$ patterns for the two red oak trees from Palmerton (Fig. 2C) and for all of the trees (even of different species; Fig. 2B–G) except one hemlock, the latter from a location in the Pocono region more distal to the industrial sources. Therefore, we believe that the first-order, consistent long-term $\delta^{13}\text{C}$ variations (increase from 1900 or even earlier to 1970 and decrease after 1970) of the core provide a reliable local or regional trend. Given the limited samples, the constraints for the second-order, short-term (<10 year) variation are more uncertain and thus less discussed here.

All trees except the Pocono hemlock show prominent elevated $\delta^{13}\text{C}$ values for the period of 1900 to 1970 (Fig. 2), indicating severe long-term environmental stress in the study region. Some of the trees show obvious decrease in ring width indices for the same period (Fig. 2E, F), whereas others do not (Fig. 2B, G). The absence of the decrease in ring width indices is likely attributable to overcorrection of the early width series during correction for the ageing effect for those trees. The pattern of elevated $\delta^{13}\text{C}$ values for the period of 1900 to 1970, in the PA–NJ forests, is distinct from patterns for more remote sites (e.g., California and Siberia; Fig. 2A), where tree-ring $\delta^{13}\text{C}$ values show steady decrease over the last 200 years (Suess effect; Feng and Epstein, 1995) corresponding to the decrease in $\delta^{13}\text{C}$ values of atmospheric CO_2 due to the burning of ^{13}C -depleted fossil fuel (Francey et al., 1999; McCarroll and Loader, 2004).

Natural factors that could cause increase in tree-ring $\delta^{13}\text{C}$ values (e.g., the juvenile effect and droughts) cannot explain the observed 70-year $\delta^{13}\text{C}$ increase of 1.3 to 4.1‰. The juvenile increase of tree wood $\delta^{13}\text{C}$ values due to different microenvironments and physiological factors at the seedling stage (see review by McCarroll and Loader, 2004) generally lasts for a short period (<40 years) with only slight increase in $\delta^{13}\text{C}$ values of about 1‰ (Duquesnay et al., 1998). The PA and NJ forests, independent of tree ages, show consistent and large increase in tree-ring $\delta^{13}\text{C}$ values for the period of 1900 to 1970 and, in these forests, most of the trees were more than 50 years old when $\delta^{13}\text{C}$ values began to increase in 1900. Drought events can also drive positive isotopic shift due to the reduced ability to discriminate against ^{13}C while the stomata of tree leaves partially close to prevent the loss of water (Ferrio et al., 2003). However, historical climatic records for the northeastern USA show no dry periods of 70-year

Fig. 2. Tree-ring width indices and $\delta^{13}\text{C}$ series of oaks, hemlocks and cedars from Pennsylvania and New Jersey, USA (B–H). “n” represents the number of available old trees used to reconstruct the ring width index. For comparison, also shown in (A) are: (1) ice core record of $\delta^{13}\text{C}$ of atmospheric CO_2 (light gray band; data from Francey et al., 1999 and standardized to the value of 1800), (2) tree-ring $\delta^{13}\text{C}$ trends controlled by the Suess effect in California (two dark gray bands; data relative to the value of 1800; Feng and Epstein, 1995), and (3) example of exponential decrease in tree-ring width due to the ageing process from central Siberia (gray line; Rigling et al., 2001).

duration. The reconstructed Palmer Drought Severity Index (PDSI) close to our study area over the last 300 years (Cook et al., 1999) show more frequent variations but no significant difference in the period of 1900–1970 comparing with earlier periods (Fig. 4B). Therefore, the possibility that these tree-ring records reflect drought conditions is seemingly excluded.

Anthropogenic deforestation may also increase the $\delta^{13}\text{C}$ values of tree wood by reducing air moisture (e.g., Henderson-Sellers and Gornitz, 1984). However, for this region, deforestation is known to have been initiated in the mid 18th century, after European settlement, and forest recovery began as early as the mid 19th century with the abandonment of cropland in the northeastern USA (Hall et al., 2002). Therefore, deforestation could possibly be responsible for the slight $\delta^{13}\text{C}$ increase of the Palmerton hemlock before 1850 but is unlikely to be the primary mechanism responsible for the significant $\delta^{13}\text{C}$ increases for the period of 1900 to 1970.

The timing of the elevated tree-ring $\delta^{13}\text{C}$ values (1900 to 1970) seemingly agrees well with the timing of the pollution period (Fig. 3), indicating that the tree-ring record reflects industrial pollution. It has been demonstrated, by experiments and field studies, that some industrial pollutants (e.g., SO_2 and O_3) can impose significant physiological stress on trees and thus influence the growth and chemical compositions (e.g., isotopes) of tree wood mostly by changing stomatal conductance or impairing the biogeochemistry of photosynthesis (Darall, 1989; Jordan, 1975; Martin et al., 1988; Savard et al., 2002, 2004; Strand, 1993). Dendro-isotopic records from two industrial localities correlate well with known emissions of toxic elements (for Palmerton; Fig. 3A) or raw steel production (for Bethlehem; Fig. 3B), indicating a causal relationship. Although far away from industrial plants, the red cedar and oak trees from JJSF still show elevated $\delta^{13}\text{C}$ values for the pollution period, which correlates well with the sulfur emission inventory and the ground-level O_3 content in the USA (Fig. 3C). These relationships suggest that industrial emission from 1900 to 1970 has impacted the forests not only at a local scale, directly adjacent to factories (within a few kilometers), but also at a regional scale (>50 km).

Since 1970, all trees have shown rapid and steady decrease in $\delta^{13}\text{C}$ values (Fig. 2). Significant increase in ring width indices was also observed for all trees except the hemlocks. Again, this 30-year enhanced growth and steady decrease in $\delta^{13}\text{C}$ values are not consistent with the higher-frequency climate changes (e.g., enhanced air moisture; see Fig. 4B) and thus cannot be explained by such factors. The $\delta^{13}\text{C}$ decrease of atmospheric CO_2 is $\sim 0.8\%$ (McCarroll and Loader, 2004) since 1970, which can account for most but not all of the recent $\delta^{13}\text{C}$ decrease of 1.1 to 2.9‰ in the PA and NJ trees. More rapid decrease in $\delta^{13}\text{C}$ values of tree wood than atmospheric CO_2 over the last few decades was also reported for a number of other sites (see Gagen et al., 2008; McCarroll et al., 2009 and reference therein) but without satisfactory explanation. We suggest that the rapid decrease in $\delta^{13}\text{C}$ values of trees (at least in PA and NJ) since 1970 is attributable to two factors: (1) the decrease in $\delta^{13}\text{C}$ of atmospheric CO_2 , and (2) the ongoing physiological relaxation due to reduced emission of industrial pollutants after passage of the Clean Air Act. The recovery of the trees from industrial impact can thus be inferred by two factors: (1) The tree wood $\delta^{13}\text{C}$ values after 1970 became atmospheric CO_2 -dominant, which was not seen for the industrial period, indicating that the industrial impact has weakened. (2) When the Suess effect is removed, the carbon isotope fractionation between tree wood and atmospheric CO_2 show gradually increasing magnitude after 1970 (Fig. 5), indicating increasing capability of isotope discrimination and thus the relaxation of trees from industrially induced physiological stress.

As a result of recovery from industrial toxins (mostly SO_2 ; and lately O_3 as well), all of the trees but the hemlocks show much more rapid growth, as indicated by the ring width indices, during the recovery period compared with that during the pollution period

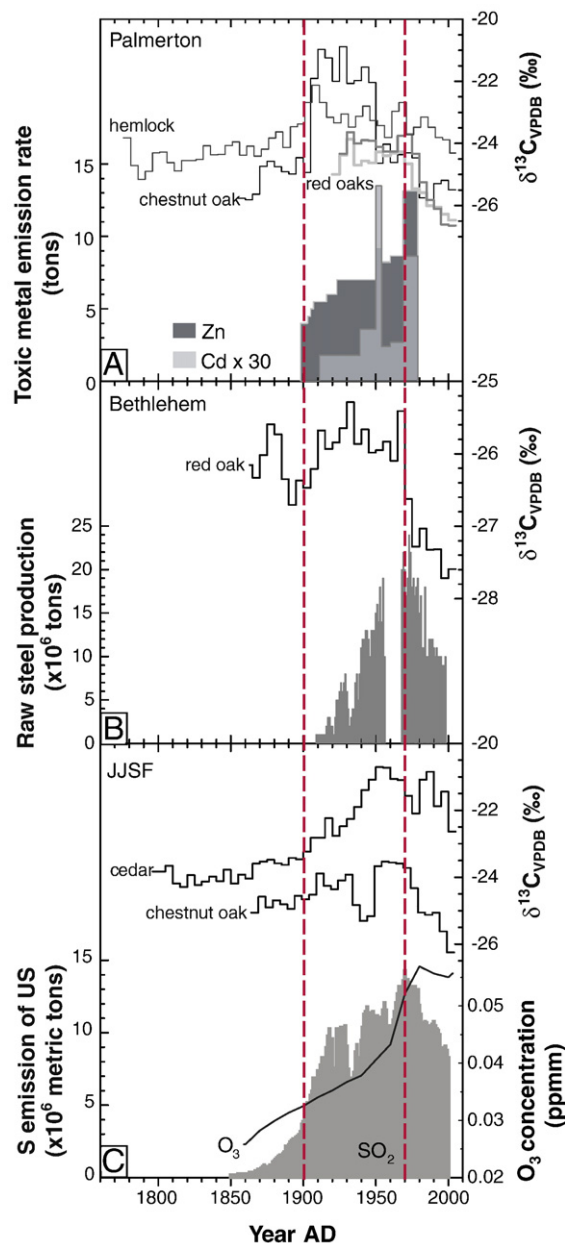


Fig. 3. Comparison of tree-ring $\delta^{13}\text{C}$ time series with local and regional pollution indices, including (A) Zn and Cd emission rates from the New Jersey Zinc Co. smelters at Palmerton, PA. (Buchaur, 1971; USEPA, 1987), (B) Raw steel productivity of the Bethlehem Steel Company at Bethlehem, PA. (Garn, 1999; data around the 1960s are missing), and (C) historic S emission inventories in USA (Smith et al., 2004) and ground-level of O_3 concentrations (based on the model by Horowitz et al., 2003 using the emission data source from Olivier et al., 1996).

(Fig. 3). The weighted average of the width indices of all trees (hemlocks excluded), presumed to represent the regional growth pattern, indicates significant growth enhancement and increased C isotope discrimination over the last 3–4 decades (Fig. 4A). Except the physiological relaxation of trees, nitrogen deposition, another important factor accompanying the industrial pollutants that could positively affect tree growth (e.g., Magnani et al., 2007; Thomas et al., 2010), is also worth of consideration here. The historical nitrogen deposition reconstructed in Massachusetts (Bowen and Valiela, 2001) shows increasing wet deposition of $\text{NO}_3^- - \text{N}$ and total N, but decreasing wet deposition of $\text{NH}_4^+ - \text{N}$ (Fig. 4C) during the industrial period. Despite of different uptake preference of N species for different trees (Templer and Dawson, 2004), nitrogen deposition may

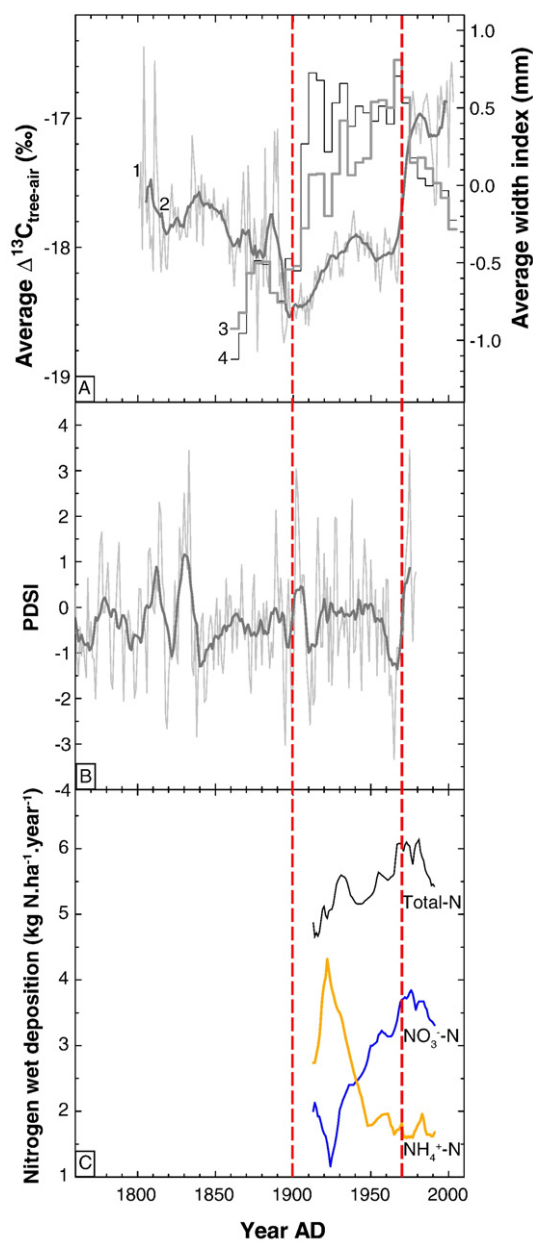


Fig. 4. (A) Averages of carbon isotope fractionation and width index for all trees except hemlock to represent the regional patterns for the past 200 years in Pennsylvania and New Jersey. 1 – Average ring width index; 2 – 10-year running mean of 1; 3 – average C isotope fractionation (Palmerton chestnut oak excluded); and 4 – average C isotope fractionation (Palmerton chestnut oak included). (B) Annual (light gray) and 10-year running mean (dark gray) Palmer Drought Severity Index (PDSI) reconstructed from New Jersey (Cook et al., 1999). (C) Wet nitrogen deposition reconstructed from Massachusetts (Bowen and Valiela, 2001).

contribute to the slightly increasing growth trend between 1900 and 1970 (Fig. 4A), but should play a less important role in the rapid growth enhancement after 1970 because the nitrogen deposition of NO_3^- -N and total N have started to decrease since 1980 when the industrial nitrogen emission was regulated (Fig. 4A, C).

Ring width indices of the hemlock trees show no increase (in Palmerton) or even decrease (in Pocono) since 1970, trends inconsistent with their recent change in $\delta^{13}\text{C}$ values (Fig. 2D, H). The cause of this decoupling of ring width and isotopic records, relative to what is observed for the other trees, is not yet clear. One possibility is that the growth of hemlock may be similar to that of spruce (both are conifer species), the latter which is more sensitive to moisture than to other

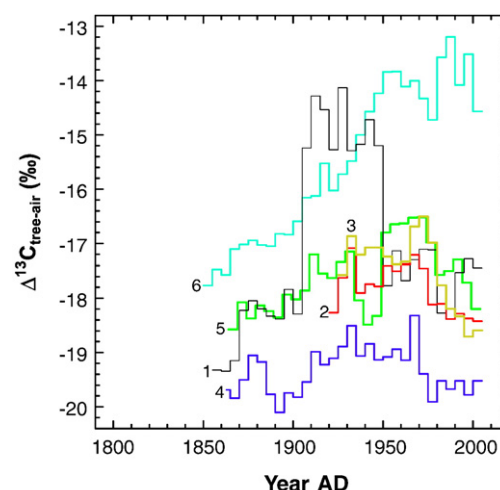


Fig. 5. Carbon isotope fractionation between individual trees and air CO_2 over the last 150 years. 1 – Palmerton chestnut oak; 2 and 3 – Palmerton red oak; 4 – Bethlehem red oak; 5 – JJSF red oak; and 6 – JJSF cedar. Carbon isotope compositions of air CO_2 are from McCarroll and Loader (2004).

environmental stresses (Buhay et al., 2008). Perhaps the industrial signal is weakened in the Pocono hemlocks because they are located on a plateau (elevation of the coring site is ~ 450 m relative to 100–350 m at the other coring sites) and deflected from wind transportation of the major pollutants (Fig. 1) and thus experienced less industrial stress. Another potential cause may be related to the pollution tolerance of hemlock trees. But the lack of experimental data makes it difficult to evaluate this possibility.

4.2. Implications for carbon storage in temperate forests

The elevated tree-ring $\delta^{13}\text{C}$ values in the pollution period, observed in the PA and NJ forests, indicates significant industrially induced physiological stress on these forests, consistent with observations for other industrial sites (Sakata and Suzuki, 2000; Savard et al., 2002). The ring width indices show less prominent growth reduction and inconsistent cross-species and cross-site signatures. Thus, whether a significant growth reduction of the regional forests occurred in the pollution period remains somewhat unclear. However, the enhanced growth in these forests, due to physiological relaxation of industrial stress after 1970, is clearly illustrated by the increase in ring width indices coupled with decrease in tree-ring $\delta^{13}\text{C}$ values. To evaluate the effect of this enhanced growth on the C budget in the temperate forest, we estimated and compared the real biomass of trees and the biomass assuming no growth enhancement after 1970. The aboveground biomass of a single tree can be calculated using the following commonly used equation (see Ter-Mikaelian and Korzukhin, 1997):

$$M = aD^b \quad (3)$$

where M is dry weight of the biomass (kg); D is diameter of the tree at chest height (cm), which can be deduced from tree-ring width series; a and b are constants, but vary with tree species and local living conditions. Ter-Mikaelian and Korzukhin (1997) compiled the regressed a and b parameters for 65 North American tree species. Two sets of these parameters based on measurements of trees in West Virginia and Upper Great Lakes, which are very close to our study area, were employed for the biomass estimation for oak trees. Whereas three sets of parameters from New Brunswick, Upper Great Lakes and Maine were employed for cedar trees because no data from closer sites are available. The site-dependant a and b parameters we employed only resulted in $<10\%$ discrepancy in our calculated

biomass inventory. The diameters used to calculate the biomass are either measured values (for true biomass with growth enhancement) or modeled ones (assuming no growth enhancement after 1970); the latter was deduced by the measured ring width series before 1970 and an assumed annual growth rate for the years after 1970. The assumed annual growth rate after 1970 was determined by three strategies: (1) for the trees more than 100 years old (e.g., cedar, JJSF chestnut oak), the assumed annual growth rates after 1970 were taken to be the average growth rate between 1900 and 1969; (2) for the trees less than 100-year old but show relatively short ageing effect (e.g., Palmerton chestnut and red oaks), the assumed annual growth rates after 1970 were taken to be the average growth rates for all years before 1970; and (3) for the trees less than 100-year old, but show relatively long ageing effects (e.g., Bethlehem red oak), the ageing effect will result in significant overestimation of the true growth if strategy (2) is applied. Therefore, the assumed annual growth rates for after 1970 were obtained by deducing the enhanced growth rate (0.7 mm/year as inferred from Fig. 4A) from the real measurements. The above three strategies should give the maximum diameters for the trees if the growth enhancement didn't occur, thus the calculated increase in biomass induced by growth enhancement after 1970 is a conservative estimate. The results indicate that the C budgets in cedar and oak trees, as of when they were cored, are 5–40% (average 26%) and 15–460% (average 66%), respectively, higher than those assuming no growth enhancement after 1970. Enhanced growth of this magnitude should have significantly increased C storage in this region. Carbon isotope fractionation between tree wood and air CO₂ indicates that the current level is 0.5 to 3‰ higher than the pre-pollution level (Fig. 5), implying that most of the trees have not yet fully recovered from the industrial stress. Thus, continued growth enhancement is expected in the coming few decades. Furthermore, management of industrially damaged/destroyed forests in abandoned, previously industrialized areas (e.g., the ongoing reclamation and restoration projects at Palmerton) can produce additional capacity for net future C storage in terrestrial ecosystems. These factors imply that the C budget, in the forests examined here, will continue to increase at least into the near future. However, it is at present difficult to more broadly estimate the contribution of the recovery to the C sink in the northern forests because of the limited number of test sites. Further studies are necessary to examine whether this recovery-related growth enhancement has been occurring in other US forests, especially in those used in CO₂ fertilization experiments after 1970. The growth enhancement rates deduced from the field experiments in relatively clean air after 1970 may represent the optimal CO₂ fertilization effect, but could be much smaller in the pollution period when the effect of toxins was more overwhelming.

5. Conclusions

Investigations of tree-ring width and wood $\delta^{13}\text{C}$ series of 70- to 230-year old living trees from Pennsylvania and New Jersey (USA) reveal significant industrially induced physiological stress during the period ~1900 to 1970. The decreased growth in response to this stress is difficult to evaluate in this study due to the relatively small sample dataset compared with the observed cross-site and cross-tree species variability, pointing to the need of additional similar work for additional sites throughout the northeastern USA. However, tree-ring width and $\delta^{13}\text{C}$ series both consistently indicate that rapid growth enhancement occurred at least in cedar and oak trees since 1970 as a result of relaxation from the pollution-related environmental stress due to the regulation of the emission of industrial pollutants. Some initial estimates indicate that this growth enhancement has resulted in increase in the carbon storage of ~26% in cedar trees and ~66% in oak trees in this study area.

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