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Records of Anthropogenic Pollution in Silesia Captured in Scots Pine Tree Rings: Analysis by Radiocarbon, Stable Isotopes, and Basal Area Increment Analysis

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Abstract The objective of this study was to analyze tree response to environmental pollution using basal area increment (BAI) tree ring stable isotopes and radiocarbon. Scots pine (*Pinus sylvestris* L.) was assessed from three fresh mixed coniferous forest habitat sites within highly urbanized and populated areas of Silesia subject to high levels of atmospheric pollution and were compared with trees from a site in Silesia where atmospheric emissions were comparatively low. The combined analysis of tree ring width and isotopic data allowed the identification of tree adaptation to environmental pollutants. Changes in BAI revealed a clear long-term decrease in wood increment from 1960 to 1980. We also observed depletion rates of carbon isotopes (^{14}C and $\delta^{13}C$) and increased water use efficiency related to atmospheric CO_2 emissions from fossil fuel combustion.

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1 Introduction

Emissions from human activities connected with home heating, motor vehicles, and industrial activities, including low and high stack emissions, impact the environment. In Poland, most electricity is generated by burning coal. The extensive use of coal as a primary energy source produces significant emissions of pollutants such as sulfur oxides, carbon oxides, particulate matter, and B(a)P (Kaleta, 2014). Between 1960 and 1980, large increases in the emission of industrial pollutants occurred in Silesia, which resulted in the introduction of industrial pollution abatement measures. However, despite actions to reduce pollution, according to the European Environment Agency (EEA, 2021), in the past decade, some cities in Silesia were among the most polluted in the EU. Atmospheric pollution not only can affect tree vigor, but its effects also can form a permanent record captured in the carbon isotope composition of tree rings. Variations in atmospheric carbon isotopic composition can be due to many factors, for example, from an increase in anthropogenic CO_2 emissions, biomass burning, releases from nuclear reactors, and the exchange of carbon within and between different natural carbon reservoirs. Fossil fuel combustion affects atmospheric carbon isotope levels through the

Suess effect, in which the admixture of carbon from fossil fuel combustion dilutes the proportion of ^{14}C and ^{13}C in the atmosphere (Keeling, 1973; Rakowski, 2011; Suess, 1955).

Trees can be used as bioindicators of ecosystem function. For example, α-cellulose has been considered a reliable component of the annual tree ring for examining carbon isotopes because it is directly related to the formation of glucose and cellulose by the plant during photosynthesis (Park & Epstein, 1961; Rakowski, 2011; Saurer & Siegwolf, 2007; Sensuła & Pazdur, 2013; Sensuła & Wilczyński, 2018; Sensuła et al., 2011). In addition, when cellulose forms, further carbon exchange between saccharide molecules and the atmosphere is blocked (Białobok et al., 1993). Pine tree rings have been used to evaluate responses to anthropogenic and natural climatic factors and have been used to document changes in tree growth and carbon isotopic composition of Scots pine (*P.sylvestris* L.) (for example, Nöjd & Reams, 1996; Krąpiec & Szychowska-Krąpiec, 2001; Wilczyński, 2006; Stravinskiene et al., 2013; Sensuła et al., 2011; Sensuła, Opała, et al., 2015; 2018; Sensuła and Pazdur, 2013).

Elevated CO_2 induces changes in stomatal conductivity (g_s) and photosynthetic rates (A_{max}), which can increase plant intrinsic water use efficiency ($iWUE = A_{max}/g_s$). Furthermore, changes in WUE are associated with quantitative changes in isotopic carbon and oxygen ratios in plants, as described by Scheidegger et al. (2000) and Saurer et al. (2004). Emissions of other pollutants may also influence tree growth and carbon isotope composition of tree rings; however, information on such emissions is unavailable due to a lack of reporting by government and industry.

The influence of climate on the isotopic composition of wood has been the subject of previous research and is beyond the scope of this study. Industrial pollution reduces tree vigor and growth and increases sensitivity to meteorological factors, increasing the heterogeneity of their annual incremental responses and weakening the strength of the climatic signal contained in tree rings and in tree basal area increment (BAI). Previous dendrochronological studies indicate that Scots pine is very sensitive to industrial pollution (Nöjd & Reams, 1996; Krąpiec & Szychowska-Krąpiec, 2001; Juknys et al., 2002; Wilczyński, 2006; Malik et al., 2012; Stravinskiene et al., 2013; Sensuła, Wilczynski, et al., 2015; Sensuła et al., 2017;, 2018; Sensuła & Wilczyński, 2018). However, most studies of tree rings evaluated changes in radial growth, which in Scots pine is characterized by a clear decrease with age (the so-called age trend), which can potentially mask changes in tree condition. In contrast to radial growth, basal area increment (BAI) is characterized by a long-term upward trend with age in healthy trees, peaking at an age of several dozen years (Erteld & Hengst, 1966; Wilczyński, 2020).

This study examines the responses of Scots pine to environmental change in four forest areas in Silesia (south part of Poland). Specifically, we assess whether BAI in younger trees is a sensitive indicator of tree vigor, and we examine the use of carbon isotopes in tree rings as a long-term record of historical changes in environment and tree condition.

2 Material and Methods

The research was carried out in four forest areas, three of which have high levels of emissions of airborne pollutants (near the towns of Łaziska (lab code: LAP), Kędzierzyn-Koźle (lab code: ZKC), and Dąbrowa Górnicza (lab code: HKF2)). The fourth forest site was in Olesno (lab code: OLE) and served as a non-polluted control, located about 100 km from large industrial areas (Fig. 1).

2.1 BAI

Fifteen 100-year-old Scots pine trees were selected for study in each of the four stands (Olesno, OLE– the control site; Łaziska, Kędzierzyn-Koźle, and Dąbrowa Górnicza– the sites affected by industrial emissions). Two increment cores were taken 1.3 m above ground level from each sample tree. Tree ring widths on increment cores were measured to the nearest 0.01 mm using the CoRecorder and CDendro programs (www.cybis.se) and were dated and rechecked using the Cofecha computer program (Holmes, 1983). The average radial increment in each year (r) was calculated using the two cores from each tree (r_1 and r_2), where $r = (r_1 + r_2)/2$. Annual basal area increment (BAI) was calculated from the average radius values as follows:

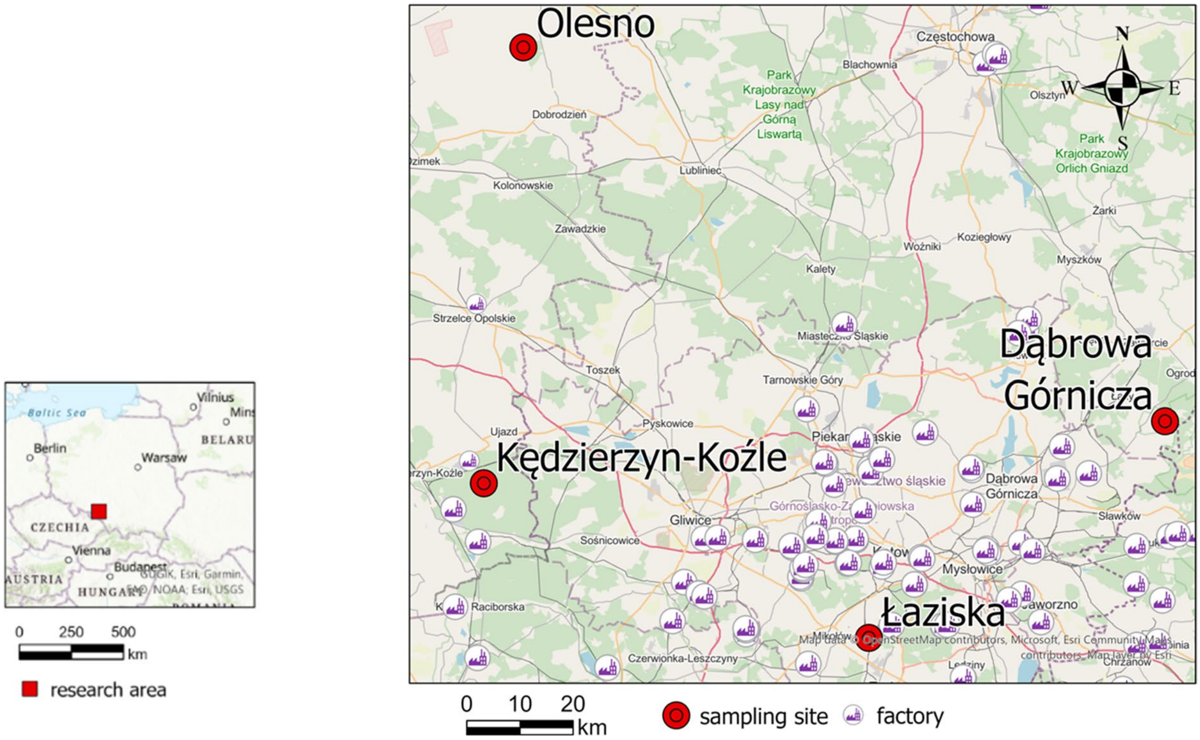


Fig. 1 Sampling sites localized nearby Łaziska, Kedzierzyn-Koźle, Dąbrowa Górnicza, and Olesno

$$\mathrm{BAI}_i = \pi r_i^2 - \pi r_{i-1}^2$$

where BAI_i is basal area increment in year i, r_i is radial increment in year i, and r_{i-1} is radial increment in year $i-1$.

At each site, the annual BAI of the 15 sampled trees was averaged for each year. In this way, site BAI chronologies were constructed. In addition, the BAI series of each tree was subjected to indexing and autoregressive modeling using the ARSTAN program (Cook & Holmes, 1986). The program fits twice a negative exponential curve or trend line to each BAI series. The BAI indices (BAII) for each year are calculated using the formula:

$\mathrm{BAII}_i = R_i/Y_i$, where R_i is the BAI in year i and Y_i is the value of a fit curve in year i.

Then each indexed series was subjected to autoregressive modeling in order to eliminate autocorrelation. To calculate r_{bar}, the residual series created by the ARSTAN program were used. This removed trends, long-term fluctuations, and autocorrelation in the BAI series. For each site, the mean inter-series correlation coefficient (r_{bar}) was estimated to quantify the confidence interval of the indexed BAI chronology for each site and to measure the common variance between individual series (Briffa & Jones, 1990).

The running r_{bar} indices were calculated and plotted for 15-year periods (Fig. 2). The average correlation coefficient of the indexed series determines the degree of homogeneity of the annual growth responses of trees from year to year. It, therefore, gives an insight into the characteristics of inter-tree variability within a stand and the occurrence of environmental disturbances (Briffa et al., 1987).

2.2 Stable Isotopes and Radiocarbon

The standard protocol described by Pazdur et al. (2013) was used for α-cellulose extraction from annual tree rings. The isotopic composition of tree rings is described by δ (in permilles), where $\delta = (R_{\mathrm{sample}}/R_{\mathrm{standard}} - 1)*1000$, and R is the ratio of the heavier to lighter isotope in the sample and in the standard.

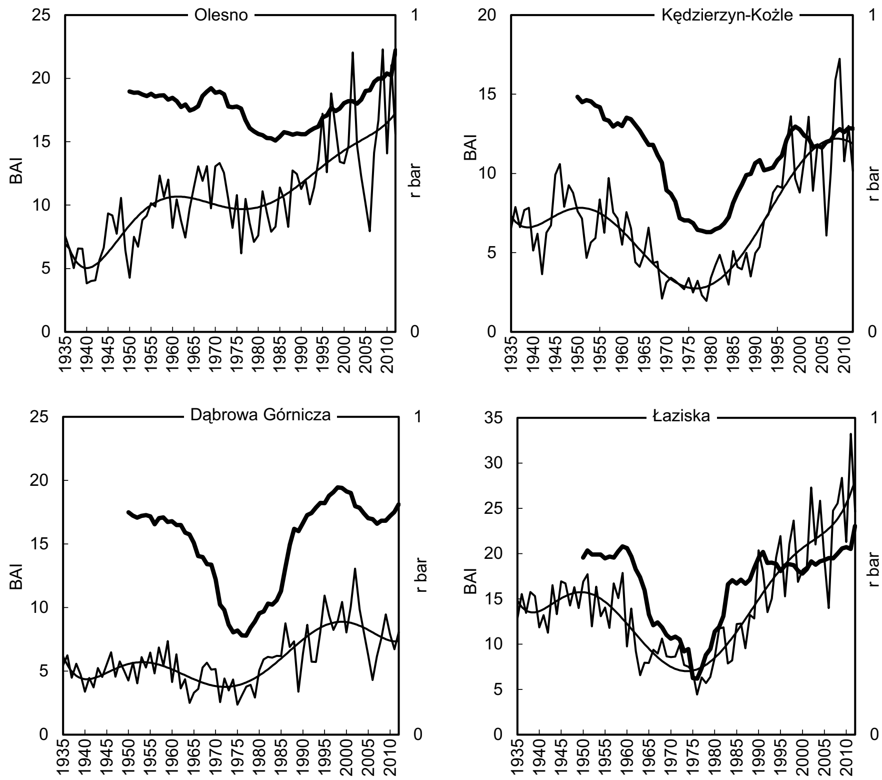


Fig. 2 The BAI chronologies (thin line) and 15-year running values of r_{bar} (thick line). Consecutive values of r_{bar} are plotted at the end of each 15-year period – the first period ranged 1941–1955 and the last period 1998–2012. Łaziska, Kędzierzyn-Koźle, Dąbrowa Górnicza, and Olesno

Intrinsic WUE, directly linked to the ratio of intercellular to atmospheric CO_2, was calculated using carbon isotopes in tree rings according to Saurer et al. (2004):

$$\Delta^{13}C_{cel} = \left(\frac{\delta^{13}C_{air} - \delta^{13}C_{cel}}{1 + \frac{\delta^{13}C_{cel}}{1000}}\right)$$

$$\text{Thus,}\ _{i}WUE = \frac{Amax}{g_s} = c_a \frac{b - \Delta^{13}C_{cel}}{1.6(b-a)}$$

where $\delta^{13}C_{cel}$ and $\delta^{13}C_{air}$ are respectively carbon isotope composition of α-cellulose and the air, c_a is atmospheric CO_2 concentration, $a \sim 4.4‰$ is isotope fractionation during CO_2 diffusion through stomata, $b \sim 27‰$ is isotope fractionation during fixation by Rubisco, A_{max} is photosynthesis rate, and g_s is stomata conductance, respectively.

Determination of carbon and oxygen isotopes in α-cellulose was carried out to assess differences in photosynthetic efficiency and stomatal conductivity from 1975 to 2012. Carbon and oxygen stable isotope compositions were measured at the Mass Spectrometry Laboratory of the Institute of Physics at the Silesian University of Technology, Poland, using continuous-flow isotope ratio mass spectrometry (ISOPRIME, GV Instruments, Manchester, UK). Samples consisted of 60 µg of α-cellulose for $\delta^{13}C$ measurements and 90 µg of α-cellulose for $\delta^{18}O$ determinations (Sensuła et al., 2017, 2018). The detailed analysis of the oxygen isotopes fractionation has already been discussed (Sensuła & Wilczyński, 2018) and is out of the scope of current research. In the current investigation, $\delta^{18}O$ data have been used to analyze the changes in WUE, as described by Scheidegger et al. (2000) and (Saurer and Siegwolf, 2007).

The α-cellulose samples were converted to graphite according to procedures described by Piotrowska (2013). The $\Delta^{14}C$ in graphite was determined by the Rafter Radiocarbon Laboratory (Lower Hutt, New Zealand) or the DirectAMS Laboratory (Bothell, WA, USA). The NIST Oxalic Acid II standard was used for normalization, and black coal was used as a blank material. Values of $\Delta^{14}C$ (‰) were calculated according to van der Plicht and Hogg (2006), in which.

$$\Delta^{14}C = \left(F^{14}Ce^{-\lambda(Ti-1950)} - 1\right)1000$$

where $F^{14}C$ is the normalized radiocarbon concentration, λ is the decay constant for radiocarbon isotopes (equal to 8267 yr^{-1}), and T_i is calendar year.

The analysis of isotopes composition of tree rings was conducted with annual resolution over the years 1975–2012 (stable isotopes), with 5-year resolution between 1975 and 2000, and with annual resolution from 2000 to 2012 (radiocarbon). A detailed description of those results was presented by Sensuła et al., (2018, 2020).

3 Results and Discussion

Trees can be biomonitors of the environment. In this regard, annual tree ring properties can be important in the analysis of local and regional changes in the environment, including the effects of industrial pollution.

3.1 Dendrochronology

Tree rings from the four research plots showed dynamic changes in basal area increment and r_{bar} values over time (Fig. 2). In the years 1935–1955 at Kędzierzyn-Koźle, Dąbrowa Górnicza, and Łaziska, there was a significant increase in BAI. In this period, r_{bar} values remained high. However, in Olesno (control) forest area, high BAI growth and r_{bar} values were maintained only up to the mid-1970s (Fig. 2). The strong decrease of r_{bar} values at Kędzierzyn-Koźle, Dąbrowa Górnicza, and Łaziska occurred in the years 1960–1990, i.e., when emissions of industrial pollutants peaked. At Olesno (OLE), decreasing BAI and much weaker r_{bar} occurred primarily in the 1970s and 1980s (Fig. 2). At the beginning of the 1990s, BAI and r_{bar} increased in all forest areas in the study, with the largest increase in both indicators at Kędzierzyn-Koźle, Dąbrowa Górnicza, and Łaziska (Fig. 2).

Industrial pollution weakens the physiological activity of trees (Emberson, 2003; L'Hirondelle & Addison, 1985; McLaughlin et al., 2002; Percy & Ferretti, 2004), reducing their vitality and growth (Juknys et al., 2002; Stravinskiene et al., 2013; Wilczyński, 2006). Pollution also decreases sensitivity to meteorological factors, increases the heterogeneity of annual growth, and weakens the climatic signal in tree ring chronologies (Wilczyński, 2006, 2020). Changes in BAI revealed a clear long-term decrease in wood increment in study trees. This period marked a peak in emissions of industrial pollutants in Silesia. In those years, the annual variability of BAI and the tree-to-tree uniformity of BAI responses to other factors decreased (Fig. 2), which may be a result of pollutants. Early in the 1990s, following a reduction in industrial pollutant emissions, Scots pine resumed increased wood growth rates. In this period also, the annual variability of BAI and the homogeneity incremental reactions of trees increased (r_{bar}). These results show that Scots pine can recover from damage caused by heavy industrial pollution.

These results confirm the usefulness of BAI and the degree of homogeneity of growth responses (r_{bar}) for evaluating the effects of industrial pollution on trees. Scots pine growing in areas of heavy pollution had, in the period of greatest emissions, clearly reduced BAI and increased heterogeneity in annual growth. Following the reduction of pollutant emissions, pine trees quickly increased both wood growth and the homogeneity of annual growth. In the early 2000s, trees in this study reached their natural maximum BAI, after which the homogeneity of annual incremental responses returned to values seen before the prior century's period of increased pollution.

3.2 Carbon Isotopes

Variations in carbon isotope concentrations in tree rings (Figs. 3 and 4) can be due to the admixture of

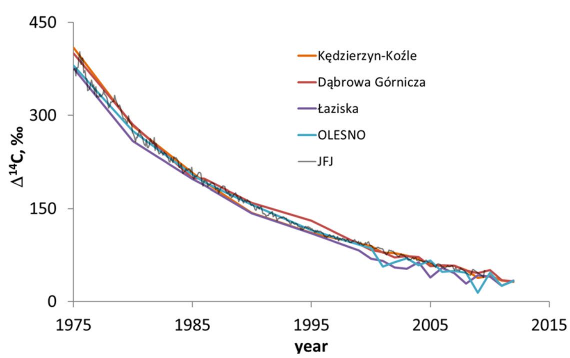
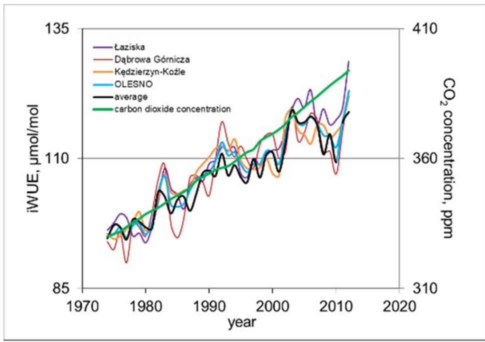


Fig. 3 The comparison of radiocarbon concentrations in Scots pine tree rings (1975–2012) growing in four forests in Silesia and atmospheric radiocarbon concentration measured at the Alpine research station in Jungfraujoch (JFJ) (Hammer & Levin, 2017), an area considered unaffected by local anthropogenic effects



$\delta^{13}C$	↑	↑	↑	↓	↓	↓	*	*
$\delta^{18}O$	↑	*	↓	↑	*	↓	↑	↓
A_{max}	*	↑	↑	↓	↓	*	↓	*
g_s	↓	*	*	*	*	↑	*	↑

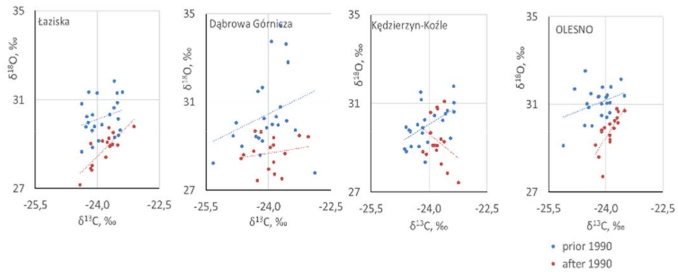


Fig. 4 **a** Variation in iWUE in α-cellulose extracted from annual pine tree rings and increases atmospheric carbon concentration. **b** Theoretical, quantitative link between isotopic carbon and oxygen in plants due to changes in stomatal conductivity (g_s) and rates of CO_2 assimilation (A_{max}), as described by (Saurer and Siegwolf, 2007) and Scheidegger et al. (2000); direction of the arrow presents a direction of the changes; "*" means no changes. **c** Variation in oxygen and carbon isotopes

atmospheric CO_2 with CO_2 from the burning of fossil fuels, as well as the exchange of CO_2 between natural carbon reservoirs. From 1975 to 2012, $\Delta^{14}C$ in tree rings decreased exponentially from year to year, from 409 to 32‰ in Scots pine at Kędzierzyn-Koźle, from 399 to33‰ at Dąbrowa Górnicza, from 375 to 34‰ at Łaziska, and from 380 to 34‰ at Olesno. The admixture of atmospheric carbon with carbon from biomass burning or with carbon from other emission sources enriched in ^{14}C in this region can be significant. In this regard, the most significant local Suess effect was observed in Łaziska.

Between 1975 and 2012, the concentration of atmospheric CO_2 increased from 330 to 394 ppm (Boden et al., 2016), while over the same period, iWUE varied between ca. 90 µmol/mol and 128 µmol/mol, a significant increase in iWUE of about 42%. According to the quantitative model described by Suarer and Scheidegger (Fig. 4b and c), pines react to environmental stress mostly by reducing stomatal conductivity. With climate change, warmer temperatures, especially in concert with reduced rainfall, could produce water stress resulting in reduced stomatal conductivity. In locations where there are large emissions of CO_2 from fossil fuel combustion, local depletion of $\delta^{13}C$ may be observed due to admixture of CO_2 from different sources (Ferrio et al., 2003). In an area where there is not a strong local Suess effect, $\delta^{13}C$ in atmospheric CO_2 is equal to ~ −8.5‰ (White et al., 2015). We observed that $\delta^{13}C$ in trees ranged from −24.4 to −23.1‰ in Łaziska, from −25.3 to −22.9‰ in Dąbrowa Górnicza, from −24.6 to −23.2‰ in Kędzierzyn-Koźle, and from −25.1 to −23.5‰ in Olesno. Mean $\delta^{13}C$ across all sites was −23.9‰. According to Zimnoch et al. (2012), in southern Poland, the $\delta^{13}C$ in coal is ~ −24‰ and in gasoline is ~ −31‰. When these sources mix with CO_2 in the atmosphere, the result is depletion of $\delta^{13}C$ in atmospheric CO_2 and in the biosphere.

4 Conclusions

In conclusion, our study indicates that Scots pine can be useful for biomonitoring environmental conditions. In particular, tree conditions can be important in the analysis of local and regional changes in the environment affected by pollution. The combined analysis of tree ring width and isotopic data allowed the identification of tree adaptation to environmental pollutants. Changes in BAI revealed a clear long-term decrease in wood increment in the years of the culmination of industrial pollution (1960–1990) and an increase in the following years. Based on these results, we believe that Scots pine is able to regenerate rapidly after reducing harmful emissions. We also observed depletion rates of carbon isotopes (^{14}C and $\delta^{13}C$) and an increase in the water use efficiency related to atmospheric CO_2 emissions from fossil fuel combustion. In the region where most energy has been produced by burning coal, the result has been that during photosynthesis, CO_2 fixed by trees will be influenced by depletion of $\delta^{13}C$ caused by the Suess effect.

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Data Availability The datasets (BAI and stable isotopes composition of pine tree rings) analyzed during this study are included in this published article and its supplementary information files. Radiocarbon datasets are available from the authors upon reasonable request.

Declarations

Conflict of Interest The authors declare no competing interests.

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