

Research article

Climatic dominance and delayed industrial modulation of inter-site intrinsic water-use efficiency in Scots pine revealed by tree-ring $\delta^{13}\text{C}$

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Abstract. Tree-ring stable carbon isotopes provide insights into long-term physiological responses of forests to environmental change. This study examined inter-site divergence in intrinsic water-use efficiency ($\Delta iWUE$) and radial growth (ΔBAI) of Scots pine (*Pinus sylvestris* L.) at two sites in northern Lithuania from 1933 to 2022. One site is located near a major historical source of industrial emissions, while the other represents a comparatively less affected environment. Tree-ring $\delta^{13}\text{C}$ data were used to reconstruct $iWUE$, and inter-site differences were analysed in relation to climatic variability and reconstructed emission histories. Both physiological and growth chronologies showed increasing divergence beginning in the early 1990s. Mean $\Delta iWUE$ increased significantly after 1990, while ΔBAI shifted from negative to positive values, indicating a reversal in relative growth performance. Inter-site differences in $iWUE$ were moderately correlated with growth divergence ($r = 0.45$, $p < 0.001$). Generalized additive models identified climatic variability as the dominant driver of $iWUE$ divergence. Differences in growing-season precipitation showed a significant nonlinear relationship with $\Delta iWUE$ and explained the largest proportion of deviance among climatic predictors. After accounting for temporal trends and climatic divergence, models including industrial emissions revealed additional nonlinear effects. One-year lagged sulfur dioxide showed the strongest relationship with $\Delta iWUE$, while lagged carbon monoxide improved model performance; nitrogen oxides had no significant effect. These results indicate that climatic variability, particularly moisture availability, primarily controls inter-site physiological divergence, while industrial emissions exert secondary, lagged influences.

Keywords: Tree-ring stable isotopes; $iWUE$; whole-wood; air pollution; climatic variability; stomatal conductance; generalised additive model (GAM); Eastern Baltic

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INTRODUCTION

Forests across the Eastern Baltic region are exposed to both rapidly changing climatic conditions and long-term anthropogenic pressures. Distinguishing between these drivers remains a central challenge in dendroclimatology, particularly where industrial activity and

late-20th century climate warming have overlapped. Traditional growth proxies such as tree-ring width (TRW) integrate multiple structural and ecological influences, including stand dynamics, tree age, and site conditions, which can obscure the physiological mechanisms underlying observed growth responses (Fritts 1976; Babst *et al.* 2019; Frank *et al.* 2010).

Stable carbon isotope ratios ($\delta^{13}\text{C}$) in tree rings provide a complementary perspective because they reflect leaf-level processes governing carbon assimilation and stomatal conductance (McCarroll, Loader 2004; Gagen *et al.* 2011). Compared to growth-based parameters, $\delta^{13}\text{C}$ is generally less influenced by structural and site-related variability and more directly records environmental controls on photosynthesis (Loader *et al.* 2003). Carbon isotope discrimination is mechanistically linked to the ratio of intercellular to atmospheric CO_2 concentrations (Farquhar *et al.* 1982), allowing reconstruction of intrinsic water-use efficiency (*iWUE*), which integrates climatic influences with changes in atmospheric composition (McCarroll, Loader 2004). Numerous studies have documented long-term changes in *iWUE* across temperate and boreal forests, commonly interpreted in relation to rising atmospheric CO_2 concentrations, climate variability, moisture availability (Francey, Farquhar 1982; Peñuelas *et al.* 2011; Frank *et al.* 2015; Guerrieri *et al.* 2019), and pollution-induced physiological stress (Pišová *et al.* 2008; Sensula, Wilczyński 2022; Trembl *et al.* 2022). As such, tree-ring *iWUE* has become a widely used indicator of long-term plant responses to environmental forcing.

Climatic influences on *iWUE* operate primarily through their effects on stomatal behavior and photosynthetic demand. Temperature influences photosynthetic capacity and evaporative demand, whereas moisture availability constrains stomatal conductance, particularly during the growing season (Farquhar, Sharkey 1982; Körner 2015). In many forest ecosystems, indices integrating precipitation and evaporative demand, such as the Standardized Precipitation-Evapotranspiration Index (*SPEI*), have proven more informative than precipitation alone for identifying climatic controls on *iWUE* (Vicente-Serrano *et al.* 2010; Labuhn *et al.* 2016). In addition, legacy effects from previous growing seasons may influence physiological responses recorded in tree rings, highlighting the importance of considering both current and prior-year climatic conditions (Kagawa *et al.* 2006; Gessler *et al.* 2014). Given the tight coupling between stomatal regulation and soil moisture availability, inter-site differences in growing-season moisture balance are expected to represent a strong control on physiological divergence.

Comparisons of *iWUE* between nearby sites provide an opportunity to isolate differential physiological responses under broadly similar climatic regimes. Inter-site differences may reflect variation in soil properties, moisture availability, microclimate, or sensitivity to environmental stressors (Andreu-Hayles *et al.* 2011; Lévesque *et al.* 2014). In regions where climate change has coincided with substantial anthropogenic influence, such comparisons offer a use-

ful framework for distinguishing dominant climatic regulation from secondary environmental modifiers. Because both climate variability and pollution influence stomatal regulation, careful evaluation of shared temporal structure and site-specific deviations is required to disentangle their relative contributions.

Atmospheric pollution has been proposed as an additional factor influencing tree physiological functioning. Gaseous pollutants such as SO_2 and NO_x may induce stomatal responses and oxidative stress, while industrial emissions can modify the broader environmental context through atmospheric chemistry and deposition processes (Baciak *et al.* 2015). However, empirical evidence for consistent pollution effects on tree-ring *iWUE* remains mixed. Reported associations vary among regions and pollutants and may partly reflect shared long-term trends in atmospheric CO_2 and climate rather than pollutant-specific physiological responses (Saurer *et al.* 1995; Leonardi *et al.* 2012).

Intrinsic water-use efficiency reflects the balance between carbon assimilation and stomatal conductance and is therefore sensitive to factors regulating stomatal behavior. Climatic drivers such as soil moisture availability and atmospheric demand exert strong and often immediate control on stomatal aperture, resulting in pronounced seasonal signals in tree-ring *iWUE* (Farquhar, Sharkey 1982; Jones 1998; McCarroll, Loader 2004; Gagen *et al.* 2011; Lévesque *et al.* 2014; Körner 2015). In contrast, pollution effects are more likely to operate indirectly through cumulative exposure, physiological adjustment, or interactions with climatic conditions rather than through instantaneous responses (Carter *et al.* 2017; Tupu *et al.* 2026). Pollutants associated with sustained combustion activity may therefore influence *iWUE* on delayed or nonlinear timescales, potentially modulating climatic responses rather than acting as primary drivers. This framework suggests that climatic variability is the primary regulator of *iWUE*, whereas pollution effects likely act as secondary influences that may appear with a temporal delay during periods of elevated industrial activity.

Disentangling climatic and pollution influences is complicated by the strong temporal structure shared by climate variables, industrial emissions, and tree physiological responses over the 20th century. Simple linear approaches may obscure nonlinear, delayed, or confounded relationships (Briffa *et al.* 1998; D'Arrigo *et al.* 2008; Matison *et al.* 2021), particularly in regions where industrial emissions and climate trends have co-evolved over recent decades. While numerous studies document long-term increases in *iWUE* driven by rising atmospheric CO_2 and climatic change (Peñuelas *et al.* 2011; Frank *et al.* 2015), fewer investigations explicitly evaluate whether pollution-related physiological effects persist after accounting for

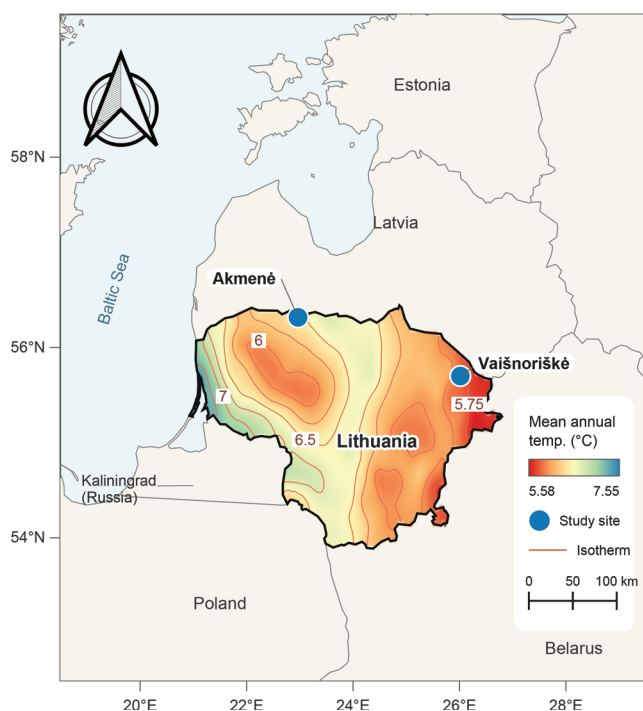


Fig. 1 Study and control site locations at Akmenė (56°19'N, 22°58'E) and Vaišnoriškė (55°42'N, 26°01'E), shown against a background of mean temperature (°C) and isotherms derived from WorldClim v2.1 data. The map was prepared using QGIS

long-term climatic and temporal trends, or whether combustion-related emissions exert distinguishable delayed influences beyond shared background forcing (Erlickytė, Vitas, 2008; Stravinskienė *et al.* 2013; Malik *et al.* 2023).

By focusing on inter-site differences in *iWUE*, this study reduces the influence of shared regional climate signals and highlights differential physiological responses to environmental forcing. Particular attention is given to the possibility that combustion-related pollution effects may be nonlinear or temporally delayed, reflecting cumulative influences rather than immediate physiological responses. The study compares Scots pine growing in Akmenė – a site influenced by historical industrial emissions – and Vaišnoriškė, a comparatively less industrially affected forest site, over the period 1933–2022 (Fig. 1). By analysing tree-ring $\delta^{13}\text{C}$ and derived *iWUE* alongside climatic variables and reconstructed emission histories, this research aims to quantify the relative contributions of climatic variability and industrial activity to inter-site physiological divergence in Scots pine.

We hypothesize that (i) inter-site differences in *iWUE* are primarily regulated by climatic variability, particularly growing-season moisture availability; (ii) industrial emissions exert secondary influences that become detectable only after controlling for shared temporal structure; and (iii) combustion-related pollutants exhibit delayed and potentially nonlinear as-

sociations with *iWUE* divergence, whereas other pollutants may exhibit weaker or inconsistent influence at annual resolution.

MATERIAL AND METHODS

Site description

The study site is located in Šapnagiai, Akmenė District, northern Lithuania (56°19'N, 22°58'E), approximately 3.1 km east of the *Akmenės cementas* plant, the largest cement producer in the Baltic region and the only cement-producing facility in Lithuania. The plant began operating in 1952 and has been a major source of atmospheric pollution in the region. Emissions increased progressively from the 1950s during the following decades, with particularly rapid growth during the 1970s and 1980s (Fig. 2). At the beginning of the 1970s, annual emissions reached approximately 27 thousand tonnes of SO_2 , 9–10 thousand tonnes of cement dust, 8.5 thousand tonnes of NO_x , and about 1 thousand tonnes of ash and other particulate matter (Erlickytė 2007). Total emissions peaked during the period 1989–1991, when overall pollutant loads amounted to approximately 60–70 thousand tonnes per year. This peak was driven primarily by exceptionally high carbon monoxide (CO) emissions (exceeding 40 thousand tonnes annually), accompanied by elevated levels of particulate matter (up to ~17 thousand tonnes), as well as NO_x and SO_2 . From the early 1990s onward, emissions declined markedly due to industrial contraction and technological modernization, falling to approximately 3 thousand tonnes by 2007. Since the early 2000s, pollutant levels have remained relatively low and stable compared to the late Soviet period, indicating a substantial long-term reduction in atmospheric emissions from the plant (Fig. 2). Industrial emission data up to 2007 were derived from Erlickytė (2007), whereas data for the subsequent period were obtained from the State Data Agency of Lithuania (<https://open-data.stat.gov.lt>).

The control site was established in Vaišnoriškė (55°42'N, 26°01'E), approximately 135 km southeast of the pollution-impacted site. The area is situated in a remote forested region with no significant industrial activity and is characterized by climatic conditions comparable to those of the study site. Daily meteorological data (air temperature and precipitation) used to evaluate potential climate-driven influences on $\delta^{13}\text{C}$ variability were obtained from the KNMI Climate Explorer. c2022. de Bilt: Koninklijk Nederlands Meteorologisch Instituut (KNMI); [accessed 2025 Nov 17]. <https://climexp.knmi.nl/start.cgi>, the official climate data portal of the World Meteorological Organization. The data were aggregated into mean annual temperature and total annual precipitation values (Fig. 3).

The climatic conditions at Akmenė and the control

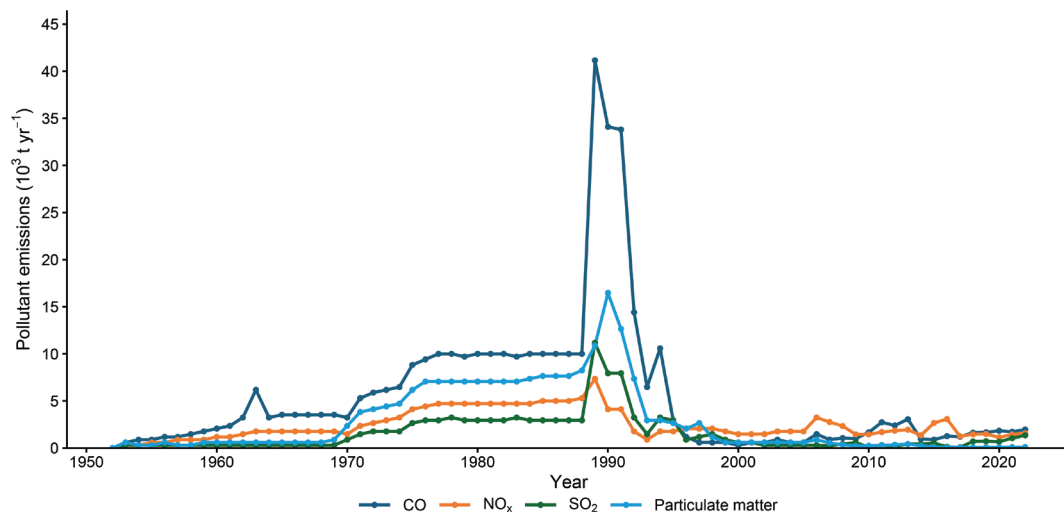


Fig. 2 Annual emissions of major air pollutants (CO, NO_x, SO₂, and particulate matter) from 1950 to 2022, expressed in 10³ t yr⁻¹. Emissions increased until the late 1980s – early 1990s, followed by a rapid decline and relatively low levels in subsequent decades

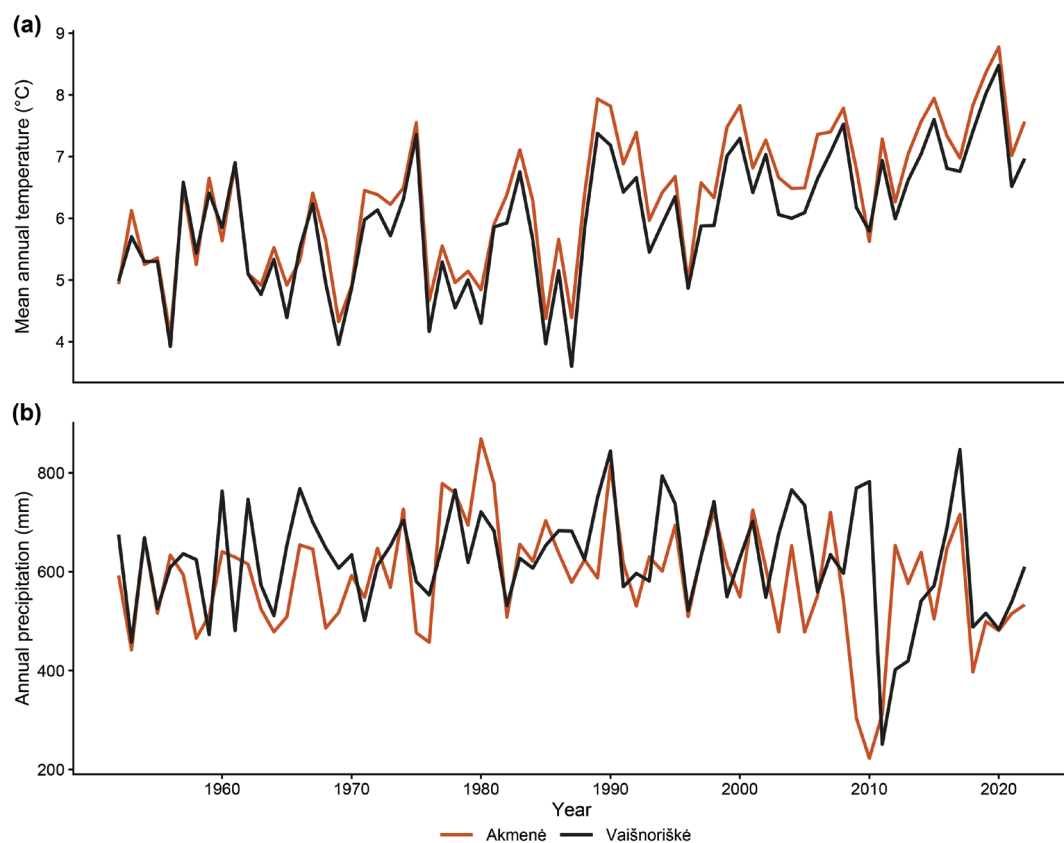


Fig. 3 Annual mean temperature (a) and annual precipitation (b) at the Akmenė and Vaišnorėškė sites from 1952 to 2022. Data obtained from the KNMI Climate Explorer (KNMI, 2025)

site in Vaišnorėškė are similar and reflect the humid climate of northern Lithuania. Over the common observation period, mean annual air temperature at both sites shows pronounced interannual variability superimposed on a clear long-term warming trend. Between 1950 and 2022, mean annual temperature increased by approximately 2.1 °C, corresponding to an average rate of ~ 0.035 °C yr⁻¹. In the earlier decades (1950s–1970s), mean annual temperatures typically ranged between

4.0 and 6.5 °C, whereas since the 1990s they have frequently exceeded 7.0 °C, reaching up to ~ 8.5 – 9.0 °C in the warmest years. Temperature records from the two sites are nearly identical in magnitude and variability, with a very strong correlation ($r = 0.994$, $p < 0.05$), indicating virtually synchronous temporal dynamics.

Annual precipitation at both locations averages 647 mm in Akmenė (representing the pollution-affected region) and 635 mm in Vaišnorėškė over the period

1950–2022. Interannual variability is considerable, with totals generally ranging from approximately 450 to 800 mm, and occasional extremes below 300 mm or above 850 mm. In contrast to temperature, no statistically significant long-term trend in total annual precipitation is evident. Precipitation series from the two sites are strongly correlated ($r = 0.725$, $p < 0.05$), demonstrating consistent regional moisture patterns despite year-to-year fluctuations.

Overall, the minimal differences in mean values, the strong inter-site correlations, and the shared warming trend confirm that Akmenė and Vaišnoriskė experience highly comparable climatic conditions, supporting robust comparisons of dendrochronological and stable isotope records between the sites.

Sampling and cross-dating

Scots pine (*Pinus sylvestris* L.) was chosen for this study because it predominates at both the impacted and reference sites and produces anatomically distinct annual rings with clearly separated earlywood and latewood, enabling precise dendrochronological and isotope-based analyses. At each location, eight dominant trees were selected for sampling. In October 2022, once cambial activity had ceased, four increment cores per tree were collected at breast height in orthogonal directions using a 5 mm Haglōf increment borer (Sweden), ensuring that the most recent annual ring was completely formed. The cores were carefully labelled and allowed to air-dry in paper holders prior to laboratory preparation.

For sample preparation, core surfaces were refined with a core microtome following Gärtner and Nievergelt (2010) to enhance ring visibility and prevent material carryover between adjacent growth increments. The prepared cores were digitized at a resolution of 3000 dpi using an Epson V700 flatbed scanner (Seiko Epson Corporation, Japan). Tree-ring widths were measured from the scanned images with CooRecorder 7.1.0.0 (Cybis Elektronik & Data AB, Sweden), and cross-dating quality was assessed using COFECHA (Holmes 1983; Grissino-Mayer 2001) to ensure accurate calendar-year assignment. Reliable cross-dating was critical for subsequent annual-scale comparisons of stable isotope ratios.

The developed tree-ring chronology spans 1933–2022 for Akmenė and 1895–2022 for Vaišnoriskė, the latter covering pre-industrial conditions. The comparatively shorter chronology at Akmenė is attributable to the lack of older individuals at the site. This limitation is not expected to bias interpretations, as previous research has demonstrated only a weak association between intrinsic water-use efficiency (*iWUE*) and tree age in Scots pine growing at similar boreal latitudes (Brienen *et al.* 2017).

Sample processing and $\delta^{13}\text{C}$ measurement

The dated tree rings were manually separated, pooled by calendar year, oven-dried at 60 °C for 6 h, and homogenized to fine powder using metal beads in 2 mL Eppendorf tubes for 20 min with a Spex Geno/Grinder 2010 mill. A total of eight dominant trees were sampled at each site, and pooling was performed to obtain a representative stand-level signal for inter-site comparison. The resulting samples, were analysed for whole-wood $\delta^{13}\text{C}$ using laser absorption isotope spectrometry. Restricting the analysis to the period with adequate replication ensured consistent and reliable inter-site comparisons. Approximately 1 mg of homogenized wood powder was weighed into tin capsules and combusted at 1020 °C in a combustion module (Costech Analytical Technologies, Inc.) coupled via a Picarro Liaison Interface A0301 to a Picarro Cavity Ring-Down Spectrometer (CRDS; G2121-i). Instrument calibration was performed using international reference materials IAEA-600, IAEA-CO-8, and SRM 4990C. Isotopic compositions were reported in delta notation ($\delta^{13}\text{C}$, ‰) relative to the Vienna Pee Dee Belemnite (VPDB) standard.

Because intrinsic water-use efficiency (*iWUE*) is commonly calculated from α -cellulose $\delta^{13}\text{C}$ values, and whole-wood measurements are typically depleted relative to cellulose, a constant offset of +1.0‰ was applied to approximate cellulose-equivalent values following Loader *et al.* (2003). While this correction does not account for potential temporal variability in whole-wood-cellulose offsets, it enables comparability with physiological models developed for cellulose-derived isotope data.

The corrected $\delta^{13}\text{C}$ values were converted to intrinsic water-use efficiency (*iWUE*) following the model of Farquhar *et al.* (1982). *iWUE* is defined as:

$$iWUE = \frac{A}{g} = \frac{c_a - c_i}{1.6} = \frac{c_a(1 - \frac{c_i}{c_a})}{1.6} \quad (1)$$

where A is the photosynthetic assimilation rate, g is stomatal conductance to water vapour, c_a is atmospheric CO_2 concentration, and c_i is intercellular CO_2 concentration.

Carbon isotope discrimination ($\Delta^{13}\text{C}$) was calculated as

$$\Delta^{13}\text{C} = \frac{\delta^{13}\text{C}_a - \delta^{13}\text{C}_p}{1 + \delta^{13}\text{C}_p} \quad (2)$$

where $\delta^{13}\text{C}_a$ and $\delta^{13}\text{C}_p$ denote atmospheric and plant isotopic compositions, respectively.

Discrimination is related to the ratio c_i/c_a according to:

$$\Delta^{13}\text{C} = a + (b - a) \frac{c_i}{c_a} \quad (3)$$

where $a = 4.4\text{‰}$ represents fractionation during diffusion through stomata and $b = 27\text{‰}$ represents fractionation during enzymatic carboxylation.

Tree-ring width (TRW) measurements were converted to basal area increment (BAI) to remove the geometrical effect of increasing stem circumference with age. BAI was calculated

$$BAI_t = \pi(R_t^2 - R_{t-1}^2), \quad (4)$$

where R_t and R_{t-1} represent stem radius at years t and $t-1$ respectively.

Inter-site physiological divergence

To examine potential shifts in tree physiological responses associated with the period of maximum regional industrial emissions, the record was divided into pre-1990 and post-1990 periods. The year 1990 was

selected as an analytical breakpoint because it coincides with the period of highest industrial emissions in the region (late 1980s – early 1990s) and the onset of divergence observed in the ΔBAI and $\Delta iWUE$ chronologies between the Akmenė and Vaišnorėškė sites. This transition coincided with the interval of highest industrial pollution in the region (approximately 1988–1993), associated with peak emissions from cement production (Fig. 4). Dividing the record at 1990 therefore provides a pragmatic separation between the period preceding the observed divergence and the subsequent phase characterized by declining emissions and changing growth and physiological responses.

Inter-site physiological divergence was quantified as the difference in intrinsic water-use efficiency between the two study sites:

$$\Delta iWUE = iWUE_{AKM} - iWUE_{VAI} \quad (5)$$

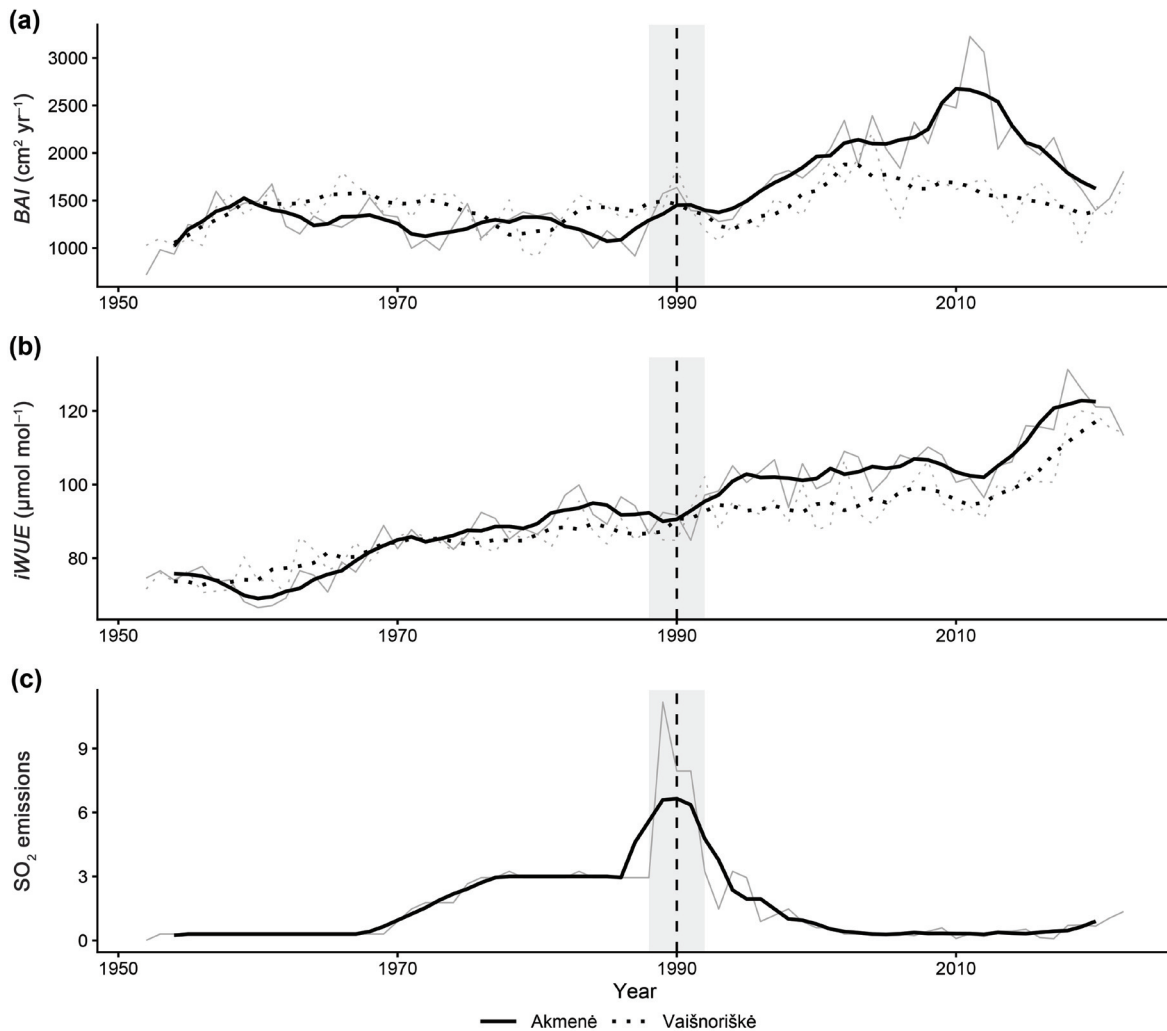


Fig. 4 Temporal variation in basal area increment (BAI), intrinsic water-use efficiency ($iWUE$), and industrial sulfur dioxide (SO_2) emissions in the study region. (a) BAI chronologies for Scots pine at the Akmenė and Vaišnorėškė sites. (b) Site chronologies of intrinsic water-use efficiency derived from tree-ring $\delta^{13}C$ measurements. (c) Annual SO_2 emissions associated with regional industrial activity. Thin grey lines represent annual values and thick lines show 5-year centered moving averages. The shaded area indicates the peak industrial pollution period (1988–1993), while the dashed vertical line marks the breakpoint year (1990) used to distinguish pre- and post-divergence periods in subsequent analyses

Using inter-site differences minimizes the influence of regional-scale drivers such as rising atmospheric CO₂ concentrations and shared climatic variability, thereby emphasizing site-specific physiological responses. Inter-site growth divergence was calculated analogously as the difference in basal area increment between sites:

$$\Delta BAI = BAI_{AKM} - BAI_{VAI} \quad (6)$$

To evaluate whether physiological and growth divergence differed between the two periods, statistical comparisons were performed on $\Delta iWUE$ and ΔBAI . Normality of variables was evaluated using Shapiro-Wilk tests and inspection of Q–Q plots (Fig. 5). Differences between the two periods were evaluated using Welch two-sample *t*-tests, which do not assume equal variances between groups. Because some variables showed minor deviations from normality, non-parametric Wilcoxon rank-sum tests were also performed to confirm the robustness of the results.

Relationships between $\Delta iWUE$ and ΔBAI were assessed using Pearson correlation analysis. Pearson and Spearman correlations were compared to ensure robustness of the results to potential deviations from normality. All statistical analyses were conducted in R (version 4.5.2) (R Core Team (2024)). R: A Language

and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria).

Climatic controls on $iWUE$ divergence

Climatic influences on $\Delta iWUE$ were first evaluated using Pearson correlation analysis between annual $\Delta iWUE$ and monthly inter-site differences in temperature (ΔT), precipitation (ΔP), and the Standardized Precipitation-Evapotranspiration Index ($\Delta SPEI$). Monthly climate variables were analysed from June of the previous growing season through August of the current growing season. For previous-season months (June–December), climatic values were assigned to the subsequent ring year to account for potential lagged physiological effects on growth and isotopic composition. To distinguish short-term and cumulative moisture influences, both three-month ($SPEI-3$) and six-month ($SPEI-6$) accumulation periods were analysed. Correlations were computed using complete cases only. To explore potential temporal instability in climate- $\Delta iWUE$ relationships, 20-year moving-window correlations were calculated. Because moving-window analysis reduces effective sample size and may introduce autocorrelation, these results were interpreted qualitatively and were not used for formal statistical inference.

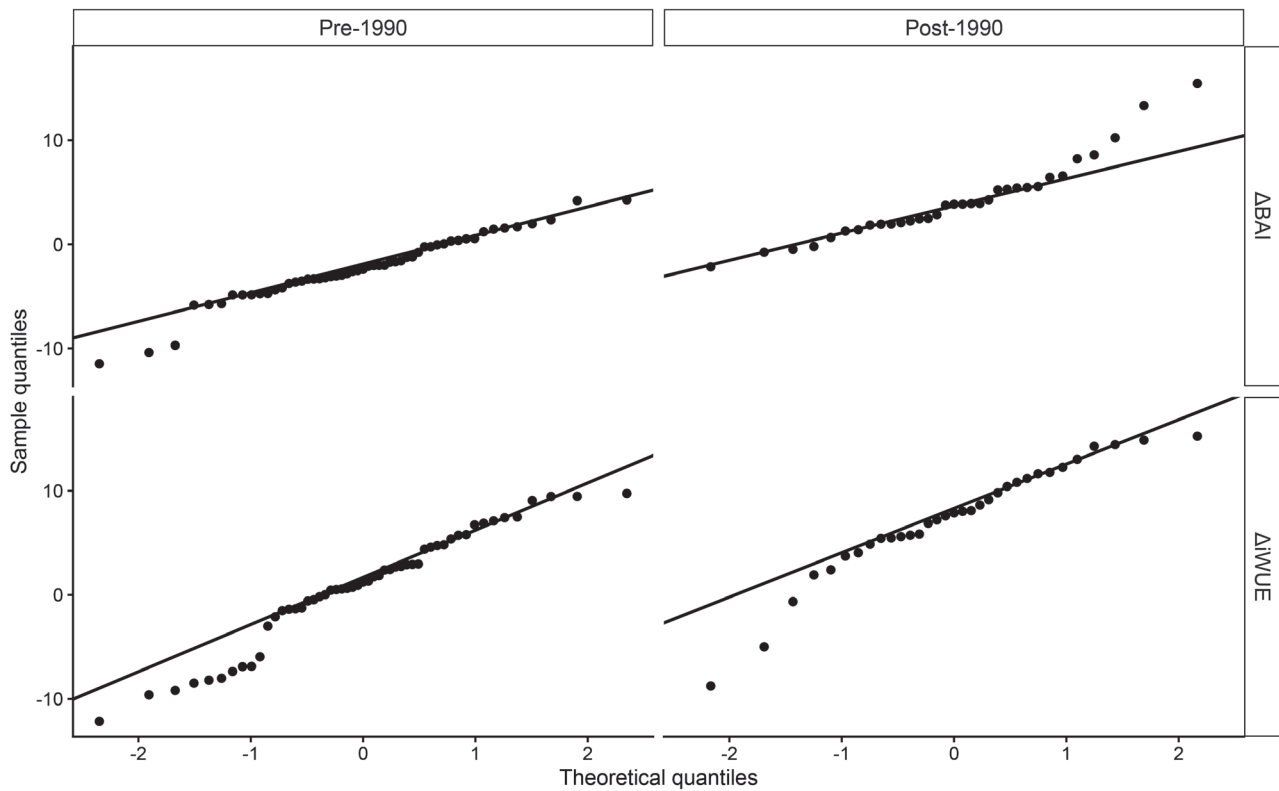


Fig. 5 Quantile–quantile (Q–Q) plots assessing normality of inter-site divergence in basal area increment (ΔBAI) and intrinsic water-use efficiency ($\Delta iWUE$) for the pre-1990 and post-1990 periods. Points lying close to the reference line indicate approximate normality of the data distributions used in subsequent statistical analyses

Generalized additive modelling of climatic and emission influences

Potential nonlinear relationships between $\Delta iWUE$ and environmental drivers were evaluated using generalized additive models (GAMs) with penalized regression splines implemented in the *R* package *mgcv*. Initial models evaluated climatic controls on $\Delta iWUE$ and included smooth terms for calendar year and inter-site differences in growing-season climate variables. Growing-season variables were derived from monthly data (May–August) and included differences in precipitation (ΔP_{gs}) and temperature (ΔT_{gs}). Drought indices (*SPEI*-3 and *SPEI*-6) were evaluated in analogous models.

To test potential anthropogenic influences beyond climatic divergence, annual industrial emission variables (CO , NO_x , SO_2) were incorporated individually as smooth terms:

$$\Delta iWUE \sim s(\text{Year}) + s(\Delta P_{gs}) + s(\text{Emission}) \quad (7)$$

Because combustion-related pollutants were strongly correlated (CO vs. SO_2 ; $r = 0.96$), each emission variable was evaluated in a separate model to avoid multicollinearity. Both same-year emissions and one-year lagged values were examined to account for potential delayed physiological responses. Model performance was evaluated using approximate *F*-tests for smooth terms, adjusted R^2 , deviance explained, and Akaike Information Criterion (*AIC*). Improvements in model fit following inclusion of emission variables were interpreted as evidence of associations independent of climatic divergence and long-term temporal trends. Statistical significance was assessed at $p < 0.05$.

Residual temporal autocorrelation was evaluated using autocorrelation function (*ACF*) diagnostics for the fitted GAMs. Residual autocorrelation was weak, indicating that the smooth term for year adequately captured the dominant temporal structure. As an additional robustness check, models incorporating a first-order autoregressive [*AR*(1)] structure were tested and yielded qualitatively similar results, supporting the validity of the inference.

RESULTS

Inter-site divergence in growth and water-use efficiency

Basal area increment (*BAI*) and intrinsic water-use efficiency (*iWUE*) chronologies revealed increasing divergence between the Akmenė and Vaišnoriškė sites beginning in the early 1990s (Fig. 6). Prior to 1990, growth at Akmenė was generally lower than at Vaišnoriškė, whereas in subsequent decades Akmenė

exhibited relatively higher growth. This shift was reflected in significant differences in inter-site growth divergence between the two periods. Mean ΔBAI increased from $-2.30 \text{ cm}^2 \text{ yr}^{-1}$ before 1990 to $4.14 \text{ cm}^2 \text{ yr}^{-1}$ afterwards (Welch $t = 8.08$, $\text{df} = 58.9$, $p < 0.001$). Inter-site physiological divergence showed a similar pattern. Mean $\Delta iWUE$ increased from $0.82 \mu\text{mol mol}^{-1}$ before 1990 to $7.39 \mu\text{mol mol}^{-1}$ afterwards (Welch $t = 5.46$, $\text{df} = 68.6$, $p < 0.001$), indicating a pronounced increase in water-use efficiency differences between the two sites.

Inter-site differences in intrinsic water-use efficiency were also positively associated with growth divergence. $\Delta iWUE$ and ΔBAI were moderately correlated across the study period (Pearson $r = 0.45$, $p < 0.001$; Fig. 7), indicating that years with greater physiological divergence between sites also tended to exhibit larger differences in radial growth. Spearman rank correlation produced similar results ($\rho = 0.49$, $p < 0.001$), confirming the robustness of this relationship.

Climate relationships with $\Delta iWUE$

Monthly correlations between $\Delta iWUE$ and climatic variables were generally weak to moderate (Fig. 8). The strongest positive associations were observed for inter-site temperature differences during the current growing season, particularly in July ($r = 0.50$) and August ($r = 0.38$), with the July correlation being statistically significant ($p < 0.001$). Moderate positive correlations were also observed during the late previous growing season, especially from July to November of the previous year ($r \approx 0.27\text{--}0.33$). In contrast, full-period correlations with inter-site precipitation differences and drought conditions (ΔSPEI -3 and ΔSPEI -6) were generally weaker and less consistent across months.

Generalized additive models (GAMs) were subsequently used to evaluate broader climatic controls on $\Delta iWUE$. A model including only differences in drought conditions between sites (ΔSPEI 6) did not significantly explain variation in $\Delta iWUE$ ($F = 1.29$, $p = 0.355$), accounting for only 3% of the total deviance. When calendar year was included in the model, a significant nonlinear temporal trend in $\Delta iWUE$ emerged ($p < 0.001$), whereas ΔSPEI 6 remained non-significant ($p = 0.63$). This model explained approximately 36% of the deviance in $\Delta iWUE$. Similarly, models including *SPEI*6 for the Vaišnoriškė site or short-term drought differences between sites (ΔSPEI 3) showed significant nonlinear temporal trends ($p < 0.001$) but no significant effects of drought variables ($p = 0.10$ and $p = 0.19$, respectively), explaining approximately 37–38% of the deviance.

In contrast, differences in growing-season cli-

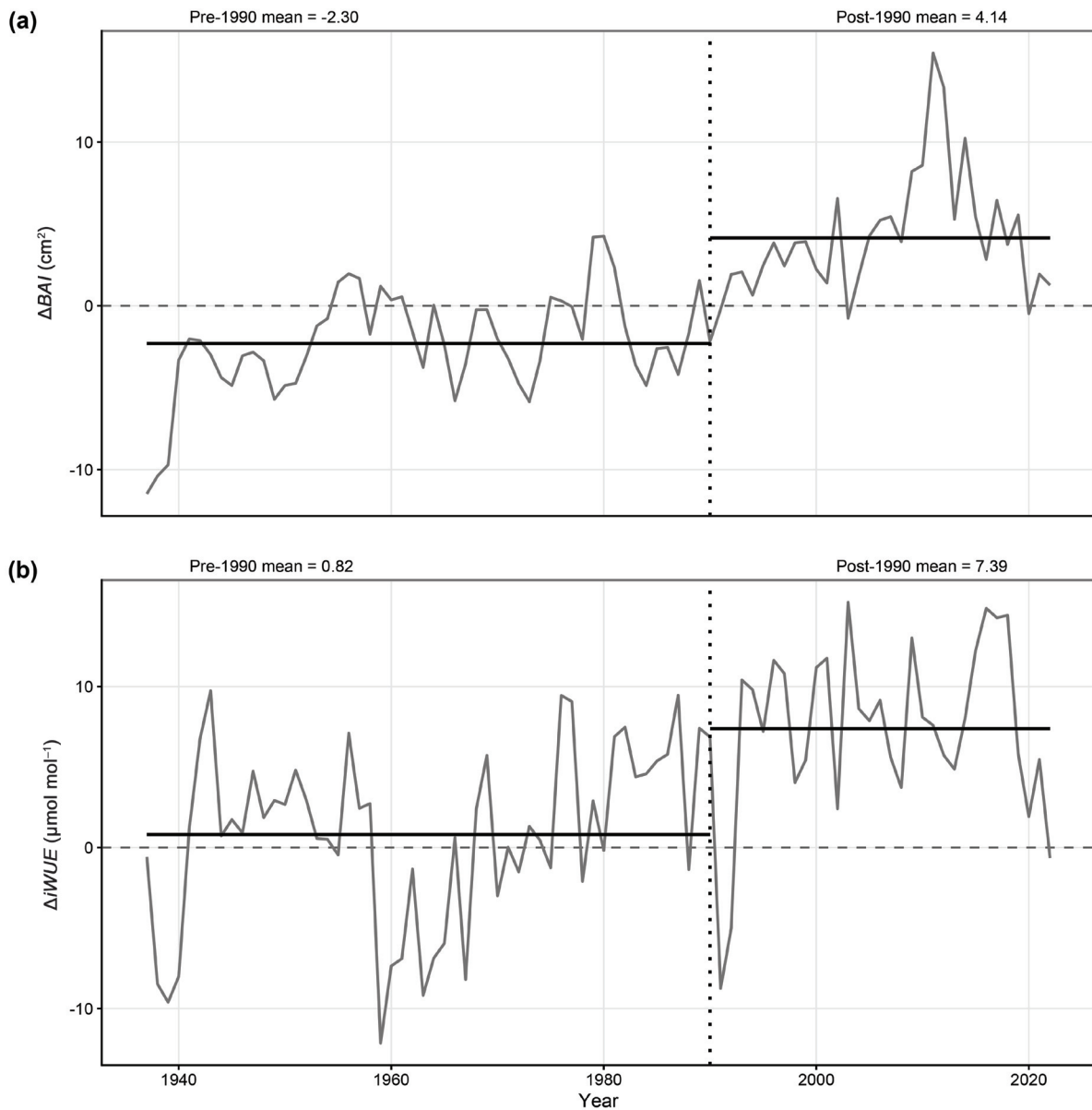


Fig. 6 Temporal dynamics of inter-site divergence in (a) basal area increment (ΔBAI) and (b) intrinsic water-use efficiency ($\Delta iWUE$) between the Akmenė and Vaišnorėškė sites. Grey lines represent annual inter-site differences. The vertical dotted line marks the 1990 breakpoint separating the pre-1990 and post-1990 periods, the horizontal dashed line indicates zero difference between sites, and the solid horizontal lines indicate the mean values for each period. Inter-site divergence increased after 1990, with mean ΔBAI shifting from negative to positive values and mean $\Delta iWUE$ showing a pronounced increase

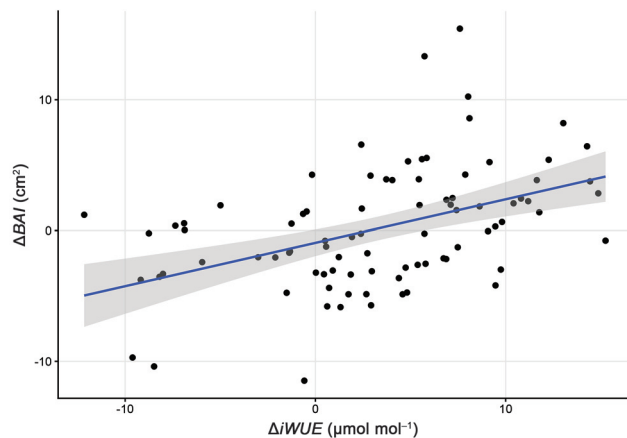


Fig. 7 Relationship between inter-site differences in intrinsic water-use efficiency ($\Delta iWUE$) and basal area increment (ΔBAI) for Scots pine at the Akmenė and Vaišnorėškė sites over the study period. Points represent annual values. The solid line shows the linear regression fit with 95% confidence interval (shaded area). A positive relationship indicates that years with larger physiological divergence between sites were associated with greater differences in radial growth

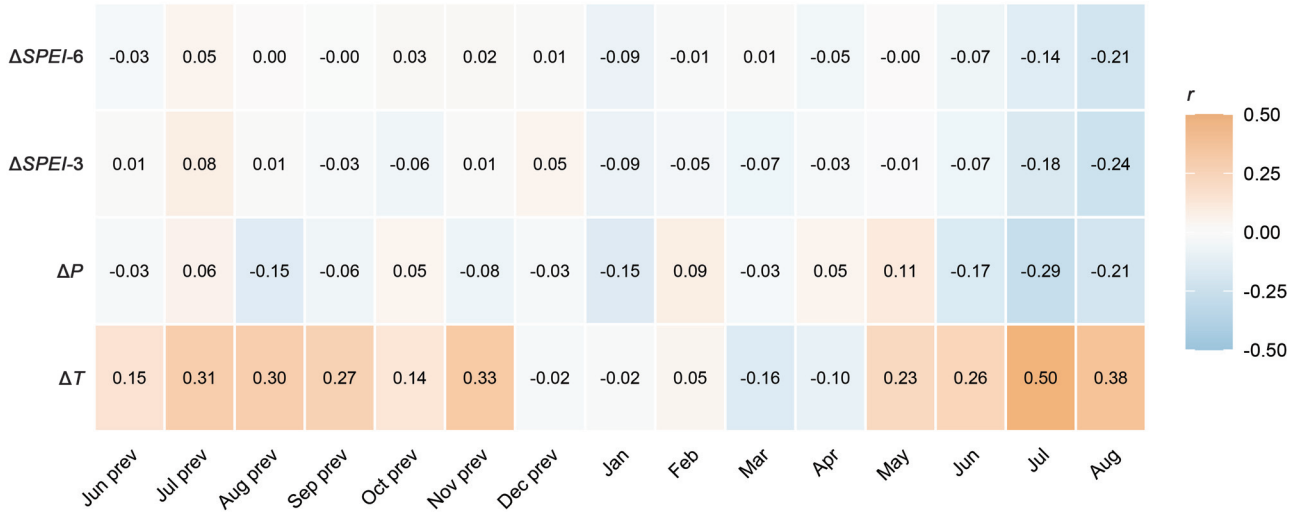


Fig. 8 Monthly correlations between inter-site differences in intrinsic water-use efficiency ($\Delta iWUE$) and climatic variables from June of the previous year to August of the current year. Climatic variables include inter-site differences in temperature (ΔT), precipitation (ΔP), and drought conditions represented by ΔS_{PEI-3} and ΔS_{PEI-6} . Numbers within cells represent Pearson correlation coefficients

mate between sites showed stronger relationships with $\Delta iWUE$. A generalized additive model including smooth terms for calendar year and growing-season precipitation differences revealed a significant non-linear effect of ΔP_{gs} ($F = 5.45$, $p = 0.0017$). The model explained approximately 63.8% of the deviance in $\Delta iWUE$, indicating that differences in growing-season moisture supply between sites were strongly associated with divergence in water-use efficiency. The partial smooth effect of ΔP_{gs} shows a nonlinear pattern, with positive effects at intermediate negative values and a gradual shift toward negative effects at higher ΔP_{gs} . Uncertainty increases toward the extremes of the prediction range (Fig. 9).

Influence of industrial emissions on $\Delta iWUE$

Simple linear regression models were first used to evaluate potential associations between $\Delta iWUE$ and annual industrial emissions (SO_2 , CO , NO_x). None of the pollutants exhibited statistically significant linear relationships with $\Delta iWUE$, and all models explained $\leq 1.6\%$ of variance (all $R^2 \leq 0.016$; $p > 0.05$), indicating that linear models explained little of the observed variability.

Generalized additive models (GAMs) were subsequently used to evaluate potential nonlinear relationships between $\Delta iWUE$ and individual pollutants. Univariate GAMs revealed no significant associations for NO_x or dust emissions. In contrast, CO emissions showed a significant nonlinear relationship with $\Delta iWUE$ in a univariate GAM, explaining approximately 24% of the deviance (Fig. 10a).

Because both $\Delta iWUE$ and emission series exhibit strong temporal structure, an additional GAM includ-

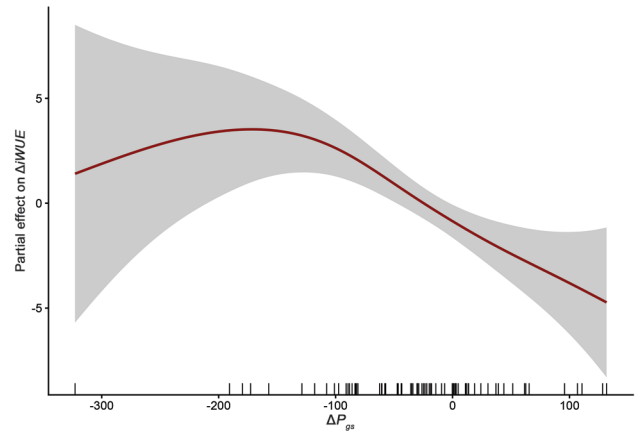


Fig. 9 Partial smooth effect of inter-site differences in growing-season precipitation (ΔP_{gs}) on intrinsic water-use efficiency divergence ($\Delta iWUE$) derived from a generalized additive model of the form $\Delta iWUE \sim s(\text{Year}) + s(\Delta P_{gs})$. The solid line represents the fitted smooth function and the shaded area indicates the 95% confidence interval. Rug marks along the x-axis indicate the distribution of observations

ing smooth terms for calendar year and CO was fitted ($\Delta iWUE \sim s(\text{Year}) + s(CO)$) (Fig. 10b). In this model, the smooth of Year was highly significant ($p < 2 \times 10^{-16}$) and accounted for the majority of structured variance, whereas the smooth term for CO was no longer statistically significant ($p = 0.13$). This suggests that part of the apparent emission signal reflects shared long-term temporal trends between $\Delta iWUE$ and regional industrial activity.

To evaluate potential delayed responses, one-year lagged emission variables were incorporated into climate-controlled GAMs of the form:

$$\Delta iWUE \sim s(\text{Year}) + s(\Delta P_{gs}) + s(\text{Emission}) \quad (8)$$

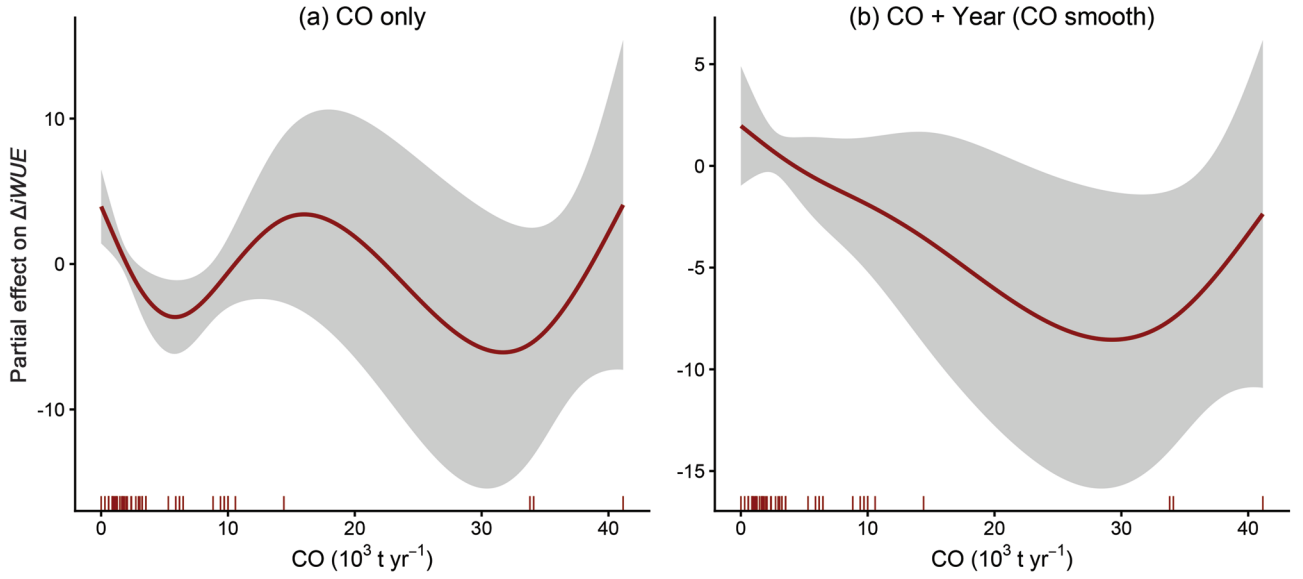


Fig. 10 Nonlinear relationships between $\Delta iWUE$ and carbon monoxide (CO) emissions derived from generalized additive models. (a) Univariate GAM showing a nonlinear association between $\Delta iWUE$ and CO emissions. (b) GAM including smooth terms for calendar year and CO, illustrating the strong temporal trend shared by $\Delta iWUE$ and industrial emissions. Shaded areas represent 95% confidence intervals and tick marks indicate data density

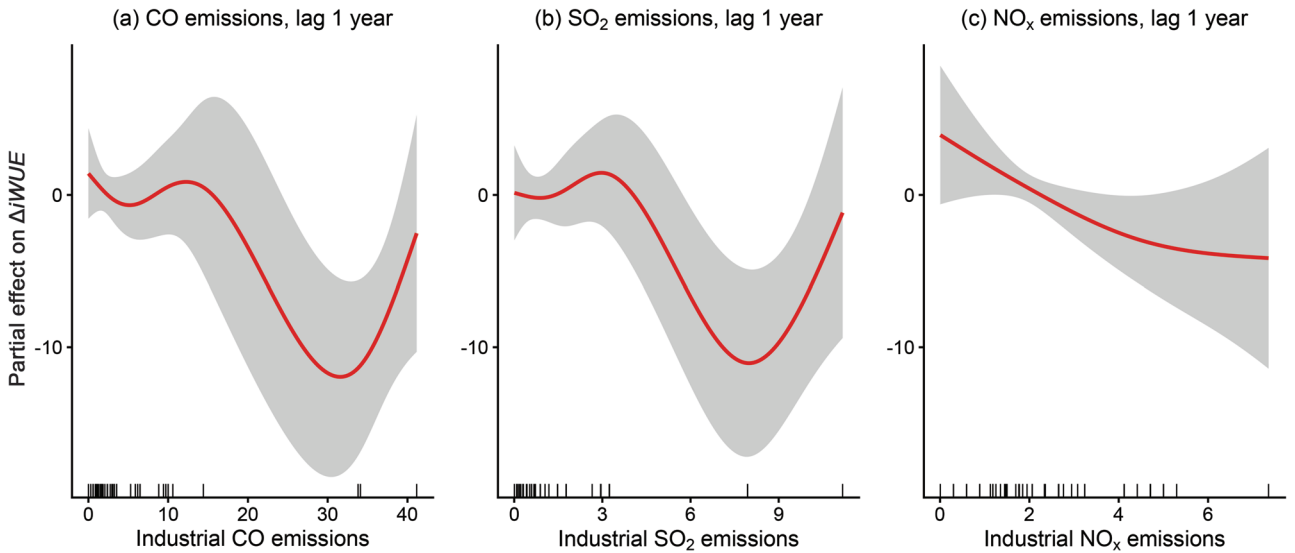


Fig. 11 Partial smooth effects of one-year lagged industrial emissions on inter-site differences in intrinsic water-use efficiency ($\Delta iWUE$) derived from climate-controlled generalized additive models. Panels show nonlinear responses of $\Delta iWUE$ to (a) carbon monoxide (CO_{lag1}), (b) sulfur dioxide (SO_{2lag1}), and (c) nitrogen oxides (NO_{xlag1}). Each curve represents the fitted smooth from models of the form $\Delta iWUE \sim s(\text{Year}) + s(\Delta P_{gs}) + s(\text{Emission})$. Red lines indicate the estimated smooth functions and grey shading represents 95% confidence intervals. Rug marks along the x-axes indicate the distribution of observations

Including lagged CO emissions revealed a significant nonlinear association with $\Delta iWUE$ ($p = 0.003$) after accounting for climatic divergence and temporal structure. The response curve indicated a nonlinear, threshold-like relationship, with increased divergence at intermediate to high lagged emission levels, although uncertainty increased toward the extremes of the emission range due to reduced data density.

Additional models incorporating other pollutants showed weaker or inconsistent support. Nitrogen oxide emissions (NO_{xlag1}) showed no significant rela-

tionship with $\Delta iWUE$ ($p = 0.13$). In contrast, lagged sulfur dioxide emissions (SO_{2lag1}) showed a significant nonlinear association with $\Delta iWUE$ ($p = 0.002$) (Fig. 11). Because combustion-related emission series were strongly correlated, each pollutant was evaluated in separate models to avoid multicollinearity.

Model comparison based on AIC indicated that the model including SO_{2lag1} provided the lowest AIC value (AIC = 405.4), followed by the CO model, whereas the NO_x model showed weaker support. CO and SO_2 emissions were strongly correlated ($r = 0.96$), indicat-

ing that both variables likely represent overall combustion intensity rather than pollutant-specific toxicity. Models including lagged CO or SO₂ emissions explained up to 66–67% of the deviance in $\Delta iWUE$, indicating improved model performance relative to climate-only models. Together, these results indicate that although climatic divergence between sites explains most of the variability in $\Delta iWUE$, lagged industrial emissions – particularly CO and SO₂ – contribute additional nonlinear effects after accounting for temporal trends and hydroclimatic differences.

DISCUSSION

The chronologies revealed a clear divergence between the Akmenė and Vaišnoriškė sites beginning in the early 1990s. This shift was reflected in both physiological and growth metrics: $\Delta iWUE$ increased significantly after 1990, while ΔBAI shifted from negative to positive values, indicating a reversal in relative growth performance between sites (Fig. 6). The positive correlation between $\Delta iWUE$ and ΔBAI further suggests that physiological divergence was accompanied by increasing differences in radial growth (Fig. 7).

Such patterns may reflect physiological adjustments of Scots pine to changing environmental conditions during the late twentieth century. Rising atmospheric CO₂ concentrations, regional climatic variability, and changing pollution levels have all been shown to influence stomatal regulation and carbon assimilation in boreal and temperate forests (Peñuelas *et al.* 2011; Körner 2015; Sensuła *et al.* 2015, 2017; Guerrieri *et al.* 2019). However, increases in intrinsic water-use efficiency do not necessarily translate directly into enhanced radial growth, as growth responses are often constrained by additional factors such as nutrient availability, stand dynamics, or climatic stress (Körner 2015; Guerrieri *et al.* 2019).

Climatic dominance in regulating inter-site $iWUE$ divergence

The present study indicates that inter-site differences in intrinsic water-use efficiency ($\Delta iWUE$) were primarily governed by climatic variability, with differences in growing-season precipitation between sites emerging as the strongest supported climatic predictor. Among the generalized additive models tested, the model including ΔP_{gs} explained the largest proportion of deviance and substantially improved model performance relative to models containing only temporal structure (Fig. 9). This result suggests that site-level differences in growing-season moisture supply played a central role in regulating physiological divergence between the Akmenė and Vaišnoriškė stands.

Exploratory monthly correlation analysis revealed the strongest individual climatic associations for inter-site temperature differences, particularly during July of the current growing season (Fig. 8). This indicates that relatively warmer conditions at Akmenė compared to Vaišnoriškė may contribute to increased divergence in $iWUE$ during certain months. However, these monthly relationships were weaker and less consistent than the integrated GAM response to growing-season precipitation differences, indicating that precipitation differences provided a more robust explanation of interannual divergence at the seasonal scale.

In contrast, drought indices based on *SPEI* did not significantly explain $\Delta iWUE$ in the GAM framework once temporal structure was considered. This suggests that, in the present dataset, direct differences in growing-season precipitation captured the relevant hydroclimatic divergence between the sites more effectively than standardized drought metrics. Accordingly, the climatic interpretation of $\Delta iWUE$ should be framed primarily in terms of differences in growing-season moisture availability rather than in terms of *SPEI*-derived drought severity.

The strong and persistent smooth of Year across models likely reflects the combined influence of rising atmospheric CO₂, regional warming, and long-term physiological adjustment common to both stands. By modelling inter-site differences and explicitly controlling for calendar year, the analysis isolates deviations from this shared background trajectory and emphasizes drivers of divergence rather than absolute long-term change.

Industrial emissions as secondary modulators of physiological divergence

Although climatic factors explained the majority of structured variance in $\Delta iWUE$, models incorporating industrial emission variables indicated an additional influence of regional pollution history. Lagged combustion-related emissions, particularly CO_{lag1}, showed significant nonlinear associations with $\Delta iWUE$ after accounting for temporal structure and climatic divergence. Importantly, same-year pollutant effects were weak or absent once calendar year was included in the models (Fig. 10). This suggests substantial temporal confounding between industrial activity and broader environmental trends such as rising atmospheric CO₂ concentrations and regional climatic change.

The emergence of significant one-year lagged effects points to delayed or cumulative physiological responses rather than immediate stomatal closure. Such delays may reflect indirect pathways, including progressive oxidative stress, altered nutrient cycling, or

interactions between pollutant exposure and drought sensitivity (Matyssek *et al.* 2017; Tupu *et al.* 2026). Given the strong correlation among combustion-related emission series, CO likely represents overall combustion intensity rather than pollutant-specific toxicity. Exploratory models using SO₂ showed similar temporal patterns and, in some cases, stronger statistical associations with $\Delta iWUE$. In contrast, NO_x effects were generally weak and inconsistent, which may partly reflect the approximately 3.1 km distance between the sampling site and the emission source (Fig. 11).

Within the $\Delta iWUE$ framework, industrial pollution does not appear to generate physiological divergence independently. Instead, pollution may modulate the magnitude of divergence that develops under moisture-driven climatic forcing. Industrial influence therefore emerges as a secondary, context-dependent factor superimposed on a primarily climate-controlled system. Similar conclusions have been drawn from dendrochronological studies of Scots pine near industrial sources in Lithuania, where growth responses reflected the combined influence of emission history and climatic variability rather than a simple direct pollution signal (Juknys *et al.* 2003; Erlickytė, Vitas 2008; Stravinskienė *et al.* 2013; Augustaitis *et al.* 2014).

Relative importance of climate and emissions

Model comparisons clearly indicate that climatic and long-term temporal factors dominate the regulation of $\Delta iWUE$. The baseline climate model explained approximately 64% of the deviance, whereas inclusion of lagged emission variables increased explained deviance to approximately 66–67%. Thus, industrial emissions contributed additional explanatory power but did not override the dominant climatic control.

This hierarchical structure aligns with physiological expectations. Moisture availability directly constrains stomatal conductance, whereas pollution effects are more likely to operate indirectly through physiological stress or interactions with drought conditions (Farquhar, Sharkey 1982; Jones 1998; Matyssek *et al.* 2014; Körner 2015). The divergence observed during the late twentieth century may therefore reflect an interaction between peak industrial activity and contemporaneous moisture stress rather than direct suppression of carbon assimilation.

Within this framework, $\Delta iWUE$ represents differential sensitivity between stands experiencing broadly similar regional forcing. Divergence increases when cumulative moisture conditions differ between sites, while industrial emissions may amplify or dampen this divergence under particular environmental conditions.

Methodological implications

The obtained results highlight the importance of explicitly accounting for long-term temporal structure when evaluating pollution effects in tree-ring isotope records. Univariate nonlinear models suggested apparent emission- $iWUE$ relationships; however, these associations were substantially reduced when calendar year was included (Fig. 10). Without controlling for temporal trends, coincident changes in industrial activity, atmospheric CO₂ concentration, and climate may be incorrectly interpreted as causal pollution signals. The modelling approach used here therefore helps distinguish genuine environmental drivers from shared long-term trends. The inter-site difference framework proved particularly useful for isolating differential responses under broadly shared climatic forcing. By removing common regional signals, this approach enhances sensitivity to site-specific modifiers while reducing the risk of spurious correlations.

Limitations and uncertainties

Several limitations should be considered. First, whole-wood $\delta^{13}C$ values were adjusted using a constant cellulose offset, which may not fully capture potential temporal variability in compound-specific fractionation (Loader *et al.* 2003; McCarroll, Loader 2004). Second, industrial emission data represent regional activity and were used as proxies for regional pollution pressure rather than direct measurements of site-level exposure. Because factors such as atmospheric transport, deposition, and local canopy interactions were not explicitly reconstructed, the models do not represent pollutant doses at the tree level. Therefore, the identified relationships should be interpreted as associations with regional industrial activity history rather than pollutant-specific dose-response relationships. The contrast between Akmenė and Vaišnorėškė reflects differences in proximity to major emission sources but remains an approximation in the absence of direct air quality or deposition measurements at the study sites. Third, strong collinearity among combustion-related pollutants limits attribution to specific chemical mechanisms. Nevertheless, the consistency of moisture-driven divergence and the reproducibility of lagged emission signals across model specifications support the interpretation that industrial emissions exert a secondary influence superimposed on dominant climatic regulation.

CONCLUSIONS

Inter-site differences in intrinsic water-use efficiency ($\Delta iWUE$) of Scots pine were primarily regulated by climatic variability, supporting our first hypothesis.

Differences in growing-season precipitation between sites emerged as the strongest climatic predictor of physiological divergence, indicating that variation in site-level moisture availability plays a central role in regulating water-use efficiency responses.

Consistent with our second hypothesis, industrial emissions showed additional but subordinate associations with $\Delta iWUE$ after accounting for shared temporal structure. Apparent same-year emission effects largely disappeared when long-term trends were included in the models, highlighting the importance of controlling for coincident changes in climate and atmospheric composition.

Supporting our third hypothesis, combustion-related emissions exhibited delayed and nonlinear relationships with inter-site physiological divergence. Lagged emission variables improved model performance and revealed threshold-like responses, suggesting indirect or cumulative physiological modulation during periods of elevated industrial activity. Among the pollutants examined, sulfur dioxide showed the strongest statistical association with $\Delta iWUE$, while carbon monoxide exhibited similar patterns consistent with overall combustion intensity, whereas nitrogen oxides showed no consistent effect.

Overall, these results indicate that climatic variability – particularly differences in growing-season moisture availability – dominates the regulation of inter-site intrinsic water-use efficiency in Scots pine, while industrial emissions act as secondary and temporally lagged modifiers rather than primary drivers.

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Declaration of competing interests

The authors declare that they have no known financial or personal competing interests that could have influenced the work reported in this manuscript. During compilation, the authors used Grammarly for language editing to improve clarity and style. The research design, data analysis, interpretation, and scientific conclusions were developed solely by the authors. The authors reviewed and approved all resulting changes and take full responsibility for the content of the publication.

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