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# Reduction determinants of Scots pine tree-ring width in the vicinity of Puławy industrial plants (central-eastern Poland)

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***Abstract***

Scots pine (*Pinus sylvestris* L.) is a tree species that reacts to meteorological conditions, industrial pollution and groundwater level. The aims of this study were to evaluate reductions and increases in Scots pine tree-ring widths (VTRW) as compared to reference sequences for sites in two types of forest habitat found in the vicinity of the chemical plant complex in Puławy, located in central-eastern Poland, and to determine the relationship between VTRW, climate and anthropogenic factors. A considerable reduction in tree-ring widths compared with reference sites was found for sites affected by chemical plant emissions from the end of the 1960s, i.e. right after the start of the Puławy industrial plants, to the beginning of the 1990s (implementation of new technologies). This period of reduction in tree-ring width lasted longer and was greater in magnitude in the pure Scots pine stands studied within a coniferous mixed forest habitat. Furthermore, the reduction in Scots pine tree-ring width was largely dependent on below-average air temperature in the period from December of the year preceding ring formation until January of the current year and above-average total precipitation from June until August above-average $SO_2$, $NH_3$ and NOx emissions and above-average surface and underground water withdrawals from company intakes. The influence of weather on VTRW was weakened by anthropogenic factors, which affected the quality of the local natural environment. Therefore, Scots pine tree-ring width may be used to evaluate the state of the natural environment and threats it is facing in areas with high levels of industrial pollution including, for example, $SO_2$, $NH_3$ and $NO_x$.

**Keywords:** temperature and precipitation conditions, air pollution, tree-ring width, forest research area, dendroecological studies, Central Europe, *Pinus sylvestris* L.

## Introduction

Scots pine (*Pinus sylvestris* L.) annual tree-ring growth occurs due to the activity of the vascular cambium, which is periodic and follows the seasons. The width of earlywood (produced in spring) and latewood (produced at the end of summer) in a given year depends on many biotic, abiotic and anthropogenic factors. Among the most important of these factors are the genetic characteristics of the tree species, ecological community relationships and soil and climate conditions (Wilczyński 2006, Hordo et al. 2009, Koprowski et al. 2012, Misi et al. 2019). Forest stand management practices and environmental pollution also influence tree growth (Erlickytė and Vitas 2008, Malik et al. 2012).

Pines tolerate many types of soils but grow best in sandy soils that are well drained and have a pH in the range of 4.0 to 7.0 (Marinich and Powell 2017). Pines do not like floodplains and sudden fluctuations in water content in the soil profile. Drought is also problematic, as it interferes with water and nutrient regulation and, in the case of older forest stands, it contributes to a pathological reduction of needles, reduces seed yield and produces dry limbs (Dueck et al. 1998, Popkova et al. 2018). The studies of Gonthier et al. (2010) showed a strong and significant correlation between tree ring widths and the aridity index in summer.

Moreover, the tree-ring width of conifers growing in areas affected by industrial pollution, especially those in the vicinity of industrial plants with high environmental impact, may be used as an indicator of environment degradation (Parobeková et al. 2016) or as an archive of past environmental pollution (Sensuła et al. 2017). The changes in the width of Scots pine tree rings can be used to determine when a period of influence from industrial pollution began and ended, the number of tree rings with reduced growth (and the degree to which their width was affected), and the intensity of the pollution influence on the stands studied (Augustaitis et al. 2014, Austruy et al. 2019, Putalová et al. 2019, Ziemiańska et al. 2019).

The chemical plant complex in Puławy was one of the industrial plants having the biggest environmental impact in Poland. It emitted pollutants during the technological process of producing mineral fertilisers (e.g. urea, ammonium nitrate) and chemical compounds (e.g. melamine, liquid caprolactam). In 1969–1971, industrial emissions caused destruction of 500 ha of the forest and visibly damaged approx. 2,000 ha of the forest (Falencka-Jabłońska 2009). Until 1994, nearly 750 ha of forest ecosystems became destroyed and as much as 85% of the forest area in the Puławy Forest District was exposed to high industrial emission (Falencka-Jabłońska 2009). The first actions reducing air pollution and revegetation activities in the deforested areas were undertaken in the second half of the 1980s.

The aims of this study were to evaluate reductions and increases in Scots pine tree-ring widths in coniferous mixed forests and broadleaf mixed forests, as well as their dependence on climate and anthropogenic factors, specifically for stands in the vicinity of Zakłady Azotowe Puławy (a complex of nitrogen plants in Puławy), located in central-eastern Poland. The study put forward a hypothesis that tree stands overgrowing a less fertile habitat are more vulnerable to the variability of meteorological conditions, deposition of $SO_2$, $NH_3$ and $NO_x$, and changes in the amounts of withdrawn surface and underground waters.

## Materials and methods

### *Research location*

The forest research areas (FRAs) were in nine forest divisions of the Puławy Forest District, viz. in the vicinity of Zakłady Azotowe Puławy (Figure 1, Table 1), within


**Figure 1.** Location of the FRAs in coniferous mixed forests (B85, B109, B116, B142, B176c – reference station) and in broadleaf mixed forests (F118, F126, F143, F176c – reference station), as well as the distribution of meteorological stations (METEO) located in the region of Zakłady Azotowe Puławy in Puławy Forest District (central-eastern Poland)

**Table 1.** Characteristics of FRAs in Puławy Forest District where Scots pine stem core samples were collected

| No. | Signature of forest research areas as in Fig. 1 / no. of samples (Items) | GPS coordinates of the forest research areas | Distance from the highest emitter of Zakłady Azotowe Puławy | | Share of the tested species in the forest research areas (%) | Habitat conditions | | |
|---|---|---|---|---|---|---|---|---|
| | | | (km) | direction | | forest habitat type | type of soil | type of vegetation cover |
| 1 | B85 / 27 | 51.482188 N 21.980831 E | 2.7 | N | 100 | B[1] | podzolic rustry soils | covered by grass |
| 2 | B109 / 25 | 51.472847 N 22.022490 E | 3.7 | NE | 100 | | typical rustry soils | covered with moss-bilberry |
| 3 | B116 / 22 | 51.473380 N 22.147932 E | 12.2 | E | 100 | | typical rustry soils | fully covered by grass |
| 4 | B142 / 23 | 51.443829 N 22.028203 E | 4.1 | SE | 100 | | typical rustry soils | covered by grass |
| 5 | B176c / 25 | 51.419045 N 21.921351 E | 5.8 | SW | 100 | | typical rustry soils | covered by grass |
| 6 | F118 / 25 | 51.476951 N 22.140849 E | 12.0 | E | 80 | F[2] | brown rustry soils | fully covered by weeded |
| 7 | F126 / 31 | 51.471712 N 22.140291 E | 11.9 | E | 70 | | typical gley-podzols | covered by grass |
| 8 | F143 / 22 | 51.443331 N 22.024899 E | 4.2 | SE | 70 | | brown rustry soils | fully covered with herbaceous plants |
| 9 | F176 c / 23 | 51.421325 N 21.921516 E | 5.7 | SW | 80 | | brown rustry soils | covered by grass |

Notes: [1] – coniferous mixed forest, [2] – broadleaf mixed forest, c – reference site (B176c, F176c).

two habitats of medium fertility: coniferous mixed forests (habitat B) and broadleaf mixed forests (habitat F). Five FRAs were chosen in habitat B (B85, B109, B116, B142, B176c), including one reference site (B176c), and four FRAs (F118, F126, F143, F176c) were selected in habitat F, including one reference site (F176c). The FRAs were located at different distances from Zakłady Azotowe Puławy – from 2.7 km to 12.2 km in the case of habitat B and from 4.2 km to 12.0 km in the case of habitat F – as well as in different directions with respect to the highest-emitting plant – from north to southwest; locations were determined by a group of selection attributes including age of forest stands and type of habitat. The reference areas were selected due to their location in relation to the chemical plant (opposite to the prevailing winds) and a relatively good health condition of the stands (based on the information from the Puławy Forest District). In habitat B, 100% of the trees in the FRAs were Scots pine, while in habitat F 70–80% were Scots pine. Detailed information on the soil types and ground cover of the analysed FRAs is presented in Table 1.

The Puławy Forest District is in the northwest part of Lubelskie Voivodeship, located in central-eastern Poland, about 140 km from Warsaw. Starting from the end of the 1960s, the area in which this dendroecological research has been conducted was subjected to industrial pollution from Zakłady Azotowe Puławy, one of the industrial plant complexes in Poland with the greatest ecological impact. The biggest degradation of the natural environment occurred east of Zakłady Azotowe Puławy, which was taken into consideration when selecting FRAs for this study.

### *Climate conditions*

The meteorological data used in the paper were collected at the meteorological station in Puławy, whereas the data about wind direction – additionally, at the station in Osiny. The stations are situated south and east of Zakłady Azotowe Puławy, respectively (Figure 1).

Thermal conditions in Puławy during the period 1930–2015 ranged from −3.1°C in January up to 18.9°C in August; sub-zero monthly mean air temperatures also occurred in February (−2.0°C) and December (−0.8°C) (Figure 2). The largest month-to-month air temperature increases (an average of 6.3°C) were recorded between March and April. The average annual total precipitation was only about 573 mm, ranging from about 30 mm per month in January–March (with a low of 28.6 mm in February) up to almost 81 mm in July. Total winter precipitation (about 94 mm) was slightly less than half of the total summer precipitation. The winds were most often from the NW (20.4%), followed by winds from the S (15.6%), SE (15.3%) and W (15.0%); the least frequent wind directions were from the NE (4.4%) and N (7.5%). Calm was recorded during fewer than 3% of days in a year.

### *Air pollution and industrial water withdrawals*

This study used annual air pollution emission data for 1970–2015 regarding pollutants such as $SO_2$, $NH_3$ and $NO_x$ that were emitted from Zakłady Azotowe Puławy, as well as data concerning the amount of surface and underground water withdrawals from company intakes in the years 1985–2015. Data regarding both air pollutant emissions and water withdrawals were shared by Zakłady Azotowe Puławy, and their variation over time was described in a previous study (Kalbarczyk et al. 2018).

$SO_2$ emission was, on average, 10.9 million tons and fluctuated from 24 million tons in 1971 to 1.7 million tons in 2015. A high ammonia emission was found in the first half of the multi-year period analysed and fluctuated from 18.5 million tons in 1970 to 0.4 million tons in 1995. The highest emission of $NO_x$ was recorded in the years 1980– 1996.

In the years 1985–2015, changes in collection of underground water from company intakes ranged from approximately 114 million m$^3$ in 1985 to approximately 41 million m$^3$ in 2015. In 1999, a water intake station from the Kurówka River was built for industrial purposes, reducing the amount of underground water withdrawn by approximately 50% by substituting it with surface water. As a result of reusing cooling water in Zakłady Azotowe Puławy, water withdrawals from the Vistula River diminished by approximately 25%.

### *Collection of tree cores*

In total, 223 Scots pine stem core samples were collected from nine FRAs, from selected trees growing in the dominant and codominant canopy layers, with one sample

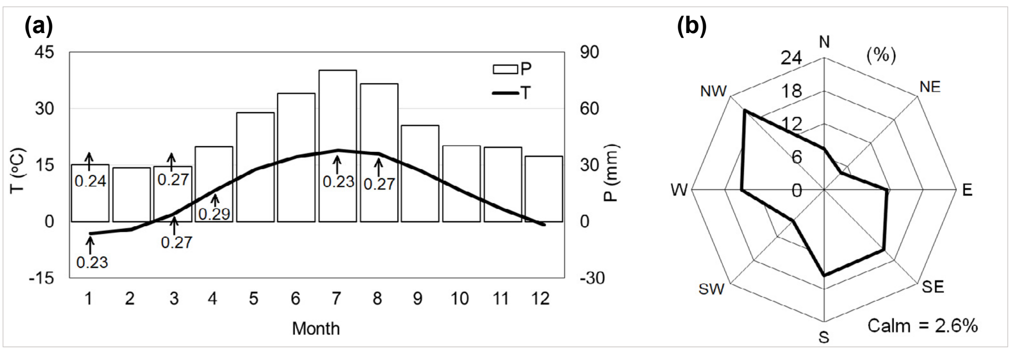


**Figure 2.** Climate conditions: average air temperature (T) and total precipitation (P) during 1930–2015 (a), and wind rose during 1951–2007 (b) in the Puławy Forest District

Note: ↑ – Pearson's correlation coefficient for a positive trend with α ≤ 0.05.

per tree drilled from west to east; 122 core samples were collected in habitat B and 101 core samples in habitat F (Table 1). Core samples were taken in July 2016 at a height of 1.3 m above ground level with the aid of a Pressler borer. Every core sample taken was given a number, and a description of the drilling location was recorded; also, the tree diameter at breast height (DBH) was measured with callipers in two directions, north-south and east-west and the average of both measurements was taken. After a period of drying at room temperature, the core samples were sanded to show the cross-section more clearly, and then a water filter (Cedro 2016) was applied to improve the visibility of the tree-ring annual increments. The tree-ring width from each year was measured using the LINTAB™ 6 measuring tool (RinnTech, Inc.) and TSAP-Win™ software package (Rinn Tech 2011) with an accuracy of up to 0.01 mm.

### *Quality assessment of individual tree-ring sequence measurements and dating of the chronologies established*

After a preliminary visual evaluation of their changes over time, annual radial growth of the collected core samples for all 223 individual sequences was assessed using the following statistical indicators: overlap ratio (*OVL*), convergence coefficient (*GLK*, %), cross-correlation (*CC*, %) and cross-date index (*CDI*) calculated based on *t* values according to Baillie-Pilcher (*TVBP*) and according to Hollstein (*TVH*). All statistical indicators were calculated using the TSAP-Win software package (Rinn Tech 2011).

Raw (TRWraw) and residual (TRWres) site chronologies were created using Program ARSTAN (Cook and Holmes 1986). Raw chronologies were created after taking the arithmetic mean of a series of tree-ring widths of the individual sequences calculated based on core samples taken from a given FRA, whereas residual chronologies were created after transformation of raw chronologies by removing the linear trend and I° autocorrelation. These transformations levelled the influence of different phases of individual tree development on the courses (or time series) of the created site chronologies. The FRA symbols are also used as codes for the site chronologies: B85, B109, B116, B142, B176c, F118, F126, F143 and F176c, where character *c* identifies the two reference chronologies, one in each habitat type. The chronologies created were evaluated using the following statistical indicators: expressed population signal (*EPS*), mean sensitivity (*MS*, %), convergence coefficient (*GLK*, %) and average correlation coefficient between the annual increment sequences (*RBAR*). Raw and residual site chronologies were described using several characteristics: arithmetic mean ($\bar{x}$, mm) and standard deviation (*SD*, mm) of the annual growth increment and maximum annual radial growth (Max, mm), as well as the autocorrelation I° coefficient (*AC1*) and Pearson's correlation coefficient (*r*) describing the age trends.

### *Variability of reductions and increases in tree-ring widths*

Changes in the pattern of reductions and increases of tree-ring widths (VTRW, %) in the Puławy Forest District were determined based on residual site chronologies for the years 1930–2015 according to the formula:

$$\mathrm{VTRW} = \frac{\mathrm{TRWn} - \mathrm{TRWc}}{\mathrm{TRWc}} \times 100\% ,$$

where TRWn is the tree-ring width for a given site chronology, and TRWc is the tree-ring width for a reference site chronology.

The choice of 1930–2015 was determined based on the age of the reference chronologies, B176c and F176c. Changes in VTRW were determined individually for habitats B and F. The values of VTRW for each year of the period of 1930–2015 were grouped into four categories. The value of VTRW was considered very small when it fell in the range of $|< 25.0\%|$, small from $|25.1\%|$ to $|50.0\%|$, medium from $|50.1\%|$ to $|75.0\%|$, and large when $|> 75.1\%|$. A negative value indicates a reduction, and a positive value indicates an increase.

### *Influence of meteorological conditions and anthropogenic factors on VTRW*

For the years common to all site chronologies (that is for 1930–2015), the statistics were calculated relating to the meteorological conditions and the reductions and/or increases in Scots pine annual radial growth as expressed by VTRW in the period from October of the preceding year through September of the year of ring formation. Analysis of Pearson's correlations for weather-VTRW relationships are summarised separately in two groups: for chronologies from habitat B (B85, B109, B116, B142) and from habitat F (F118, F126, F143). However, the relationship between VTRW and anthropogenic factors within a year was analysed for the periods 1970–2015 and 1985–2015, which was due to the availability of data provided by Zakłady Azotowe Puławy. The dependent variable in each case was the change in tree-ring width as expressed in VTRW, while the independent variables were: mean monthly air temperature, total precipitation, annual emissions of gaseous air pollution ($SO_2$, $NH_3$ and $NO_x$) and their combinations ($NH_3 + NO_x$ and $SO_2 + NH_3 + NO_x$), and finally the annual amount of surface and underground water withdrawals from company intakes used in the production of fertilisers and chemicals.

The combined influence of meteorological conditions and anthropogenic factors on VTRW was evaluated based on non-hierarchical cluster analysis, with grouping of objects (or data points) into clusters using the *k*-means method. The clusters created were evaluated using a single-factor analysis of variance for independent groups, with the share of every variable in the division of clusters established based on Fisher's test.

All input variables ($Z$) were standardized according to the formula:

$$Z = \frac{x_i - \bar{x}}{SD} ,$$

where $x_i$ is the variable value observed, $\bar{x}$ is the arithmetic mean value, and $SD$ is the standard deviation.

## Results

### Individual sequences

The overlap ratio, *OVL*, calculated for all FRAs, ranged from 74.6 to 135.8 in the case of habitat B and from 73.5 to 137.3 in the case of habitat F (Table 2), which confirms that annual tree rings were matched correctly to calendar years. The *GLK* coefficient of convergence was at least 70% for all individual sequences within a given FRA. Large values were also calculated for *CC*, which ranged from around 76.0% to 89.8%. *CDI* values ranged from 52.0 to 79.2 in habitat B and from 65.1 to 83.3 in habitat F.

### Site chronologies

Among the nine site chronologies created (TRW), the oldest were 147 years old (beginning in 1869) and 146 years old (beginning in 1870), for B142 and F143, respectively, which are located both about 4 km to the southeast of Zakłady Azotowe Puławy (Table 3). The youngest site chronologies, B176c and F176c, from stands located about 5.8 km to the southwest of the plant, were only 86 years old and began with the year 1930. The average annual increments of the raw site chronologies created (TRWraw) ranged from 1.38 mm for B142 to 2.32 mm for B176c in the case of habitat B, and from 1.44 mm for F143 to 2.44 mm for F176c in the case of habitat F.

The expressed population signal (*EPS*) used to evaluate to what extent the sample collected represents the hypothetical ("without disturbance") chronology, ranged from 0.90 to 0.97 for raw site chronologies (TRWraw) and from 0.84 to 0.95 for residual site chronologies (TRWres).

**Table 2.** Evaluation of collected Scots pine core samples used to build site chronologies in Puławy Forest District in 2016

| Indicator | Forest research areas (FRAs) | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | coniferous mixed forest (B) | | | | | broadleaf mixed forest (F) | | | |
| | B85 | B109 | B116 | B142 | B176c | F118 | F126 | F143 | F176c |
| OVL | 86.0 | 74.6 | 92.9 | 135.8 | 75.5 | 95.5 | 106.2 | 137.3 | 73.5 |
| GLK (%) | 76.0 | 71.6 | 77.9 | 75.8 | 74.2 | 73.8 | 76.0 | 70.0 | 75.6 |
| CC (%) | 81.5 | 76.0 | 78.6 | 86.5 | 77.6 | 85.0 | 86.2 | 80.0 | 89.8 |
| CDI | 60.5 | 52.0 | 69.0 | 79.2 | 67.4 | 69.1 | 83.3 | 65.1 | 62.9 |

Notes: OVL – overlap ratio, GLK – convergence coefficient (%), CC – cross correlation (%), CDI – cross-dating index calculated on the t value according to Baillie and Pilcher (TVBP) and according to Hollstein (TVH).

**Table 3.** Statistical profiles of Scots pine site chronologies (TRW) in Puławy Forest District

| Type of chronology | Chronology code | Time span (number of years | Characteristics | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | $\bar{x} \pm SD$ (mm) [1] | Max (mm) [1] | *EPS* | *MS* (%) | *GLK* (%) | *RBAR* | *AC (1)* | *r* for the linear trend |
| raw | B85 | 1916–2015 (100) | 1.75 ± 0.97 | 4.51 | 0.97 | 20.1 | 76.0 | 0.54 | 0.890 $^{3}$ | –0.66 $^{3}$ |
| res | | | 1.00 ± 0.22 | 1.90 | 0.92 | 22.5 | 77.0 | 0.34 | –0.0071 $^{ns}$ | –0.022 $^{ns}$ |
| raw | B109 | 1929–2015 (87) | 2.25 ± 1.40 | 8.10 | 0.96 | 21.7 | 71.6 | 0.54 | 0.89 $^{3}$ | –0.71 $^{3}$ |
| res | | | 1.00 ± 0.24 | 1.75 | 0.90 | 25.5 | 72.9 | 0.34 | –0.0011 $^{ns}$ | 0.031 $^{ns}$ |
| raw | B116 | 1919–2015 (97) | 2.11 ± 1.39 | 7.52 | 0.93 | 19.2 | 77.9 | 0.41 | 0.901 $^{3}$ | –0.71 $^{3}$ |
| res | | | 0.99 ± 0.18 | 1.49 | 0.91 | 22.6 | 77.8 | 0.33 | –0.031 $^{ns}$ | –0.029 $^{ns}$ |
| raw | B142 | 1869–2015 (147) | 1.38 ± 1.02 | 5.49 | 0.92 | 19.8 | 75.8 | 0.40 | 0.87 $^{3}$ | –0.80 $^{3}$ |
| res | | | 1.00 ± 0.19 | 1.71 | 0.87 | 23.8 | 75.7 | 0.32 | –0.036 $^{ns}$ | –0.0098 $^{ns}$ |
| raw | B176c | 1930–2015 (86) | 2.32 ± 0.98 | 5.38 | 0.94 | 18.2 | 74.2 | 0.40 | 0.81 $^{3}$ | –0.77 $^{3}$ |
| res | | | 1.00 ± 0.17 | 1.37 | 0.84 | 21.6 | 75.7 | 0.38 | –0.0011 $^{ns}$ | –0.046 $^{ns}$ |
| raw | F118 | 1914–2015 (102) | 2.18 ± 1.44 | 6.50 | 0.92 | 21.3 | 73.8 | 0.40 | 0.84 $^{3}$ | –0.76 $^{3}$ |
| res | | | 1.01 ± 0.25 | 1.55 | 0.89 | 22.7 | 74.5 | 0.32 | –0.031 $^{ns}$ | 0.106 $^{ns}$ |
| raw | F126 | 1902–2015 (114) | 2.09 ± 1.29 | 6.03 | 0.96 | 20.0 | 76.0 | 0.50 | 0.90 $^{3}$ | –0.62 $^{3}$ |
| res | | | 1.01 ± 0.25 | 1.57 | 0.95 | 22.7 | 76.6 | 0.44 | –0.021 $^{ns}$ | –0.098 $^{ns}$ |
| raw | F143 | 1870–2015 (146) | 1.44 ± 0.86 | 5.31 | 0.90 | 16.6 | 70.0 | 0.39 | 0.81 $^{3}$ | –0.43 $^{3}$ |
| res | | | 1.01 ± 0.26 | 1.68 | 0.86 | 19.4 | 71.7 | 0.31 | –0.0012 $^{ns}$ | –0.051 $^{ns}$ |
| raw | F176c | 1930–2015 (86) | 2.44 ± 1.47 | 6.47 | 0.96 | 19.8 | 75.6 | 0.52 | 0.84 $^{3}$ | –0.86 $^{3}$ |
| res | | | 1.00 ± 0.19 | 1.41 | 0.92 | 21.9 | 77.7 | 0.35 | –0.012 $^{ns}$ | –0.022 $^{ns}$ |

Notes: Chronology – raw (TRWraw), residual (TRWres), $\bar{x}$ – arithmetic mean (mm), *SD* – standard deviation (mm), Max – highest value in the time series (mm), *EPS* – expressed population signal, *MS* – mean sensitivity (%), *GLK* – convergence coefficient (%), *RBAR* – average correlation coefficient between the sequences of annual tree rings, *AC (1)* – autocorrelation Io, *r* – Pearson's correlation coefficient, $^{3}$ – significant at $\alpha \leq 0{,}01$, $^{ns}$ – insignificant at $\alpha \leq 0.1$, [1] – the unit applies only to raw chronology.

The relative difference in the widths of two consecutive rings, which describes the tree's sensitivity to any factors influencing their increase, was determined using the mean sensitivity indicator (*MS*), with *MS* for TRWraw ranging from 16.6% (for F143) to 21.7% (for B109), whereas for TRWres, it was between 19.4% (for F143) and 25.5% (for B109). The *GLK* coefficient for both TRWraw and TRWres was at least 70%, and the average correlation coefficient between the annual increment sequences (*RBAR*) fell within the range of 0.39–0.54 for TRWraw and 0.31–0.44 for TRWres. The autocorrelation I° coefficient for all TRWraw was significant at $\alpha \leq 0.01$ and ranged from 0.81 to 0.90, but for all TRWres, it was negative and insignificant at $\alpha \leq 0.1$. As expected, the trend in tree-ring width of Scots pine was negative with respect to increasing age for the TRWraw site chronologies, but insignificant for $\alpha \leq 0.1$ in the case of TRWres. Pearson's correlation coefficient for TRWraw ranged from −0.43 for F143 up to −0.86 for F176c.

### *Reductions and increases in tree-ring width*

The graphs (Figure 3) show that the longest reduction periods occurred in the years 1966–1996 in the case of habitat B and in the years 1961–1990 in the case of habitat F, these periods were defined to allow interruptions from one year to no more than four years, with increases not exceeding 10%. In the case of the site chronology for B85, located 2.7 km to the north of the plant, the longest period of reduction in Scots pine radial growth occurred between 1970 and 1991; the longest reduction periods for the TRW of the other habitat B sites were 1968–1995 for B109, 1966–1990 for B116 and 1966–1996 for B142.

Figure 3 shows that slightly different lengths of the periods of tree-ring width reduction, as compared to the habitat B, were recorded in the habitat F. They occurred in 1961–1987 in the case of TRW of the codes F118 and F126, and in the case of TRW of the code F143 – in 1966–1990. In habitat B, the average reduction of tree-ring width, determined for the longest periods of occurrence, amounted to 32% and ranged from 22% in TRW of B116 to 45% in TRW of B85. The reduction values of the analysed TRWs, situated at a different distance from Zakłady Azotowe Puławy, amounted to 45% (for TRW of B85, 2.7 km), 33% (for TRW of B109, 3.7 km), 28% (for TRW of B142, 4.1 km) and 22% (for TRW of B116, 12.2 km).

In habitat F, the average reduction of tree-ring width amounted to 21% and did not differ significantly among individual site chronologies, varying from 19% in TRW of F126 to 23% in TRW of F143. Other notable periods of tree-ring width reductions lasting at least four years were recorded for habitat B between 1933 and 1937 (for TRW of B85), between 1940 and 1945 (for TRW of B109) and between 1933 and 1937 (for TRW of B116), whereas for habitat F, such periods were recorded in 1940–1944 and 1999–2002 (for TRW of F118), in 1940–1943, 1994–1999 and 2001–2005 (for TRW of F126), and in 1940–1944 (for TRW F143).

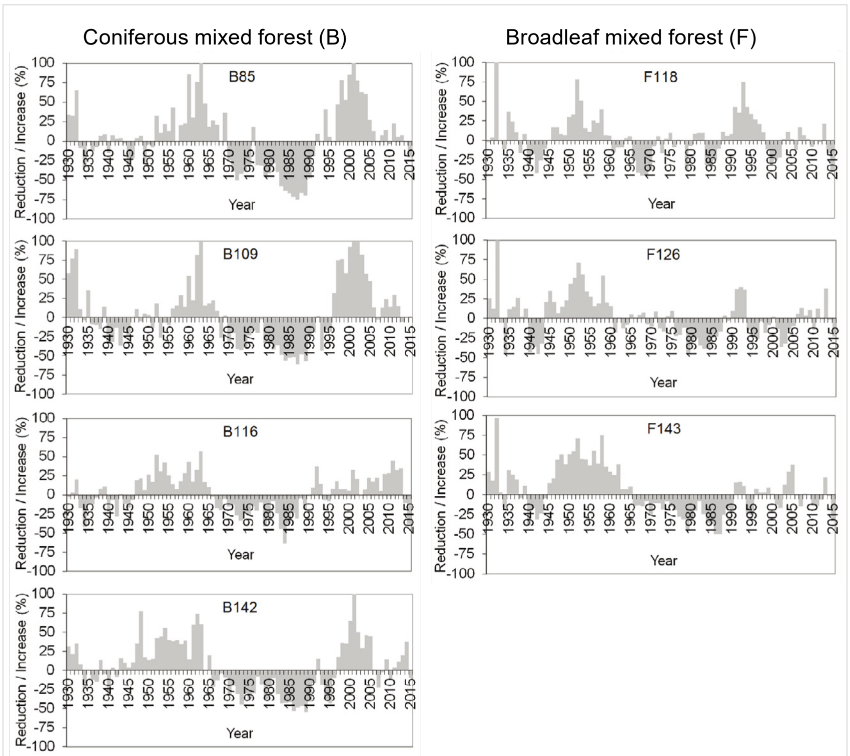


**Figure 3.** Reduction and/or increase of annual Scots pine tree-ring width in the FRAs in the Puławy Forest District in the years 1930–2015

In the years 1930–2015, there were also periods of increased annual radial growth, which were particularly notable before the construction of Zakłady Azotowe Puławy, as well as in some TRW after implementation of the environmental protection programme between 1985 and 2004 and later (Figure 3). In the coniferous mixed forest sites (habitat B), periods of increased Scots pine annual radial growth lasting at least four years were calculated for the following years: 1952–1969 and 1992–2006 (for TRW of B85); 1930–1933, 1956–1967, 1996–2006 and 2008– 2011 (for TRW of B109); 1947–1965 and 1996–2013 (for TRW of B116); and finally 1930–1933, 1943–1965, 1997–2006 and 2011–2014 (for TRW of B142). In the broadleaf mixed forest sites (habitat F), such periods of increased tree-ring widths were recorded for the following years: 1934–1937, 1945–1960 and 1988–1998 (for TRW of F118); 1935– 1939, 1944–1960, 1965–1968, 1990–1993 and 2006–2009 (for TRW of F126); and 1930–1933, 1945– 1965 and 1996–2000 (for TRW of F143).

In the period 1930–2015, among the seven site chronologies analysed (TRW), there were more years with reduced tree-ring widths than increased widths only in the TRW of B109, while the TRW of F143 had a balance between reduction and increase, and increases dominated in most years for five TRWs: B85, B116, B142, F118 and F126 (Figure 4). In the period analysed, no large (< −75%) reduction in annual increments was found in any TRW, while medium-sized reductions were found in four TRW, and small or very small reductions were found in all TRW examined. Medium-sized reductions of secondary growth were recorded in the TRW of B85 (9%), small reductions in the TRW of B109 (20%) and TRW B142 (20%), and very small reductions in the TRW of B116 (34%) and TRW F143 (34%). By contrast, increases in tree-ring width in all four size categories were found, with increases in the very small size category being most frequent: from 24% for TRW of B142 to 41% for TRW of F118. On average, there were half as many small increases compared with very small increases, ranging from 6% for TRW of B109 to 19% for TRW of B142, with medium and large increases respectively amounting to 5% and 4%, more often in habitat B.

Reductions in the tree-ring widths for habitat B occurred simultaneously across the four TRW in 24 years of the time series: 1930, 1936, 1937, 1940, 1970–1975 and 1977–1990; and in habitat F, reductions occurred simultaneously across the three TRW in 22 years: 1934, 1940– 1943, 1969–1970, 1973, 1976, 1978–1980, 1984– 1987, 2001– 2002, 2010, 2012 and 2014–2015 (Figure 5). These reductions of tree-ring width were simultaneous across both habitats in only 10 of the years: 1940, 1970, 1973, 1978–1980, and 1984–1987.

An increase of tree-ring width in habitat B occurred simultaneously across the four TRW in 26 years of the time series: 1931–1932, 1947, 1952, 1956, 1958–1963, 1965, 1992, 1997–2006, 2009 and 2011–2012. In habitat F, increases occurred simultaneously across the three TRW in 28 years: 1931–1932, 1935–1937, 1939, 1945–1960, 1965, 1991–1993, 2008–2009 and 2013. Simultaneous increases of tree-ring width across both habitats analysed were found in 10 of the years: 1931–1932, 1947, 1952, 1956, 1958–1960, 1965, 1992 and 2009.

### *Climate-VTRW relationship*

The strength and, sometimes, also the direction of influence of meteorological conditions on VTRW differed between the habitats explored (Figure 6). A different direction of air temperature impact on the magnitude of VTRW was found for two months out of the twelve explored. For habitat B, in October air temperature of the preceding year positively influenced VTRW, while December air temperature negatively influenced VTRW; in habitat F was found

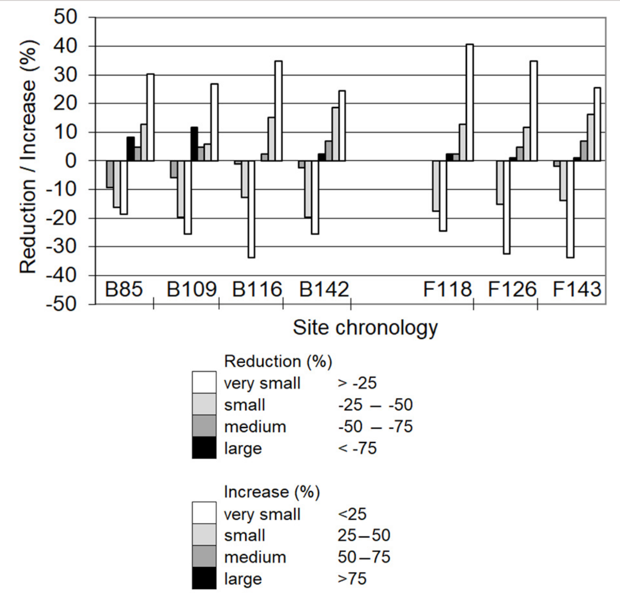


**Figure 4.** Frequency of occurrence of reduction and increase of Scots pine tree-ring width in the FRAs in the Puławy Forest District in the years 1930–2015

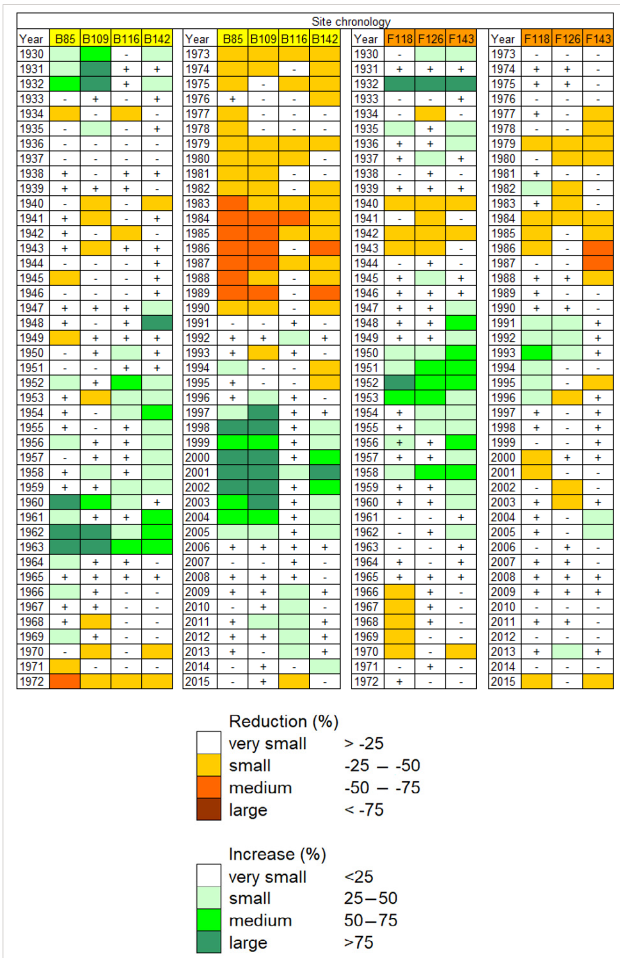


**Figure 5.** Identification of the degree of reduction and increase of Scots pine tree-ring width in the FRAs in the Puławy Forest District in the years 1930–2015

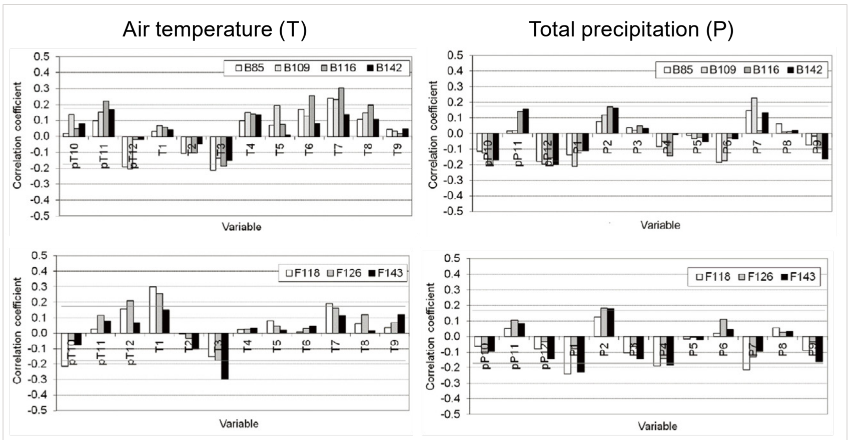


**Figure 6.** Relationship between reduction and increase of annual Scots pine tree-ring widths in the FRAs in the Puławy Forest District and meteorological conditions in the years 1930–2015

Notes: p – previous year, Pearson's correlation coefficient at $\alpha \leq 0.1$ and for $N = 86$ amounts to $\pm$ 0.18, where $N$ is the number of degrees of freedom (–), T1–T12 – air temperature in consecutive months of the year, P1–P12 – precipitation in consecutive months of the year.

the opposite relationship. In November of the preceding year, as well as the period from April to September, air temperature positively influenced VTRW in both habitats – that is, above average air temperature (T) favoured an increase in tree-ring width. In habitat B, July temperatures had the strongest and most positive influence on VTRW, whereas the strongest negative influence was observed in December of the preceding year and in March of the current year; however, in habitat F, the strongest positive influence was found in January, and the strongest negative influence was found in March.

The influence of total monthly precipitation (P) on VTRW differed by habitat for three months, March, June and July: in habitat B, the relationship was negative in June and positive in March and July, whereas it was the opposite in habitat F (Figure 6). Pluvial conditions, except for June and July, positively influenced VTRW in November of the preceding year and in February and August of the current year, and negatively in October and December of the preceding year, in January, during April–May and in September of the current year.

### *Relationship between VTRW and anthropogenic factors*

The influence of air pollutant emissions produced during the manufacturing of mineral fertilisers and chemicals in the years 1970–2015 on the level of reduction or increase of Scots pine tree-ring widths as quantified by VTRW was significant and negative in five out of the seven considered site chronologies (Figure 7). Slightly higher Pearson's correlation coefficients were found between VTRW and $NH_3 + NO_x$, and between VTRW and $SO_2 + NH_3 + NO_x$ than between VTRW and individually analysed instances of air pollution. Significant correlation coefficients ($r$) for the relationship between VTRW and $NH_3 + NO_x$ ranged from −0.58 for TRW of F143 to −0.78 for TRW of B85 and B109, and for the relationship between VTRW and $SO_2 + NH_3 + NO_x$, from −0.50 for TRW of F143 to −0.73 for TRW of B142. From the correlation analysis, among the separately analysed instances of air pollution, the strongest influence on VTRW was exerted by $NH_3$ ($r$ ranged from −0.45 for TRW of F143 up to −0.68 for TRW of B109), followed by $NO_x$ ($r$ ranged from −0.36 for TRW of B116 up to −0.60 for TRW of B85). $SO_2$ was most strongly and negatively associated with VTRW in the case of TRW of B142. The weakest negative influence of air pollution was found for two site chronologies created in habitat F, which is TRW of F118 and TRW of F126, which are located about 12 km to the east of Zakłady Azotowe Puławy.

Correlation coefficients confirmed significant VTRW dependencies on the amount of surface and underground water withdrawals by Zakłady Azotowe Puławy within the

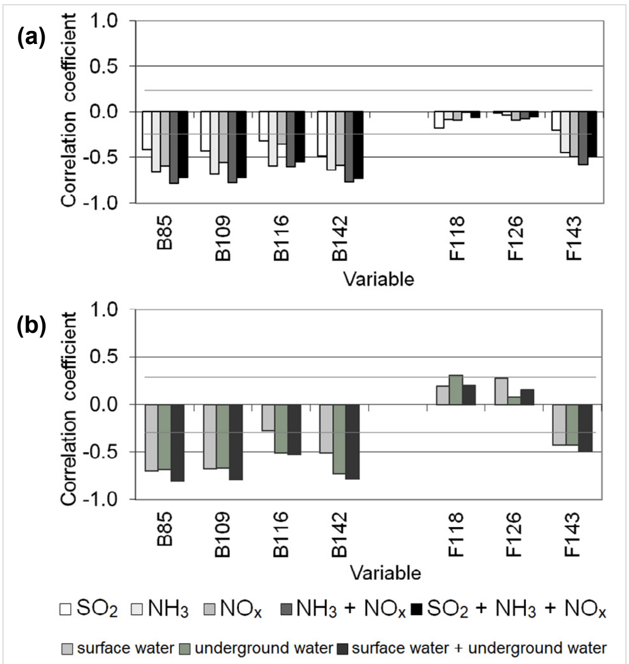


**Figure 7.** Relationship between reduction and increase of annual Scots pine tree-ring widths in the FRAs in the Puławy Forest District and gaseous pollution emissions from Zakłady Azotowe Puławy in the years 1970–2015 (a) and the amount of surface and underground water withdrawn from company intakes and used in the production of fertilisers and chemicals in the years 1985–2015 (b)

Note: Pearson`s correlation coefficient at $\alpha \leq 0.1$ and for $N = 45$ amounts to $\pm$ 0.24, and for N = 30 – $\pm$ 0.29, where $N$ means is the number of degrees of freedom (–).

period 1985–2015 (Figure 7), with a larger water withdrawal favouring a reduction in tree rings of five chronologies created; the exceptions were F118 and F126, which are located in habitat F. Surface water withdrawal most strongly and negatively determined TRW of B85 ($r = -0.70$), followed by TRW of B109 ($r = -0.68$), which were located closest to the plants, with the least effect on TRW of B116 ($r = -0.27$), located 12.2 km from the plants. The relationship between VTRW and underground water withdrawal was similar to the relationship between VTRW and surface water withdrawal, with regards to direction and strength. After considering in the assessment the influence of total surface and underground water withdrawals on VTRW, the closest relationships had correlation coefficients ranging from −0.50 for TRW of F143 up to −0.80 for TRW of B85.

### *Joint influence of climate and anthropological factors on VTRW*

The first cluster analysis model shows that changes in the size of Scots pine tree rings in the period 1930–2015 in FRAs of both habitats analysed in Puławy Forest District significantly depended on the meteorological variables group (Table 4). Air temperature had a significant influence on VTRW in the period between June and August (T6–8; $F \approx 53$, $\alpha \leq 0.01$), as did total precipitation in the period between December and January (pP12–P1; $F \approx 9$, $\alpha \leq 0.01$).

Considering additional variables that characterise changes in air pollution emissions from Zakłady Azotowe Puławy during 1970–2015, the second model indicates that $NH_3$ emissions ($F \approx 78$, $\alpha \leq 0.01$) and air temperatures ($F \approx 31$, $\alpha \leq 0.01$) in the period between June and August had the strongest influence on the changes of tree-ring widths (Table 5). VTRW was also influenced significantly (at the level of $\alpha \leq 0.01$) by other factors ($SO_2$, $NO_x$, and pP12–P1), for which the values of Fisher's test were determined to be, respectively, 13.4, 7.1 and 5.2.

**Table 4.** Influence of a selected group of meteorological variables on the reduction and/or increase of annual Scots pine tree-ring widths in the FRAs in Puławy Forest District in the years 1930–2015

| Variable | *SS* | $Df_{SS}$ | *SSE* | $Df_{SSE}$ | Fisher's test | α |
|---|---|---|---|---|---|---|
| Bx | 41.6 | 2 | 43.4 | 83 | 39.8 | 0.01 |
| Fx | 43.8 | 2 | 41.2 | 83 | 44.2 | 0.01 |
| T6–8 | 47.6 | 2 | 37.4 | 83 | 52.9 | 0.01 |
| pP12–P1 | 5.1 | 2 | 79.9 | 83 | 8.6 | 0.01 |

Notes: Bx – changes in the size of tree rings expressed by VTRW for habitat B (chronology average: B85, B109, B116, B142), Fx – changes in the size of tree rings expressed by VTRW for habitat F (chronology average: F118, F126, F143), T6–8 – air temperature from June to August, pP12–P1 – the total precipitation from December to January, *SS* – sum square error of between-group variation, $Df_{SS}$ – number of degrees of freedom for sum square error *SS*, *SSE* – sum square error of within-group variation, $Df_{SSE}$ –number of degrees of freedom for sum square error *SSE*, α – level of probability.

In the third cluster analysis model, based on the years 1985–2015, in which one additional variable was taken into account (i.e. the amount of surface and underground water withdrawn from company intakes, SUW), tree-ring widths of these Scots pine forest stands were mostly determined by anthropogenic factors: $NO_x$ ($F \approx 151$, $\alpha \leq 0.01$), SUW ($F \approx 146$, $\alpha \leq 0.01$), $NH_3$ ($F \approx 38$, $\alpha \leq 0.01$) and $SO_2$ ($F \approx 18$, $\alpha \leq 0.01$) (Table 6). Meteorological variables had less of an impact on VTRW than anthropogenic factors. The Fisher's test values for meteorological variables ranged from 4 in the case of pP12–P1 up to 7 in the case of T6–8. In the second and third cluster analysis models constructed, a stronger influence of meteorological variables and anthropogenic factors on VTRW was found for habitat B (Tables 5–6).

Below-average air temperature in the period from June to August and above-average total precipitation in the period from December to January promoted a reduction in Scots pine tree-ring width in the FRAs of both analysed habitats in Puławy Forest District (Figure 8a). The inclusion of anthropogenic factors in the cluster analysis models, which characterise the unfavourable influence of Zakłady Azotowe Puławy on the environment, demonstrates a weakened influence of meteorological conditions

**Table 5.** Influence of a selected group of meteorological variables and gaseous pollution from Zakłady Azotowe Puławy on the reduction and/or increase of annual Scots pine tree-ring widths in the FRAs in Puławy Forest District in the years 1970–2015

| Variable | *SS* | $Df_{SS}$ | *SSE* | $Df_{SSE}$ | Fisher's test | α |
|---|---|---|---|---|---|---|
| Bx | 43.2 | 2 | 12.4 | 43 | 74.8 | 0.01 |
| Fx | 4.2 | 2 | 18.9 | 43 | 6.5 | 0.01 |
| T6–8 | 35.2 | 2 | 24.2 | 43 | 31.4 | 0.01 |
| pP12–P1 | 9.2 | 2 | 38.2 | 43 | 5.2 | 0.01 |
| $SO_2$ | 17.3 | 2 | 27.7 | 43 | 13.4 | 0.01 |
| $NO_x$ | 11.1 | 2 | 33.9 | 43 | 7.1 | 0.01 |
| $NH_3$ | 35.3 | 2 | 9.7 | 43 | 78.1 | 0.01 |

Notes: $SO_2$ – sulphur dioxide emission, $NO_X$ – nitrogen oxides emission, $NH_3$ – ammonia emission. The remaining explanations as in Table 4.

**Table 6.** Influence of a selected group of meteorological variables and anthropogenic factors on the reduction and/or increase of annual Scots pine tree-ring widths in the FRAs in Puławy Forest District in the years 1985–2015

| Variable | *SS* | $Df_{SS}$ | *SSE* | $Df_{SSE}$ | Fisher's test | α |
|---|---|---|---|---|---|---|
| Bx | 23.1 | 2 | 15.7 | 28 | 20.6 | 0.01 |
| Fx | 5.3 | 2 | 9.3 | 28 | 7.9 | 0.01 |
| T6–8 | 10.5 | 2 | 21.8 | 28 | 6.7 | 0.01 |
| pP12–P1 | 6.8 | 2 | 22.3 | 28 | 4.3 | 0.01 |
| $SO_2$ | 12.4 | 2 | 9.4 | 28 | 18.3 | 0.01 |
| $NO_x$ | 37.2 | 2 | 3.4 | 28 | 151.4 | 0.01 |
| $NH_3$ | 10.5 | 2 | 3.8 | 28 | 38.0 | 0.01 |
| SUW | 27.3 | 2 | 2.6 | 28 | 146.3 | 0.01 |

Notes: SUW – amount of surface and underground water withdrawn from company intakes and used in the production of fertilisers and chemicals. The remaining explanations as in Table 4.

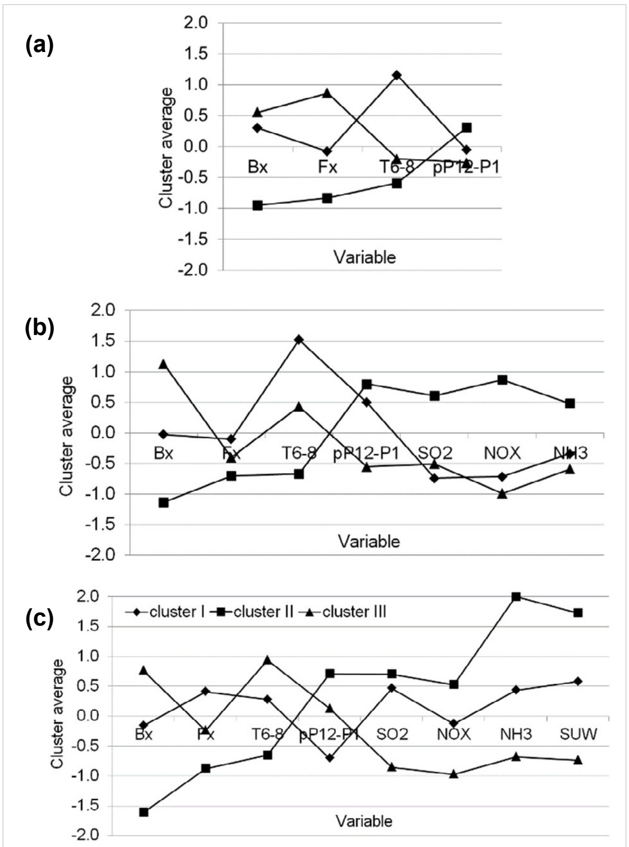


**Figure 8.** Cluster average for chosen variables (Tables 4–6) describing aggregate impact of meteorological variables and anthropogenic factors on reduction and/or increase of annual Scots pine tree-ring widths in the FRAs in the Puławy Forest District in the years: 1930–2015 (a), 1970–2015 (b) and 1985–2015 (c)

Note: Explanations as in Tables 4–6.

on VTRW (Tabels 4–6, Figure 8bc). As expected, reductions in tree-ring widths were demonstrated to occur with above-average emission of $SO_2$, $NO_x$ and $NH_3$, as well as with above-average withdrawals of surface and underground water from company intakes. Under conditions of strong human impact on the environment, below-average temperatures in the period from June to August (T6–8) and above-average precipitation conditions in the period from December to January (pP12–P1) favoured the formation of narrower tree rings.

## Discussion

The identified reduction periods in pine stands near Puławy correspond with the period of launching and operating of Zakłady Azotowe Puławy. Gaseous air pollution, such as from sulphur dioxide, ammonia and nitric oxides, disturbs the physiological processes of trees, thereby slowing down or totally inhibiting their primary and secondary growth (Liu et al. 2011, Sensuła et al. 2015, Cedro and Cedro 2018). Indeed, once toxic substances are incorporated into woody plants, physiological effects may be observed on various levels of biological organisation, including cells, entire plants and even the plant community (Jim and Chen 2008, Sha et al. 2010, Safdari et al. 2012).

The end of the TRW reduction period in Puławy Forest District was determined to be in the 1990s, which was when the provincial authorities implemented an environmental protection programme in the years 1985–2004, which was possible in the later years thanks to the introduction of new legal regulations, both from the state and the EU. At the same time, after system changes in Poland after 1989 and the start of economic transformation, there was a decrease in the production of mineral fertilisers and chemicals, and subsequently, after the introduction of new production technologies and gas reduction systems, the amount of industrial pollution decreased. There was large variability of gaseous pollution emissions from Zakłady Azotowe Puławy during 1970–2015 (Kalbarczyk et al. 2018).

According to Rutkiewicz and Malik (2018), the effects of physiological disorders are recorded in the form of reduced tree-ring width, with the aid of which one may determine both the beginning and end of such influences. In such a period, it is possible to evaluate the direction of industry influence on the natural environment, as well as the strength of such influence, as this study has done. Due to the periodic activity of cambium in trees, the influence of exogenous factors is still recorded in their secondary growth, or radial increments of secondary xylem, with annual precision, and this manifests as a decrease in tree-ring width (Luong et al. 2013, Sensuła et al. 2017). Such reductions in secondary growth have been determined by various methods, including visual evaluation of chronology curves, regression models and comparison of stand chronologies with a reference chronology (Malik et al. 2012, Ziemiańska et al. 2019), as done in this study.

Numerous dendrochronological experiments for evaluating the changes of air quality and other environmental elements have demonstrated the negative influence of emissions on tree stands (Danek 2008, Sensuła et al. 2017, Putalová et al. 2019). Such studies have shown that as distance from emission sources increases, the negative reactions of trees get progressively smaller, and as emissions decrease, tree-ring widths reach a value which is like the width from the preceding period, or even exceed it (Wilczyński 2006, Danek 2008). The impacts of pollution and the scale of the environmental changes caused by the influence of humans on the growth and development of trees are determined by the concentration of phytotoxic substances and physiological and biochemical features of the plants themselves, as well as the influence of biotic and abiotic factors that modify the state of the environment (Fritts et al. 1991, Sensuła et al. 2015, Fu et al. 2020). Abiotic factors include, among others, climatic fluctuations, clearly observed in the Little Ice Age. In Poland in the XX century there were only brief, several-year-long periods characterised by very unfavourable atmospheric conditions during the formation of tree-ring widths (Kaczka 2004).

TRW reduction was also influenced by surface and underground water withdrawals by company intakes, which lowered the level of groundwater in the FRAs in Puławy Forest District. Reduction of Scots pine tree-ring width was also favoured by below-average air temperature between June and August and by above-average precipitation between December and January. However, the influence of temperature and precipitation conditions on TRW was significantly smaller in Puławy Forest District than in non-industrial areas. Still, meteorological conditions had a crucial influence on the interaction of anthropogenic factors with VTRW as they determined the movement and settlement processes of industrial pollution resulting from dry and wet deposition. The significant influence of meteorological conditions on TRW was reported by Dueck et al. (1998), Hordo et al. (2009), Augustaitis et al. (2014) and Rimkus et al. (2019). Short, mild winters as well as warm and quickly starting springs favour wide tree rings in the season of growth and development of Scots pine (Koprowski et al. 2012, Cedro and Cedro 2018, Rimkus et al. 2019). At the end of winter, as day length increases, pine stands lose their frost resistance, becoming sensitive to low air temperatures. Needles, branches and trunks freeze and experience frost damage, drying out by cold winds and mechanical damage from snow, which all impair tree health and reduce tree-ring growth rates (Cedro and Cedro 2018). Furthermore, recent studies show that observed climate changes expressed in terms of air temperature increases limit tree water access in the summer, resulting in the formation of narrower tree rings (Babst et al. 2019, Fu et al. 2020).

## Conclusions

Long-term periods of reduction of Scots pine tree-ring width in Puławy Forest District were observed in all FRAs studied during the years starting from the launch of Zakłady Azotowe Puławy until introduction of new technologies and environmental protection laws. A clear decrease of tree-ring width with respect to reference sites was determined for the period from the end of the 1960s to the beginning of the 1990s. The reduction in ring width lasted for longer and was more intensive in the coniferous mixed forest habitat (habitat B), which was completely dominated by Scots pine. With the decrease of industrial emissions in the Puławy Forest District, and the concurrent decrease in the amount of surface and underground water withdrawn from company intakes, there was not only a reversal of the observed trend of diminishing tree-ring width, but tree rings even began to exceed the widths observed before the period of anthropogenic influence.

The reduction of Scots pine tree-ring width was significantly dependent on meteorological conditions – below-average air temperatures in the period from December of the year preceding ring formation to January of the current year, and above-average total precipitation in the period from June until August – as well as anthropogenic factors – above-average $SO_2$, $NH_3$ and $NO_x$ emissions and above-average surface and underground water withdrawals by Zakłady Azotowe Puławy. The influence of weather on VTRW variability was weakened by anthropogenic factors, which determined the quality of the local natural environment.

Scots pine tree-ring widths may thus be used to evaluate the state of and threats to the natural environment in areas with high levels of industrial pollution, for example, $SO_2$, $NH_3$ and $NO_x$.

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