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The Impact of Air Pollution on the Growth of Scots Pine Stands in Poland on the Basis of Dendrochronological Analyses †

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Abstract: The aim of this study was to evaluate Scots pine stand degradation caused by the pollutants emitted from Zakłądy Azotowe Puławy, one of the biggest polluters of the environment in Poland for over 25 years (1966–1990). To assess the pollution stress in trees, we chose the dendrochronological analysis We outlined three directions for our research: (i) the spatio-temporal distribution of the growth response of trees to the stress associated with air pollution; (ii) the direct and indirect effects of air pollution which may have influenced the growth response of trees; and (iii) the role of local factors, both environmental and technological, in shaping the growth response of trees. Eight Scots pine stands were selected for study, seven plots located in different damage zones and a reference plot in an undamaged stand. We found that pollutant emission caused disturbances of incremental dynamics and long-term strong reduction of growth. A significant decrease in growth was observed for the majority of investigated trees (75%) from 1966 (start of factory) to the end of the 1990s. The zone of destruction extended primarily in easterly and southern directions, from the pollution source, associated with the prevailing winds of the region. At the end of the 1990s, the decreasing trend stopped and the wider tree-rings could be observed. This situation was related to a radical reduction in ammonia emissions and an improvement in environmental conditions. However, the growth of damaged trees due to the weakened health condition is lower than the growth of Scots pine on the reference plot and trees are more sensitive to stressful climatic conditions, especially to drought.

Keywords: Scots pine; tree-ring; air pollution; growth reduction; climate change; Poland

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1. Introduction

Air pollution and climate change are regarded as key stressors that entail a global threat to forest health and sustainability [1–3]. Interactions between these factors—synergistic on the one hand and antagonistic on the other—that result in direct and indirect changes to forest ecosystems have been described in many studies over the last few decades [4–11]. The synergistic, mutually reinforcing interaction of these factors results in a cumulative negative impact on the metabolism and physiological processes of trees [12]. High concentrations of air pollutants (mainly SO_2 and NO_2) can damage trees directly through the foliage and indirectly through the soil [1,3]. Typical symptoms include disturbances in photosynthesis and stomatal conductance, shifts in carbon dioxide allocation and water use efficiency, and leaf loss. The mechanism and effects of the impact of toxic chemicals on the course of physiological processes in plants have been described in numerous studies [12–18]. Additionally, the deposition of pollutants influences the way in which trees respond to other abiotic and biotic stressors, an example being increased sensitivity to drought and pathogen attacks [19].

Global warming is causing an increase in extreme weather events, and in particular in severe droughts—further compounded by unusually high temperatures [20]. Increasingly, trees are exposed to water stress which can cause physiological damage [21]. Trees weakened by the deposition of contaminants and, at the same time, by unfavourable weather conditions become predisposed to secondary stress (from insects, diseases, or fires). Interactions between these factors cause a gradual reduction in tree vigour and growth, which—in extreme cases—may lead to dieback of trees or certain species of trees and to changes in the ecosystem [22]. According to Manion's concept [22], air pollution can be a factor that both predisposes forests to dieback and initiates this process. A schematic view of the integrated impact of air pollution, climatic conditions, and pathogens is presented in Figure 1.

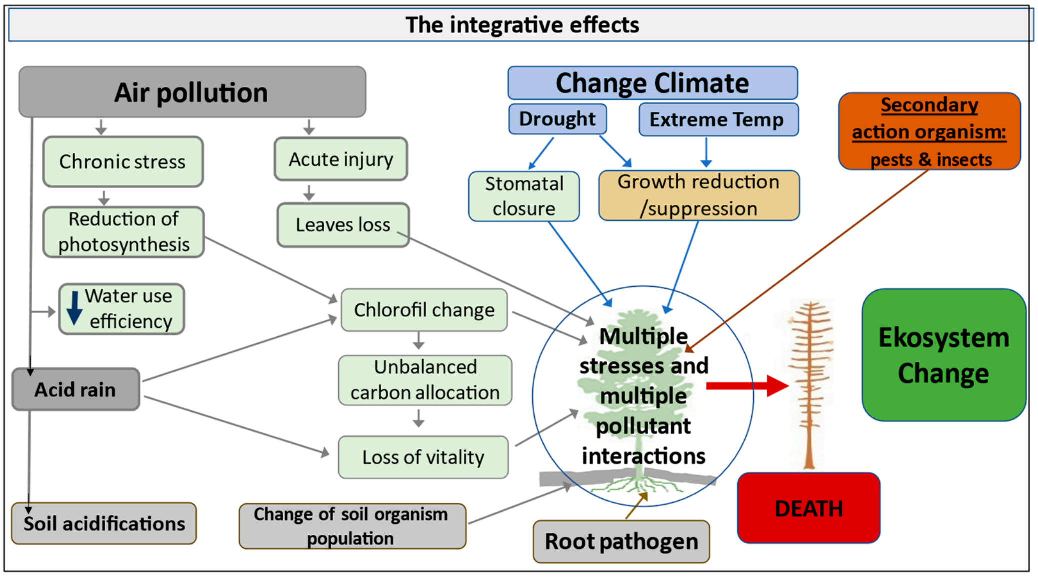

Figure 1. A schematic, integrated influence of air pollution, climatic conditions, and pathogens on trees. Explanation of the colours used in the figure: green—changes taking place in plants; grey—changes in the soil environment; brown—a growth reaction recorded in tree rings.

Despite the significant changes that have occurred over the recent decades in the chemistry, concentration, and geographical distribution of air pollutants (reduction in SOX emissions by 80% and NOX emissions by 30%), the influence of these pollutants on the structure and functioning of forest ecosystems is still visible [3,6,16,18,19,23,24]. A significant proportion of forest ecosystems remain at risk due to the excessive deposition of nitrogen compounds (ammonia NH3) and nitrogen oxides (NOX) [3,17,23–25].

To assess the pollution stress in trees, many biochemical, physiological, and morphological methods are used [14–19,26]. Among the morphological methods, the most common approach is tree-ring analysis [27–35]. A retrospective analysis of annual ring widths and other tree-ring parameters (e.g., chemical composition of the wood, stable isotopes) makes it possible to obtain information on the state of the environment at a high time resolution. Hence, tree rings are often used to assess the long-term effects of air pollution in the environment [29–38]. These studies have indicated that trees growing in a polluted environment have narrower increments or, in extreme cases, do not produce any increments in a given year [36–38].

It is commonly believed that on a global scale, the second half of the 20th century was the period with the greatest pressure of industrial pollution on forest ecosystems e.g., [1–3,23]. Since the 1960s, Poland has been classified as one of the countries with the worst air pollution indicators in Central Europe. For almost forty years, significant amounts of toxic pollutants have been emitted into the atmosphere, causing damage to forests and, in extreme cases, resulting in the dieback of entire stands [39]. The degree of forest stand degradation has varied spatially depending on the amount, type, and concentration of the pollutants as well as the duration of emission, distance from the emitter, and local orographic and climatic conditions. Most of the forest areas exposed to long-term stress related to environmental pollution suffered chronic damage and progressive deterioration of tree health, sometimes resulting in death [34–40]. The most severe damage affected forests growing in areas having a high concentration of industrial plants and subjected to the long-term negative influence of various toxic substances. This problem has been widely described in the literature [30,35–41].

Against this background, the rapid degradation of forest ecosystems caused by pollutant emissions from one plant—the nitrogen fertiliser factory in Puławy (Zakłady Azotowe Puławy)—was unique both in Poland and in Central Europe. The sudden interference with the environment in the form of high concentrations of toxic substances emitted by the plant resulted in rapid degradation of many hectares of forest within a period of several years and in the creation of a "biological death zone." Initially very abrupt, the changes in the forest environment later became chronic. A more detailed description of the damage is provided in the further sections of this paper. The degradation of the environment caused by the operation of the nitrogen fertiliser factory in Puławy has been described in many publications, e.g., [42–49]. However, the ecological interaction between air pollution and the resistance of forests to the abiotic stress that comes with climate change, especially drought, has not been examined. The role of other environmental and technological factors that could result in such extensive degradation of the forest has not been analysed to date, either.

Hence, in this study, we decided to expand the existing knowledge on the impact of pollutants emitted by the fertiliser plant in Puławy on the degradation of the pine stands growing in the surrounding area. We outlined three directions for our research: (i) the spatio-temporal distribution of the growth response of trees to the stress associated with air pollution; (ii) the direct and indirect effects of air pollution which may have influenced the growth response of trees; and (iii) the role of local factors, both environmental and technological, in shaping the growth response of trees. We set ourselves the goal of testing the following hypotheses: (i) that the emission of pollutants had caused a long-term reduction in the annual growth of pine, with the distribution and intensity of this reduction varying spatially and temporally; (ii) that the extent and spatial coverage of the reduction in the growth of pine stands can be attributed to the amount and type of pollutants, but also to local factors, especially anemometric and habitat conditions; and (iii) that the negative impact of pollutants on the growth response persists for a very long time and that even after a radical reduction in emissions, the trees continue to show reduced resistance to abiotic stress related to climate change (especially drought). To test the above hypotheses, we chose the dendrochronological analysis methods used in previous spatial-temporal studies on changes in the growth responses of trees growing in other industrial areas of Poland see [34–41]. The following reasons determined the choice of pine as the species to be studied: (i) pine is the dominant species in Poland and in the study area (accounting for 58% and 71% of forests in Poland and in the Puławy Forest District, respectively); (ii) pine as a species is very sensitive to air pollution (exposure of needles to pollution may lead to the decline of trees, e.g., [14,15,29–32,35,36]); and (iii) the spatial and temporal growth responses of pine to air pollution are being analysed for other regions of Poland. The last premise and the choice of the dendrochronological method also make it possible to use the results of this study to obtain a geographical and historical view of the impact of industrial pollution on the pine stands growing in Poland.

2. Study Area, Materials and Methods

2.1. Study Area

2.1.1. The Nitrogen Fertiliser Factory in Puławy

The nitrogen fertiliser factory in Puławy (Zakłady Azotowe Puławy) is located in central-eastern Poland (51°27′ N & 21°58′ E), in a region with prevailing westerly winds (Figure 2). The area had no significant air pollution prior to the launch of the factory. The industrial plant was built on the western edge of a large forest complex, in an area covered by oligotrophic habitats types, dominated by Scots pine, formed on sandy soils low in nutrients and suffering from periodical acute water deficits [45,50]. The facility was commissioned in the autumn of 1966, and the first signs of damage to pine stands were already observed in the early spring of 1967. In the same year, many trees died within an area of 70 hectares [41]. The zone of forest destruction kept rapidly increasing in size: within the next three years, an area of 500 hectares of the forest on the eastern side of the plant was completely degraded (with all trees being dead), creating a "biological death zone". The extent of damage to tree stands in the remaining areas was varied, with more than 75% of dry or severely damaged trees in the most affected zone. From 1970 onwards, as the devastation of the environment progressed, the zones of destruction continued to expand outwards from the fertiliser plant. Currently, severely damaged forests (with more than 75% of trees affected) cover an area of 1200 hectares, and forests with moderate damage (31% to 75% of trees) and minor damage (5% to 30% of trees) cover approximately 500 hectares and 7000 hectares, respectively [50]. The amount of air pollutants emitted from the factory in the different years is shown in Figure 2 (bottom).

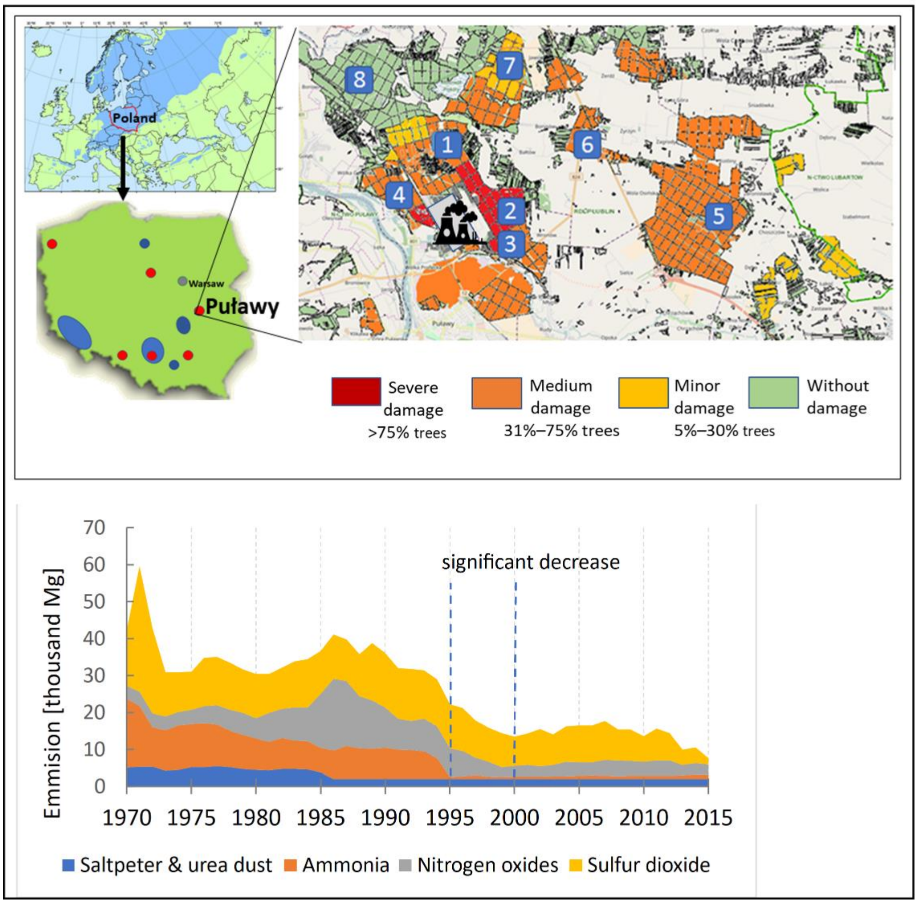

Figure 2. (**Top**): Location of the study plots. Also marked on the map of Poland are the locations of Polish industrial plants which emit toxic pollutants that cause damage to forests in Poland; Explanation: red circles—nitrogen plants, blue sign- other plants or the industrial region. Information on the impact of pollution on forests in the regions shown on the map can be found in the following studies: 30; 35–38; 40–49. (**Bottom**) Emission of gaseous pollutants (SO_2, NH_3, NO_x) from the emitters at Zakłady Azotowe Puławy in 1970–2015.

However, in addition to emissions resulting from the normal manufacturing process, there were also uncontrolled emissions associated with equipment failures as a result of which toxic substances were released into the atmosphere at concentrations well in excess of the annual average values. In the early years of the plant's operation, the deposition of pollutants reached 1000–1200 kg ha^{-1} yr^{-1} (42,46,48,51). Beginning in the early 1990s, pollutant emissions began to decrease gradually as a result of modernisation, with a radical decrease—especially in the amount of ammonia—taking place in 1995. In 2014, there was a further reduction in pollutant emissions. The current level of emissions is 15% of the value recorded at the beginning of the plant's operation [51].

2.1.2. Climatic Conditions

The meteorological data used in this study—originating from the Puławy Meteorological Station—was acquired courtesy of the Polish Institute of Meteorology and Water Management (IMGW-PIB). The monthly average air temperatures, monthly precipitation totals and monthly average wind speed and direction values for 1951–2015 were used to determine the climate characteristics.

The 1951–2015 period was characterised by an annual average temperature of approximately 8.2 °C (with data points ranging from 6.1 to 10.2 °C) and a mean annual precipitation total of around 572 mm (with data points ranging from 403 to 797 mm per year). The growing season (GS) began in April and lasted until the end of October. The average GS air temperature was 14.0 °C (with data points ranging from 12.4 to 15.3 °C), and the mean precipitation total in this period was 368 mm (with data points ranging from 207 to 570 mm). The lowest level of precipitation was observed between the mid-1980s and mid-1990s and in the first decade of the 21st century. The wind conditions in the region were characterised by a relatively low average wind speed (2.1 m/s) and a high frequency of calm periods, especially between June and September. The dominant wind directions were north-west and south (Figure 3).

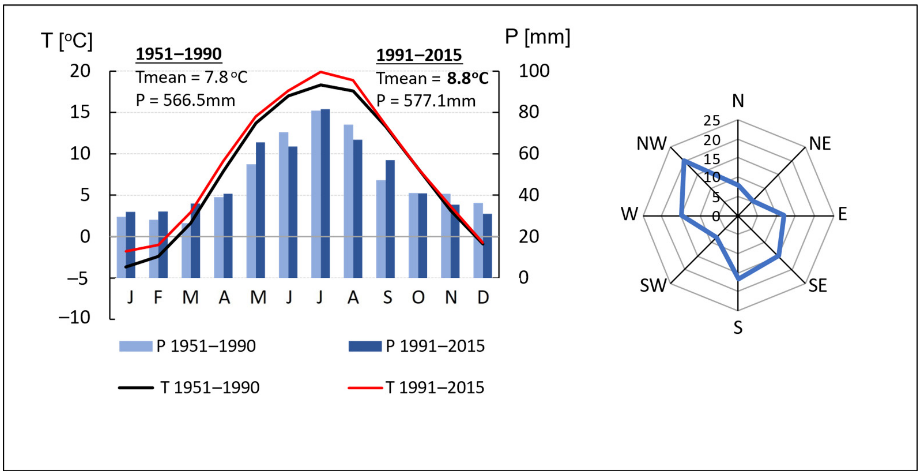

Figure 3. Climate diagram and wind rose for study area, constructed on the basis of data from meteorological station in Puławy for the years 1951–2015.

Within the research area, climate change mainly manifested itself in an increase in air temperature (Figure 3). In the years 1951–2015, a statistically significant increase in the annual average air temperature (0.26 °C per 10 years) and the average temperatures in the winter months (December to February), in spring (March) and in summer (June to August) was recorded in the study area. The greatest increase in air temperature was recorded for March, February and January (0.512, 0.49 and 0.33 °C over 10 years, respectively). In the case of precipitation, the changes were not statistically significant; there was a slight upward trend in the total annual rainfall with a negative trend in summer rainfall, especially in June and August.

2.2. Research Materials and Methods

2.2.1. Study Plots and Sample Acquisition

Eight Scots pine stands were selected for the study: seven plots located in different damage zones and a reference plot in an undamaged stand (Figure 2). When selecting the plots, we applied the following criteria: (i) all stands under study must be growing in the same habitat; (ii) the stands must be of similar age; (iii) the stands must be growing in different damage zones; and (iv) the stands must be located in different compass directions in relation to the emitter. The first two criteria made it possible to largely eliminate the impact of habitat and age, whereas the third and fourth criteria allowed us to determine the temporal and spatial distribution of the growth reduction and the influence of wind direction on the degree of that reduction. All the stands under study represented an oligotrophic mixed coniferous forest habitat and were about 120 to 130 years old (Supplementary Material, Table S1). At each site, 20 trees were sampled with a Pressler increment borer at breast height, one core per tree. The sampling was performed at the end of the 2015 growing season (in November). In total, we collected cores from 160 pines growing at different distances and in different compass directions from the factory. The cores were prepared for measurement using standard dendrochronological procedures [52,53]. The samples were dried, placed in wooden mounts and sanded with progressively finer abrasive paper (80, 120, 180, 240 and 400 grit).

2.2.2. Data Analyses

The sanded core samples were scanned at a resolution of 2400 dpi using a standard scanner (EPSON Expression 10,000XL). Tree-ring widths were measured to the nearest 0.01 mm using Coo Recorder software [54], and individual growth sequences were created for each tree. Cross-matching checks were performed using the COFECHA program [55]. Then, TRW chronologies (raw and residual) were produced for each test site using the ARSTAN program [56]. The residual chronologies were obtained from double detrending (using a negative exponential curve followed by a cubic spline function with a rigidity of 64 years and 50% frequency cut-off); autoregressive modelling was also applied [53]. The homogeneity of the growth reactions and the strength of the environmental signal in the chronologies were estimated using the expressed population signal (EPS), correlation coefficient, GLK (Gleichläufigkeit) coefficient and t values between all pairs of series included in the chronology [52,53,57]. The following statistics were used to characterise the site chronologies (raw and residual): mean value, measures of variability, mean sensitivity, and autocorrelation [52,53]. These statistics have been calculated for three periods: a period before starting factory (1931–1966); a period of extremely high air pollution (1967–1995) and a period with a decrease in the emission of ammonia and a gradual decrease in air pollution (1996–2015). The Kruskal–Wallis test was used to determine the significance of differences between the respective research and control plots [58].

The Schweingruber method [59] was used to evaluate the impact of air pollution on the pine stands under study. This method relies on the analysis of characteristic years and abrupt changes in tree-ring width. The abrupt changes reflect major shifts in ecophysiological conditions that lead to the stimulation or inhibition of cambial activity over several successive years [59]. By using this method, it is possible to determine the exact onset and duration of abrupt changes in tree-ring width. With this method, the duration and degree of TRW reductions are calculated from the ratio of the sum of the reduced ring widths to the sum of the ring widths from the period preceding the reduction. The size of the reduction is classified as follows: RI (30–50%)—low reduction; RII (50–70%)—strong reduction; and RIII (above 70%)—very strong reduction. This research methodology has been successfully used in numerous studies on the impact of industrial emissions on forest stands in Poland [30,35,36,40,41]. The reduction in annual ring width was determined for each sample using the Quercus program [60]. This was done by comparing the dendrogram of each individual sample with the chronology developed for the reference site (plot no. 8).

The climate–growth relationships were investigated by calculating bootstrapped multivariate response functions between residual chronology and climate variables: monthly average air temperatures and monthly precipitation totals [52,53]. Response function analysis is a correlation and multiple regression model that links growth indices (as dependent variables) with climate parameters (as explanatory variables). The analyses were performed with reference to each study plot for the period spanning from June of the year preceding ring formation to September of the year of the current growth (16 months in total) using DendroClim2002 software [61]. The significance of the correlations was determined at p = 0.05. The climate data originated from the meteorological station in Puławy (see 2.1.2). Taking note of reports in the literature that the deposition of pollutants may potentially be a factor in the varying sensitivity of pine to climatic conditions [31,38], we performed dendroclimatic analyses for all plots separately for three periods: 1931–1966 (a period of 35 years before the commissioning of the factory); 1967–1995 (a period of 30 years of extremely high air pollution) and 1996–2015 (with a decrease in the emission of ammonia and a gradual decrease in air pollution from 1995 onwards) (Figure 4).

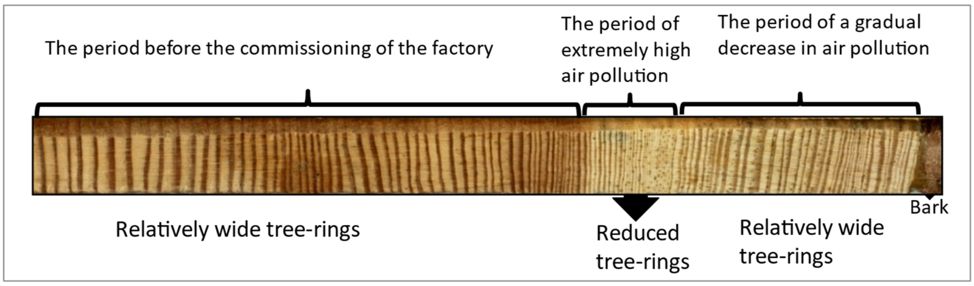

Figure 4. Illustration of a zone with reduced radial growth (sample taken from a pine growing on the 1st plot).

3. Results and Discussion

Our dendrochronological analyses have indicated the existence of significant and prolonged reductions in the annual ring widths of the Scots pine trees growing in all the forest stands under study (Figure 5). This points to a persistent, chronic decline in the vitality of the trees being examined. The decline began in the late 1960s, following the commissioning of the nitrogen fertiliser plant, which is when the pollutant emissions began. Reductions occurred in a majority of the trees under study (Table 1), although they were differentiated spatially and temporally.

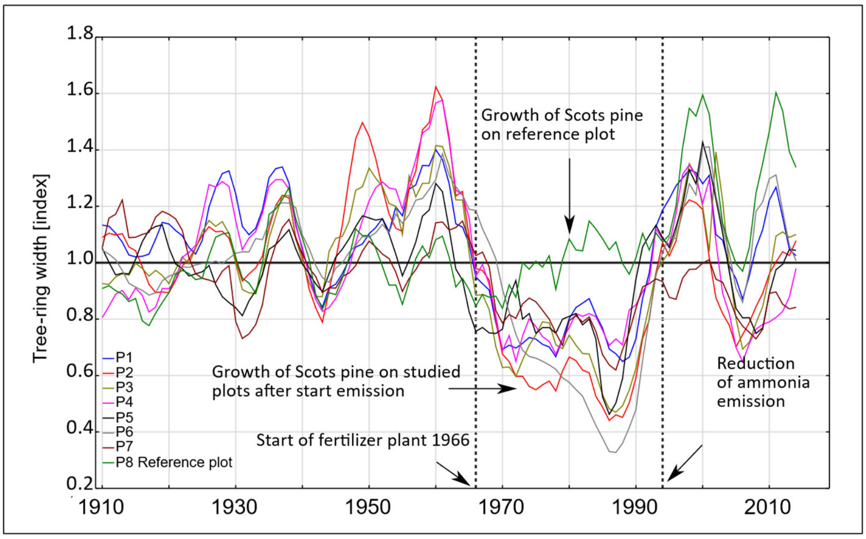

Figure 5. Chronologies of the Scots pines growing in the vicinity of Zakłady Azotowe Puławy. commissioning of the nitrogen fertiliser plant, which is when the pollutant emissions began. Reductions occurred in a majority of the trees under study (Table 1), although they were differentiated spatially and temporally.

Table 1. Characteristics of tree-ring chronologies, and results of calculation of growth changes under influence of air pollution from the nitrogen fertilizer factory in Puławy.

Plots	% of Absent Rings	% Trees with Reduction > 30%	Average Tree Ring Width a Period [index]		
			1931–1965	1966–1995	1996–2015
P1	5.7	66.7	1.14	0.72	0.89
P2	9.1	89.7	1.22	0.61	0.71
P3	6.7	76.9	1.2	0.66	0.82
P4	10.1	88.1	1.18	0.72	0.85
P5	3.9	62.5	1.04	0.8	0.96
P6	4.1	66.5	1.15	0.71	0.96
P7	1.7	41.2	1.00	0.85	0.97
P8-Reference	0.9	0	1.03	1.01	1.01

The most rapid and severe response to the pollution and the most pronounced reduction in growth (and thus decline in vitality) occurred within a radius of 3 km from the plant (plots 1 through 4) (Figure 6). In this zone, the share of trees with reduced growth was as high as 88%–89%. The ring width reductions occurred abruptly in the first two years after the commissioning of the plant and persisted over an extended period of time (more than 30 years); they were not directly linked to the prevailing wind directions. In plots 1 through 3, the reductions in growth persisted until around 1995 and were followed by an increase in growth. Then, after 2003, there occurred another growth reduction, although less pronounced. In plot 4, the reduction in growth began in 1968 and was still visible in 2015 (Figure 6).

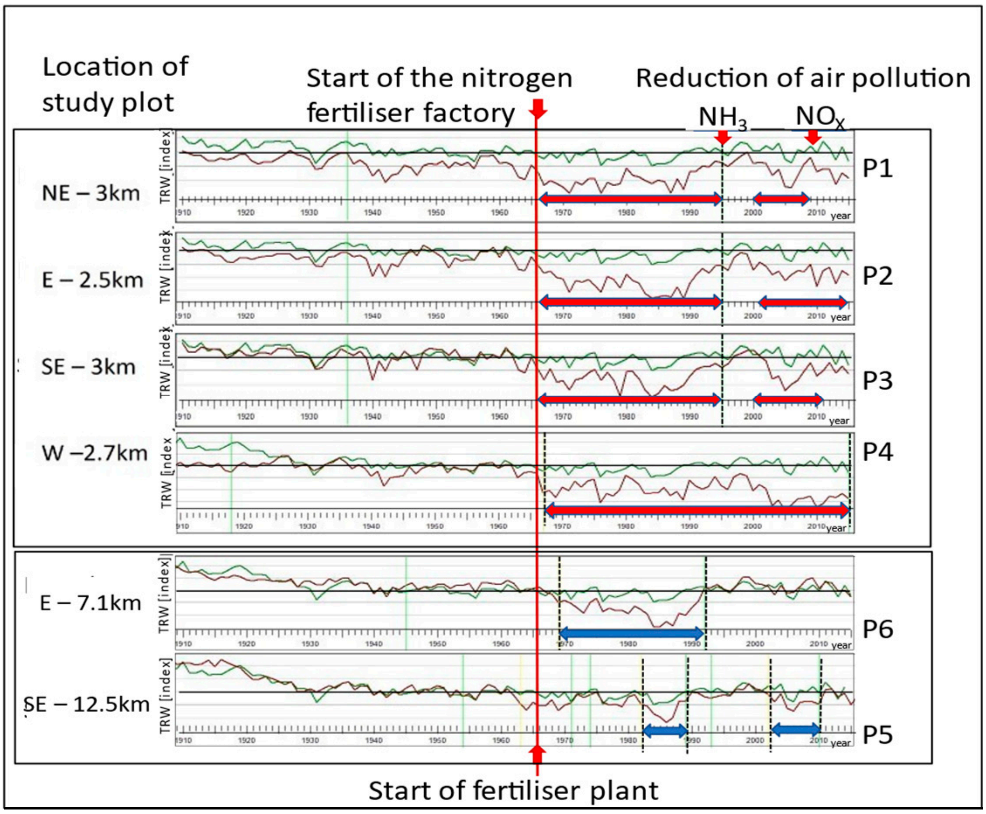

Figure 6. Reductions in the radial growth of the Scots pines growing in the vicinity of Zakłady Azotowe Puławy (selected plots). Explanation of markings: Brown lines—pine chronologies on

research plots, green line pine growth on the reference plot. Red horizontal arrows indicate the period of word reduction on surfaces located in the zone up to 3 km from the factory, blue arrows indicate the period of reduction in rings on surfaces located further from the factory. Vertical green lines indicate which the period to be taken into account when calculating the rate of growth reduction, reduction, following the Schweingruber method [59], e.g., diagram P1—the reduction period lasts from 1967 to 1995 (28 years), hence the preceding period 1938–1966 was used for the calculation of the reduction rate. See explanation in the text, methods section.

The spatial and temporal distribution of the growth reduction between the different study plots is shown in Figures 7 and 8. This distribution points to a clear relationship between the size of the reduction on the one part and the distance from the emitter and the prevailing wind direction on the other. Within a radius of 3 km from the source of the emissions, reductions in growth occurred almost simultaneously in a majority of the trees being studied (66.7% to 88.1%); the reductions lasted for many years and were strong or very strong (above 50%). The largest share of trees showing a very strong reduction in growth (above 70%) could be found in plot 2, which is located approximately 2.5 km east of the emitter, whereas the pines growing in the northern part of the zone (plot 1) showed a lesser degree of reduction. In the stands located further away from the emitter, the extent of damage to the trees generally decreased with distance from the emitter. However, trees growing in stands located in the prevailing downwind direction were affected more severely than those growing at a similar distance from the factory but located to the north of it (Figure 7).

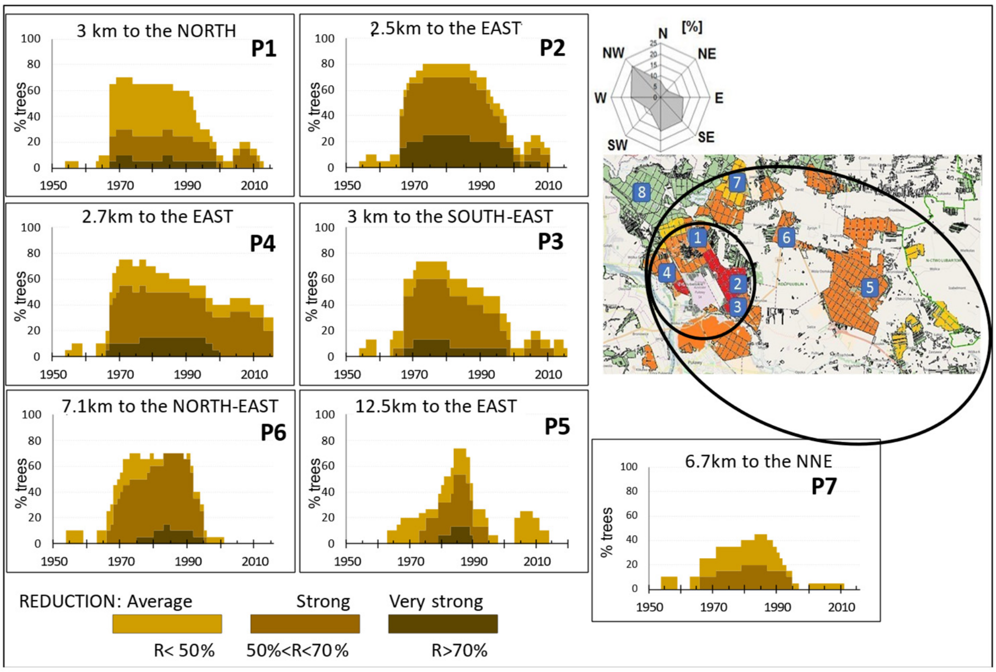

Figure 7. Graph showing growth reductions at the Scots pines in the different study plots.

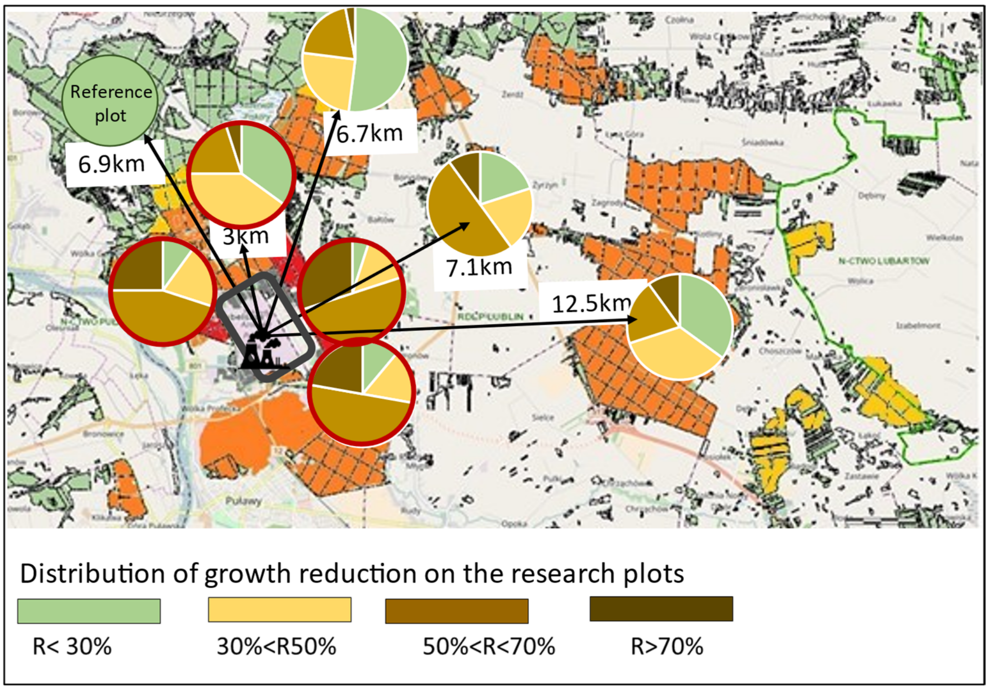

Figure 8. Spatial distribution of the reductions in the tree-ring width of the Scots pines.

In comparison with forest complexes growing near other nitrogen factories in Poland, the reductions in tree-ring width and the damage to pine stands in Puławy were much greater [30,62]. A similar degree of damage can be observed in forests growing in Silesia, which is the most polluted region of Poland. There, however, the negative effect of air pollution was more prolonged and the amount of pollution was significantly greater [36,63,64]. For that reason, we decided to search for an answer to the question of what other factors—apart from the excessive emissions of toxic substances—could have caused such reductions in growth and determined their spatial distribution.

We believe that the severe damage to the stands growing around the nitrogen factory in Puławy might have resulted from the synergistic influence of three factors: the emission of toxic substances, the relatively low height of the exhaust stacks that emit nitrogen compounds and the anemometric conditions that prevail in the region (Figure 9). The concentrations of gaseous pollutants are inversely proportional to the height of the emitter. Toxic nitrogen compounds were emitted from six stacks with a height of 47 m (ammonium nitrate) and five stacks with a height of 30 m (gaseous ammonia) [47]. Some of the nitrogen compounds also permeated into the atmosphere from the surface of industrial effluent tanks. The excessive emissions of nitrogen-based pollutants from the relatively low stacks under low wind speed conditions (with an average wind speed of 2.1 m/s) and the frequent periods of lull, especially in summer, resulted in the formation of particularly high concentrations of various pollutants near the factory itself (Figure 10). Consequently, the stands located in the immediate vicinity of the factory suffered the greatest amount of damage, regardless of their location in relation to the prevailing wind direction. Sulphur dioxide was emitted from a 160 metre-tall exhaust stack, which resulted in lower SO_2 levels in the vicinity of the nitrogen fertiliser factory itself, but the pollutants were transported over distances of up to 120 km (Figure 10). Therefore, one could conclude that the toxic nitrogen compounds (especially ammonia) were the main factors behind the degradation of the pine stands around Zakłady Azotowe Puławy.

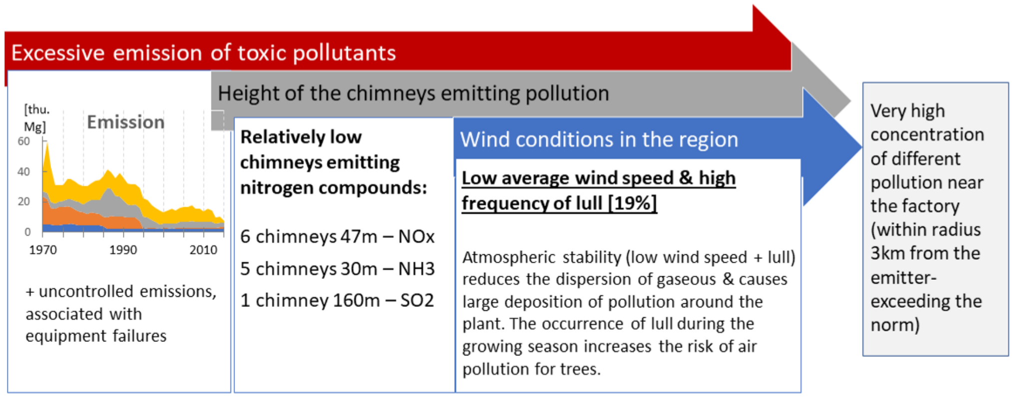

Figure 9. Possible factors determining the reductions in the annual ring width of the Scots pines.

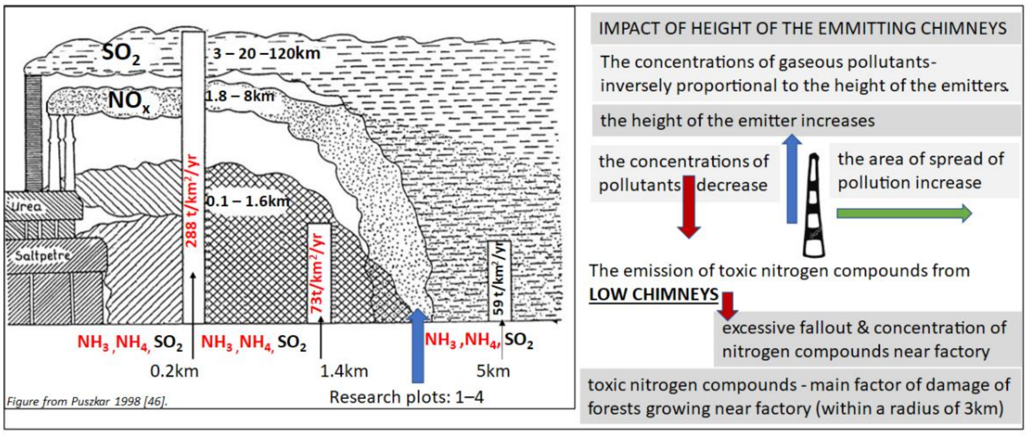

Figure 10. Extent and amount of air pollution emitted by Zakłady Azotowe Puławy.

In our opinion, this degradation—as recorded in the annual rings in the form of an abrupt and prolonged reduction in radial growth—is the result of both direct and indirect effects of toxic nitrogen compounds on the entire forest ecosystem. Our concept of this impact is shown in Figure 11. The sudden introduction of very high concentrations of nitrogen compounds into the forest environment caused a rapid, excessive toxic accumulation of nitrogen in pine needles, thus disturbing metabolic processes, causing a failure of the assimilation apparatus and death of apices, and ultimately leading to the dieback of some of the trees [43,44]. This, in turn, resulted in decreased stand density. In some of the pines, the dieback of needles and shoots caused a thinning of the crown. This permitted greater penetration of toxic pollutants and increased deposition of toxins on the needles, which was an ongoing process. At the same time, the decrease in stand density and the thinning of the crown allowed more sunlight to penetrate to the forest floor, causing changes in the microclimate—in particular by increasing the difference between the maximum and minimum air temperature. This entailed an increase in potential evaporation, decrease in mean air moisture level, increase in soil temperature, drying up of the soil, extension of the drought period and increase in water deficit [65].

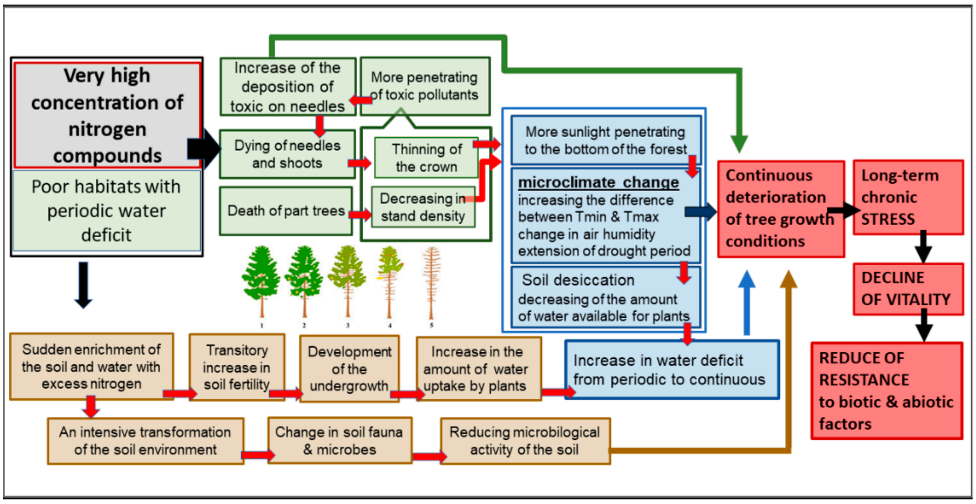

Figure 11. Concept of the direct and indirect impact of air pollution on the growth of the Scots pine stands under study. Explanation of the colours used in the figure: green—changes taking place in pine trees; blue—changes in the microclimate and water available; brown—changes in the soil environment; red—changes in vitality of Scots pine.

Very high concentrations of nitrogen compounds resulted in a sudden enrichment of the soil with excess nitrogen, which—as a consequence—caused an intensive transformation of the soil environment [46–48]. Prior to the construction of the factory, the soils in the area had generally been poor and characterised by periodic deficits of water and certain nutrients, especially nitrogen. The dieback of pine needles and shoots resulted in more sunlight penetrating to the forest floor, which—combined with a transitory increase in soil fertility—enabled extensive development of the undergrowth as well as bushes and birch and oak trees [42,46,47]. Changes to the composition of plant communities contributed to a rapid depletion of nutrients and an increase in water deficit. This was accompanied by changes to soil fauna and microbes and a reduction in the microbial activity of the soil [46].

Changes in the pine forest stands, microclimate and soil environment resulted in continuous deterioration of pine tree growth conditions, long-term chronic stress, a decline in vitality and decrease in resistance to biotic and abiotic factors. Interestingly, unlike the Scots pine, the emission from the nitrogen fertiliser factory in Puławy had a beneficial effect on oak and larches trees growing in the experimental Forest Range Ruda in Puławy (ca 3 km from factory), [66]. In the case of larch, an increase in the width of the annual rings was observed during the first decades of exposure to pollution, while oak increased its growth throughout the pollution period. According to Karolewski et al. [66], it is related both to the lower sensitivity of these trees to pollution than Scots pine, as well as to the fertilizing effect of nitrogen compounds.

As noted in the literature, pine trees weakened by air pollution are very often colonised by pests [22]. However, as the pest threat to the pine stands under study had been relatively low from the 1970s onwards [4,5], the impact of pests could not be investigated.

In regards to the climate, the question is whether the damaged trees are in fact more sensitive to climatic conditions.

A response function analysis performed for the 1951–2015 period showed that the main determinants of growth in the pines under study were temperature conditions in winter and early spring and precipitation in summer (Figure 12A). The above-average air temperatures of the January–March period contributed to the production of wide annual rings in the next growing season. Another factor that determined the trees' good health and the production of wide annual rings was the supply of water in June–August, that is during the period of greatest cambial activity in pine [31]. Droughts in that period were a strong inhibitor of growth. The dominant role of late-winter and early-spring temperatures in the development of annual rings in pine has also been noted by other authors [34,49,62–64,67,68]. The existing body of research confirms the existence of a common climate signal for Central Europe that differentiates the growth rhythm of this species [67]. As winter ends and daylight lengthens, pine loses its frost resistance and becomes sensitive to low temperatures. The freezing of needles, branches and trunks (resulting in frost damage), the drying-off caused by cold winds and the mechanical damage from snow all contribute to the deterioration of the trees' health and reduction in growth dynamics during the next growing season.

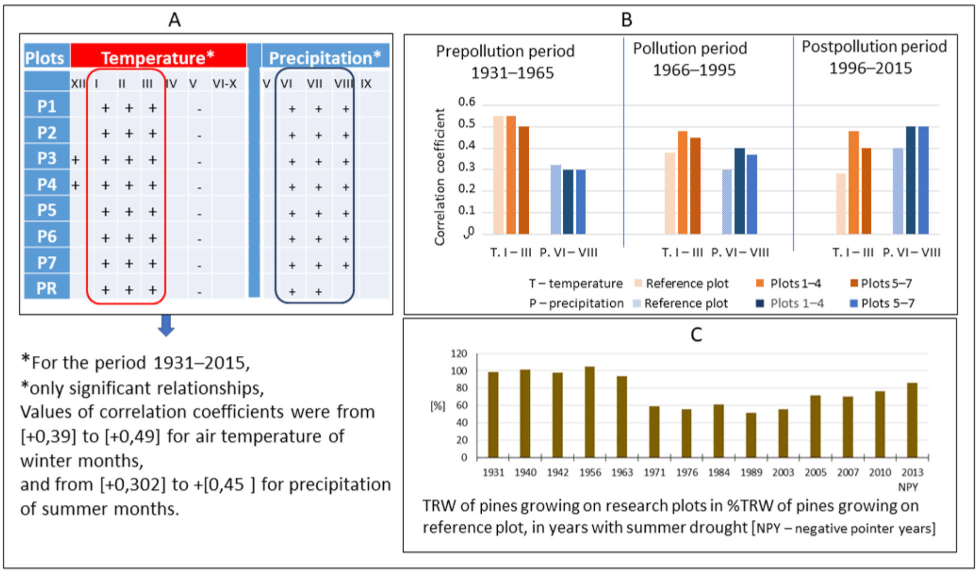

Figure 12. Climate—growth relationships for Scots pine growing on study plots. (**A**)—Results of correlation and response analyses for index pine chronologies from study plots (the values of correlation coefficients in Supplementary Material Figure S2); (**B**)—correlation coefficients between index chronologies, and winter temperature and summer precipitations, for different period; (**C**)—Tree-ring width of Scots pines growing on research plots (plots 1–4) in %TRW of Scots pine growing on reference plot, only years with extreme drought.

Analyses performed for shorter time periods—covering the years prior to the commissioning of the plant (1931–1965), the years of increased emission of airborne pollutants (1966–1995) and the years of successive reduction in toxic emissions (1996–2015)—demonstrated that the impact of climatic conditions on pine growth was similar in trend for all plots, but differences between the plots in terms of the strength of linkages began appearing in the late 1960s. Interestingly the correlation coefficient between TRW and mean air temperature of winter months decreases over time in the reference plot, while that with summer precipitation increases, witnessing the ongoing climate change with milder winters and drier summers (12B). Pines exposed to toxic pollutants were more vulnerable to cold winters and prolonged summer droughts. The reduction in industrial emissions and improvement of environmental conditions in the last decade of the 20th century resulted in the formation of wider annual rings.

Nevertheless, the trees are still weakened: they show reduced immunity to climatic stress and are more sensitive to adverse weather conditions, especially drought (Figure 12C). Similar observations concerning pine stands have been made by Oleksyn [69], Augustiastis [70,71], Vacek [26] and Putalova [34]. Furthermore, Vacek [26] demonstrated that the most severe damage can be attributed to the synergistic interaction between chemical stress and climatic stress, in particular in connection with a severe drought.

4. Conclusions

The extent and spatial coverage of forest ecosystem degradation in the Puławy area can be attributed to the amount and type of pollutants and to a number of local factors, especially the anemometric and habitat conditions and the height of the exhaust stacks (Figure 13). The high frequency of periods of lull combined with the low height of the stacks emitting the toxic pollutants multiplied the negative effects of the emissions. A radical reduction in pollutant emissions improved the environmental conditions, enabling the tress to grow once again, but the prolonged period of strong anthropopressure caused a long-term reduction in the trees' resistance to abiotic factors. Our research indicates that in areas with prolonged exposure to a high concentration of pollutants, the adverse impact of pollution on forests persists for a very long time and may be observed even 20 years after a radical reduction in emissions. These forests have reduced resistance to abiotic stress related to climate change, especially drought. Therefore, a greater impact of climate change—and in particular of extreme events—on the dieback of the trees growing in areas with strong anthropopressure can be expected.

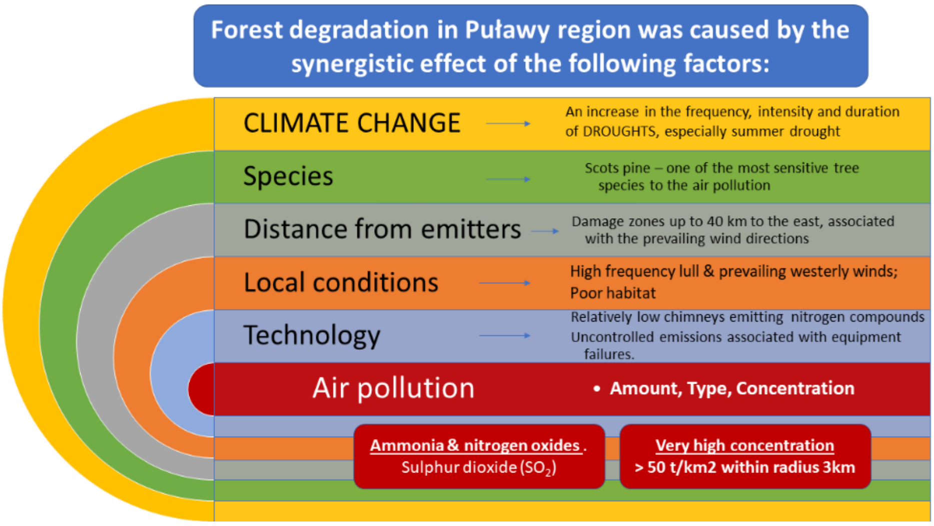

Figure 13. Factors determining forest degradation in Puławy region.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/f12101421/s1, Figure S1: The Tree-ring reduction caused by air pollution from Zakłądy Azotowe Puławy, Figure S2: Climate -growth relationships between residual chronologies and air temperature and precipitation, Table S1: Description of study plots in Puławy Forest District. GPS, Coordinates of the Study Plot.

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