

## Article

# Impact of Industrial Pollution on Radial Growth of Conifers in a Former Mining Area in the Eastern Carpathians (Northern Romania)

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**Abstract:** The research aims to evaluate the impact of local industrial pollution on radial growth in affected Norway spruce (*Picea abies* (L.) Karst.) and silver fir (*Abies alba* Mill.) stands in the Tarnița study area in Suceava. For northeastern Romania, the Tarnița mining operation constituted a hotspot of industrial pollution. The primary processing of non-ferrous ores containing heavy metals in the form of complex sulfides was the main cause of pollution in the Tarnița region from 1968 to 1990. Air pollution of Tarnița induced substantial tree growth reduction from 1978 to 1990, causing a decline in tree health and vitality. Growth decline in stands located over 6 km from the pollution source was weaker or absent. Spruce trees were much less affected by the phenomenon of local pollution than fir trees. We analyzed the dynamics of resilience indices and average radial growth indices and found that the period in which the trees suffered the most from local pollution was between 1978 and 1984. Growth recovery of the intensively polluted stand was observed after the 1990s when the environmental condition improved because of a significant reduction in air pollution.

**Keywords:** air pollution; increment cores; Norway spruce; radial growth series; silver fir



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## 1. Introduction

Climate change and air pollution represent the main drivers of global change, significantly impacting forest health and sustainable development [1]. Accelerated industrial development after World War II increased pollutant inputs in many parts of the globe, and Central and Eastern Europe were significantly affected [2]. The changes in political regimes in Eastern Europe and national and world environmental policy after the 1900s allowed for a significant reduction in air pollution in most regions. Nonetheless, investigating the environmental impact of air pollution resulting from anthropogenic industrial activities remains critically important [3].

Air pollution negatively affects forest ecosystems and soil quality worldwide. Industrial emissions from the Ivano-Frankivsk and Chernivtsi regions (Ukraine) have led to high concentrations of heavy metals in forest soils, high levels of tree crown defoliation and ecosystem changes such as biodiversity decline or reduced productivity [4]. Soil acidification in the region has risen progressively due to the increased content of heavy metals [5], which indirectly influences forest ecosystem vegetation [6]. In Germany, the

areas near recently halted mining operations were investigated to determine the uptake of heavy metals by forest trees in heavily contaminated ecosystems. The researchers also described the levels of damage caused by heavy metal toxicity [7]. A similar study in the Czech Republic analyzed the health status of Norway spruce (*Picea abies*) forests according to the phenology and radial growth of trees in relation to air pollutants, especially NO<sub>2</sub> and SO<sub>2</sub> [8]. The distribution and accumulation of heavy metals in forest soils in the Rozocze National Park (SE Poland) was found to be related to anthropogenic pollution through local and background emission sources [9]. In the Carpathian Mountains, the results of long-term monitoring activities indicated that the combined effects of O<sub>3</sub>, SO<sub>2</sub> and NO<sub>2</sub> could negatively affect forest stands and highlighted the association between air pollution levels and tree growth [10]. Furthermore, elevated levels of N and S deposition at the levels found in the Carpathian Mountains may have negatively affected forest health status and biodiversity, including visible leaf injury, losses in stand growth and productivity and higher sensitivity to biotic and abiotic stressors [11,12]. The accumulation of heavy metals with accompanying S and associated soil and foliar nutrient imbalances and reduced soil water holding capacity can restrict the recolonization of plant communities in the forest ecosystem [13]. In the recent past, some industrial activities in Romania (Copșa Mică, Zlatna, Baia Mare) were known to create regional hotspots of pollution, negatively affecting forest vegetation [14–17]. Toxicity thresholds for the forest environment in Romania were highlighted based on air quality analysis [18].

Trees are sensitive to environmental factors, and any changes in growing conditions are reflected in tree ring parameters. The reduction in tree growth is generally associated with unfavorable climate conditions and an increase in specific ecosystem competition. Furthermore, air pollution can be associated with narrow growth rings for several decades.

The evolution of forest ecosystems affected by past pollution in highly industrialized areas and damage dynamics assessments offer crucial knowledge needed to develop management strategies for the conservation and improvement of the environment. Thus, to accurately assess how severely trees or forests have been affected by pollutants, it is critical to study the issue in well-defined ecological areas. For the northeastern part of Romania, the Tarnița mining exploitation constituted a hotspot of industrial pollution. The primary processing activity of non-ferrous ores containing heavy metals in complex sulfide forms was the main cause of pollution in the Tarnița region. Tarnița mining exploitation began in 1968, and the amount of production increased until 1990 through the exploitation of new deposits. With the political regime change in Romania, concomitant with the economic recession after the fall of communism, there was a sharp decline in mining activity, followed by a cessation in 1998 [18].

Considering this specific pollution history, the aim of this research was to evaluate the impact of local past industrial pollution on radial growth in affected Norway spruce (*Picea abies*) and silver fir (*Abies alba*) stands in the Tarnița study area, Suceava (northern Romania).

## 2. Materials and Methods

A network of experimental plots was established in five representative yield management units in the Tarnița region, Suceava county, within the Stulpicani Forest District (FD) (Table 1). This network of experimental plots included 30 plots (15 for silver fir and 15 for Norway spruce) from which radial increment cores were collected (40 trees for each species per plot). In order to highlight the level of pollution intensity on the silver fir and Norway spruce stands in the Tarnița area, the 30 plots were located spatially at different distances from the main source of pollution. Taking into account previous research [19,20], in which several stands located at different distances from the source of pollution were analyzed and it was concluded that the 2 km length was the approximate distance to and from which the stands were affected by pollution to varying degrees of intensity, we tested this hypothesis and we considered in this study that intensively polluted stands are located at a maximum distance of 2 km from the main source of pollution, moderately polluted stands are located between 2 and 6 km and largely unpolluted stands are located at a distance greater than

6 km. Thus, based on the results obtained in these studies, in order to test our hypothesis, we considered these distances as limits between different pollution intensities.

**Table 1.** The main characteristics of the experimental plots network from Tarnița region.

Forest Management Unit (u.a.)	Yield Management Unit (UP)	Distance from the Polluting Source (km)	Area (ha)	Age	Composition *	Exposure	Slope (Centesimal Degrees)	Altitude (m)	Canopy Cover	Yield Class	Standing Volume (m <sup>3</sup> /ha <sup>-1</sup> )
73C	V Tarnița	6.0	4.90	95	9MO1BR	NE	28	1120	0.8	2	585
62A	V Tarnița	3.1	6.34	105	4MO3BR2FA1PAM	NE	25	985	0.7	2	416
39A	V Tarnița	0.5	4.13	85	8MO2FA	NE	16	860	0.4	2	304
111E	V Tarnița	1.2	4.67	105	4MO4BR 2FA	SE	26	980	0.6	3	396
118A	V Tarnița	2.5	18.45	140	4MO4FA2BR	S	26	1125	0.5	3	314
18F	V Tarnița	3.2	19.54	140	5MO3FA2BR	NW	36	1125	0.6	3	370
14C	V Tarnița	1.6	6.87	95	5BR3FA2MO	S	26	900	0.7	3	402
126I	V Tarnița	1.2	5.2	100	6MO2BR 2FA	SE	16	875	0.6	2	346
5A	VI Botoșana	4.5	29.84	95	7MO2BR1FA	N	27	905	0.7	1	572
17B	VI Botoșana	3.1	10.66	75	8MO2BR	N	18	1000	0.8	2	589
45A	VI Botoșana	6.9	8.57	115	6MO2BR2FA	N	18	775	0.7	2	601
61A	VI Botoșana	6.0	16.22	110	4MO3FA2BR1PAM	SE	22	1000	0.7	2	482
43B	IV Porcăreț	6.2	24.2	95	6MO3BR1FA	NE	24	990	0.7	2	513
22A	II Negrileasa	9.7	28.78	95	5MO4BR1FA	N	22	850	0.7	2	560
4B	VIII Gemenea	12.0	18.10	110	8MO2BR	NW	12	825	0.6	2	482
101C	VIII Gemenea	12.1	13.03	125	8BR2MO	SW	33	740	0.6	2	474

\* Degrees of participation (in tenths) of the species in the mix forest stand; Norway spruce (MO), silver fir (BR), European beech (FA), sycamore maple (PAM).

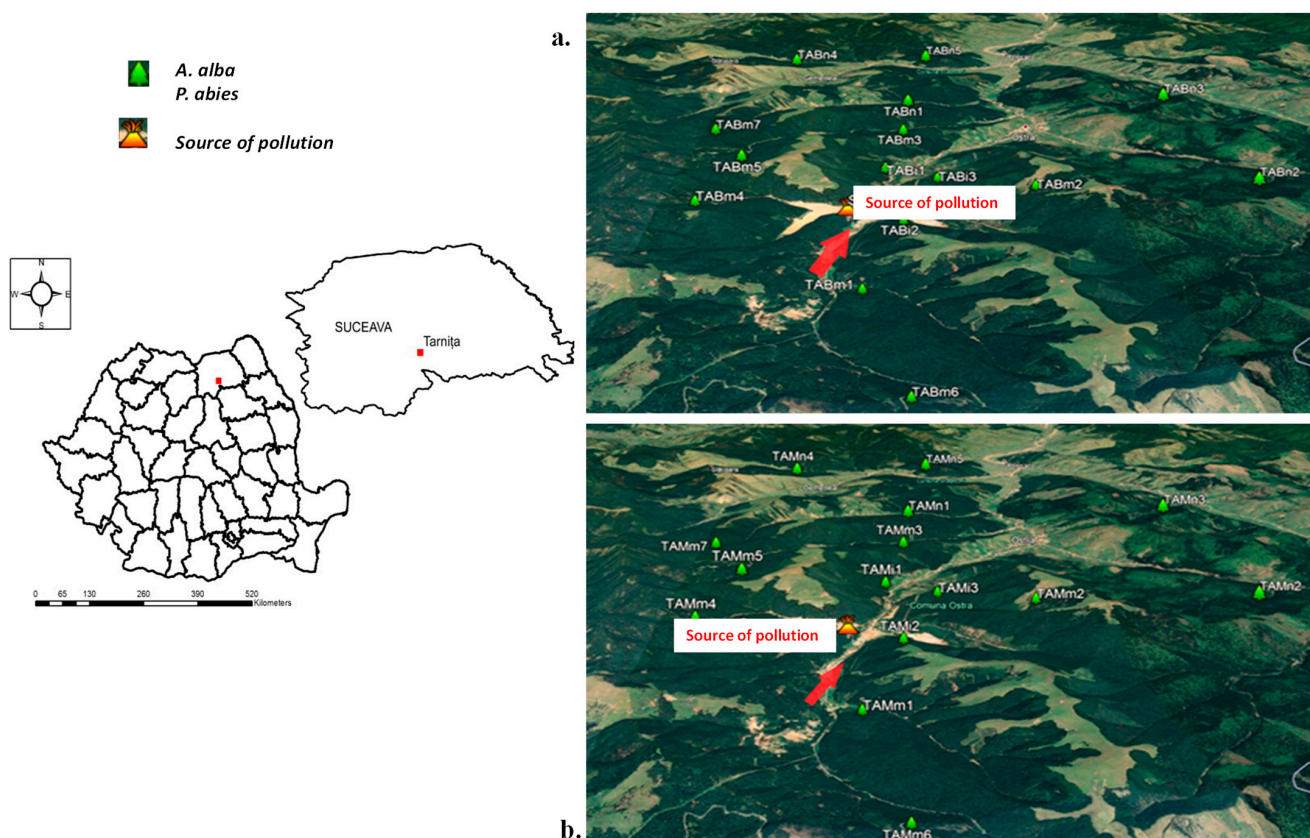
Concerning the characteristics of the studied stands, the trees included in the forest stands of the network within the Tarnița study area were between 75 and 157 years old. The tree stand composition was generally a mixture of Norway spruce (the predominant species), silver fir and European beech (*Fagus sylvatica* L.). Plot altitude varied between 750 and 1150 m. The studied stands had a canopy cover between 0.6 and 0.8 and were mostly of higher productivity (classified in the 2nd relative yield class). The volume per hectare was between 346 and 601 m<sup>3</sup>.

For the classification of the radial growth series in relation to the distance from the polluting source, both the distance from the source and the predominant direction of airmasses (NE–SW) were taken into account. Thus, there were six series of growth (three silver fir and three Norway spruce) located in the intensively polluted area, 14 series in the moderately polluted area (seven silver fir and seven Norway spruce) and 10 series in the largely unpolluted area (five silver fir and five Norway spruce). In the direction of the main valley, the stands were intensively polluted up to distances of 6–7 km (Figure 1).

The following specific statistical parameters were calculated, both for radial growth series and tree ring index series [21,22]: the period covered by each series with a minimum replication of 10 individual series, sample depth, mean tree ring width, average sensitivity (average percentage change of annual ring width relative to the next annual ring [21]) and average Rbar (correlation coefficient between all individual series).

The degree of reduction and recovery of growth due to the influence of local industrial pollution was determined through the resilience indices, presented as a 5-year moving average. Tree resilience is its post-disturbance ability to reach the level of radial growth it experienced before the disturbance, calculated as the ratio between the pre- and post-disturbance growth [23]. The tree resilience calculations were performed for each growth series analyzed, revealing the capacity of trees to grow after the disturbing events that caused reduced radial growth during certain periods.

Radial growth indices of intensively and moderately polluted trees were compared to the radial growth indices of the unpolluted trees, considered reference values. The calculations and analyses were performed for the period common to all analyzed series from 1951 to 2018. The highlighting and quantification of the growth changes of the stands in the areas affected by the industrial pollution were performed using software such as CooRecorder 7.4 [24], CDendro 7.6 [24], TsapWin [25], COFECHA [26,27] and R studio [28].



**Figure 1.** Study location: (a) Silver fir from Tarnița region; (b) Norway spruce from Tarnița region (the red arrow indicates the source of pollution and the predominant direction of airmasses in the area is NE–SW, according to the valley orientation).

### 3. Results

#### 3.1. The Statistical Parameters of the Series of Average Radial Growth

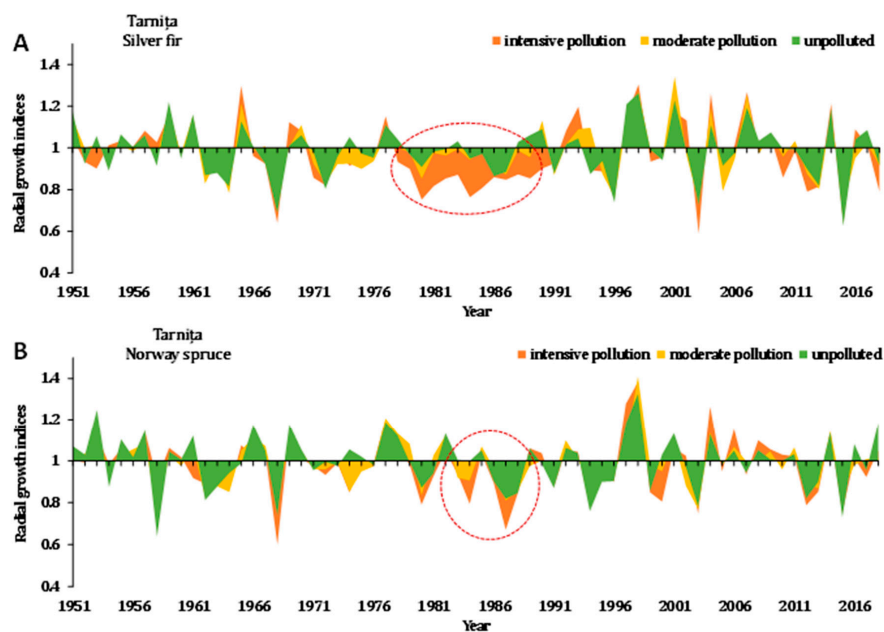
The main statistical parameters for all average radial growth series studied are shown in Table 2. The length of the analyzed silver fir series from the Tarnița area varied between 75 and 157 years, with an average ring width value between 1.704 and 3.346 mm. The mean values of sensitivity varied from 0.196 to 0.253, and the mean Rbar values of the residual series were between 0.219 and 0.411. The Norway spruce growth series lengths varied between 72 and 143 years, similar to silver fir. The average value of the tree ring width varied between 1.812 and 3.447 mm. The Norway spruce Rbar values were lower than the silver fir, varying between 0.194 and 0.364 (Table 2).

#### 3.2. Analysis of Growth Changes of Trees

The average radial growth of the silver fir in the Tarnița area showed a similar dynamic regardless of the plot distance from the pollution source (Figure 2A). The exception was the period 1978 to 1990, during which the radial increments of intensively polluted silver fir were significantly lower than those from unpolluted areas. The average radial growth values of silver fir in moderately polluted areas during this period were intermediate between intensively polluted and unpolluted plots. According to average radial growth indices, the Norway spruce trees were the most affected by local pollution from 1978 to 1984 (Figure 2B). The silver fir trees in the Tarnița area demonstrated average radial growth dynamics (Figure 2A) that indicated they were more affected by the pollution than the Norway spruce.

**Table 2.** Statistical parameters of the average radial growth series for silver fir and Norway spruce in the Tarnița area.

Serial Code	No. of Cores	Overlapping Period > 10 Cores	Average Radial Growth	Average Sensitivity	Average Rbar	Location (FD/UP/u.a.)
TABi1	43	1943–2018	3.205	0.231	0.411	Stulpicani/V/126I
TABi2	40	1925–2018	2.881	0.236	0.311	Stulpicani/V/39A
TABi3	40	1861–2018	1.704	0.253	0.374	Stulpicani/V/14C
TABm1	41	1922–2018	3.104	0.216	0.219	Stulpicani/V/62A
TABm2	40	1930–2018	2.912	0.198	0.291	Stulpicani/V/18F
TABm3	41	1921–2018	2.686	0.211	0.280	Stulpicani/VI/5A
TABm4	43	1879–2018	2.409	0.209	0.234	Stulpicani/V/118B
TABm5	40	1927–2018	2.729	0.185	0.237	Stulpicani/VI/17B
TABm6	42	1896–2018	2.041	0.193	0.322	Stulpicani/V/73C
TABm7	41	1895–2018	2.578	0.226	0.275	Stulpicani/VI/61A
TABn1	40	1912–2018	2.882	0.195	0.256	Stulpicani/VI/45A
TABn2	40	1893–2018	2.301	0.203	0.343	Stulpicani/IV/43B
TABn3	40	1905–2018	2.632	0.228	0.319	Stulpicani/II/22A
TABn4	40	1919–2018	2.765	0.196	0.238	Stulpicani/VIII/4B
TABn5	40	1911–2018	3.346	0.216	0.390	Stulpicani/VIII/101A
TAMi1	43	1946–2018	3.447	0.221	0.313	Stulpicani/V/126I
TAMi2	40	1935–2018	2.993	0.210	0.304	Stulpicani/V/39A
TAMi3	41	1907–2018	1.872	0.297	0.314	Stulpicani/V/14C
TAMm1	40	1920–2018	2.738	0.228	0.209	Stulpicani/V/62A
TAMm2	40	1926–2018	2.473	0.198	0.397	Stulpicani/V/18F
TAMm3	40	1918–2018	2.463	0.217	0.306	Stulpicani/VI/5A
TAMm4	42	1891–2018	2.303	0.204	0.304	Stulpicani/V/118B
TAMm5	41	1938–2018	3.108	0.177	0.194	Stulpicani/VI/17B
TAMm6	42	1891–2018	1.812	0.193	0.311	Stulpicani/V/73C
TAMm7	40	1916–2018	2.680	0.213	0.365	Stulpicani/VI/61A
TAMn1	40	1909–2018	2.657	0.224	0.322	Stulpicani/VI/45A
TAMn2	41	1893–2018	2.201	0.210	0.248	Stulpicani/IV/43B
TAMn3	40	1901–2018	2.357	0.270	0.364	Stulpicani/II/22A
TAMn4	40	1915–2018	2.075	0.219	0.282	Stulpicani/VIII/4B
TAMn5	42	1875–2018	2.361	0.228	0.237	Stulpicani/VIII/101C

**Figure 2.** The average series of radial growth indices developed for each of the 3 categories of stands studied (the dotted circle represents the time interval in which the trees were most affected by air pollution); (A) silver fir; (B) Norway spruce.

From 1978 to 1990 (Figure 3A), only those resilience indices corresponding to the analyzed trees in the intensively polluted area had negative values. The analysis of the resilience indices (Figure 3B) revealed that the Norway spruce trees were also affected by the local pollution from 1978 to 1990, but to a much lesser extent than the silver fir.

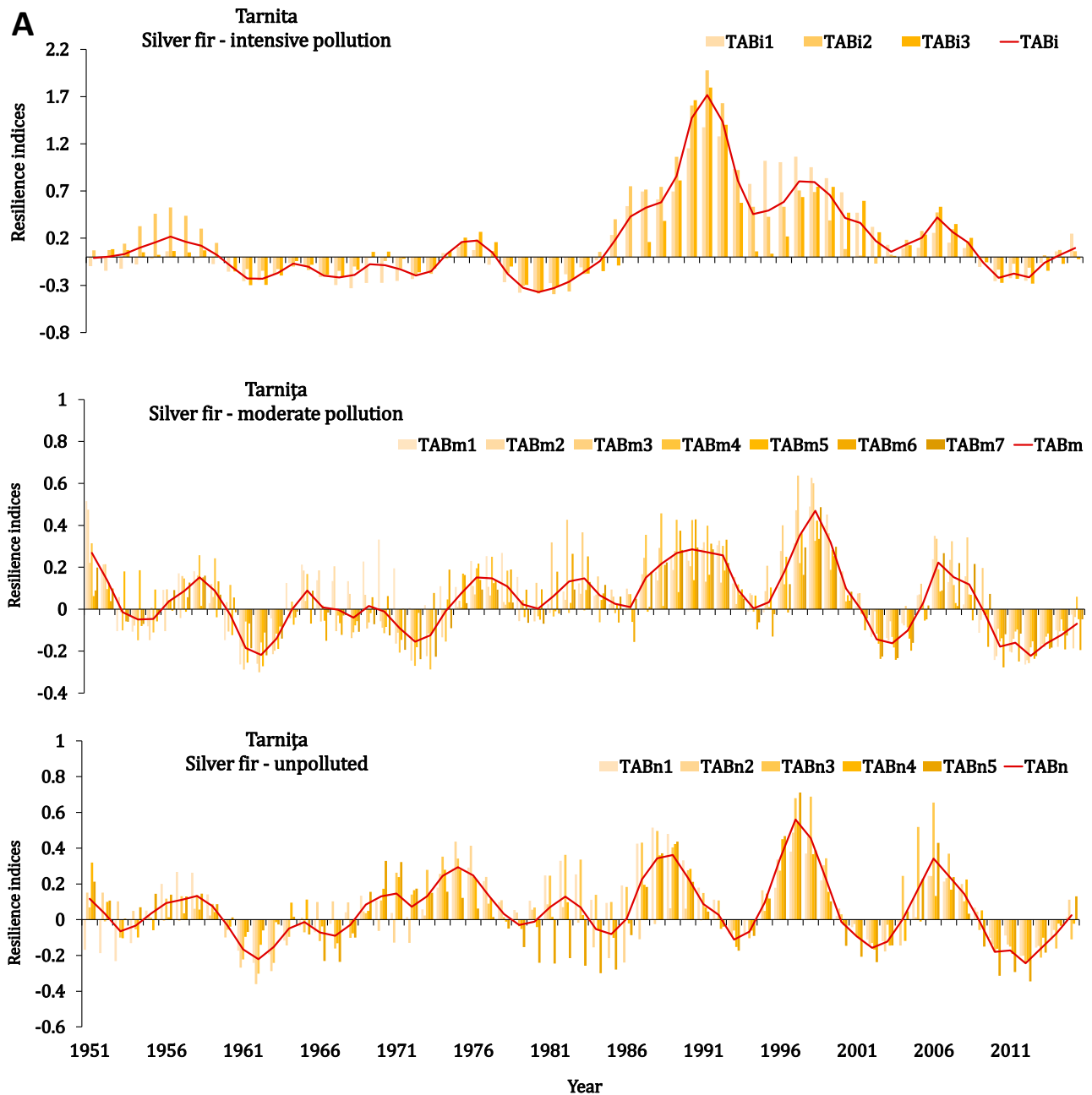
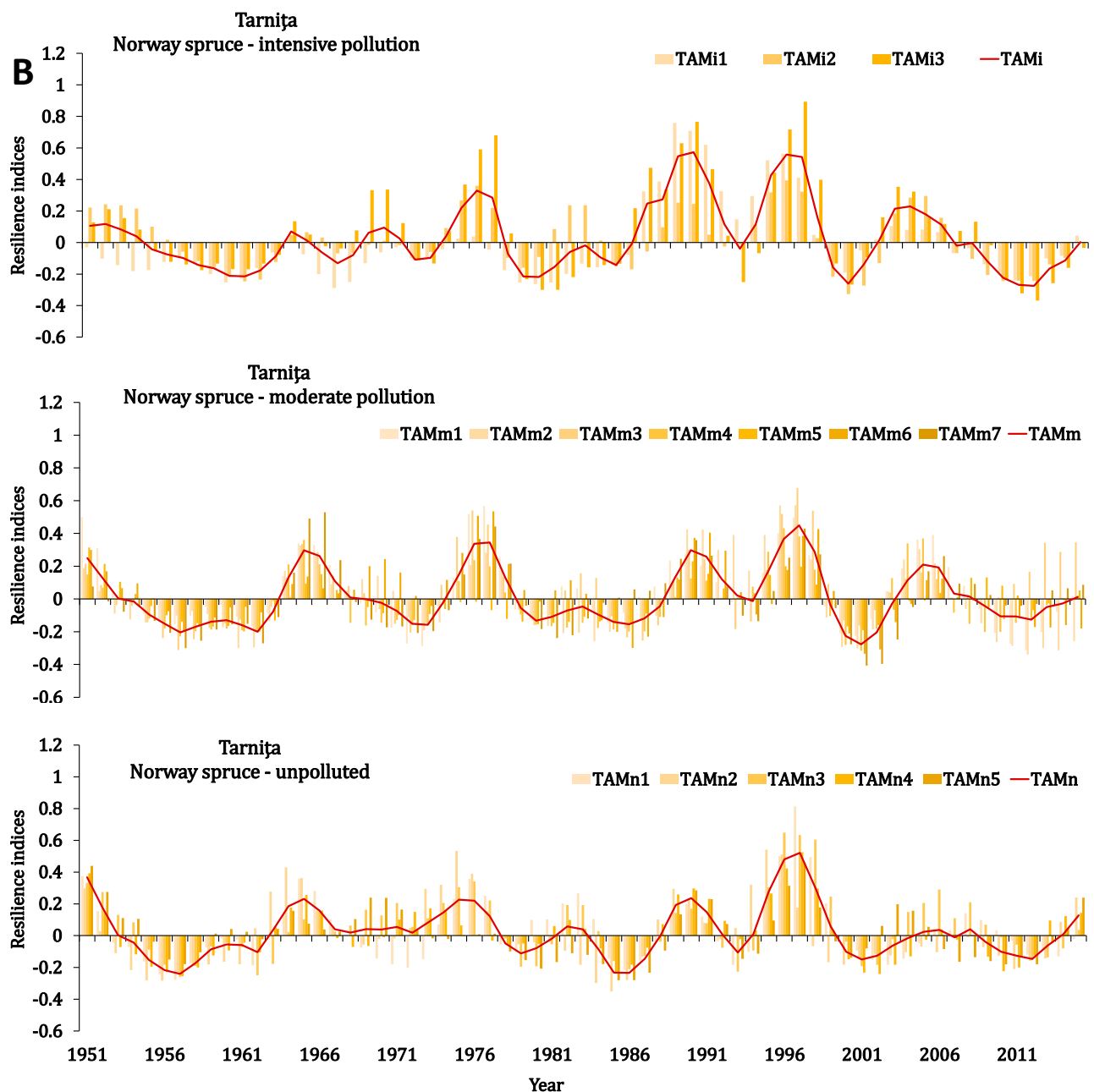


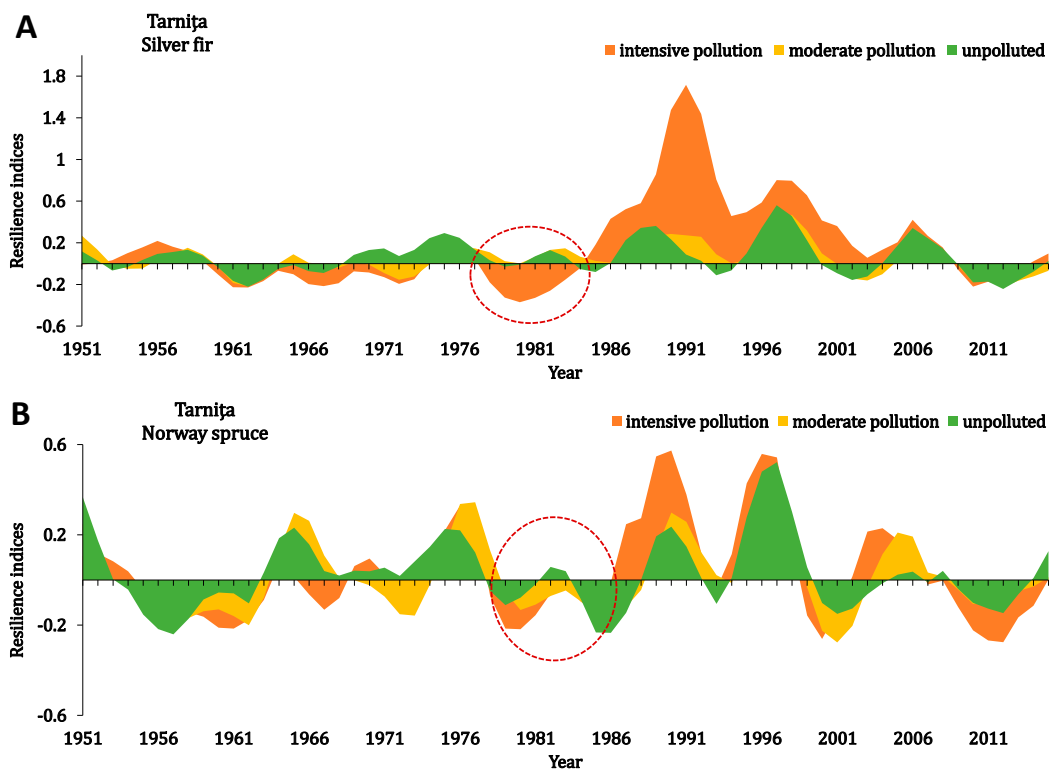
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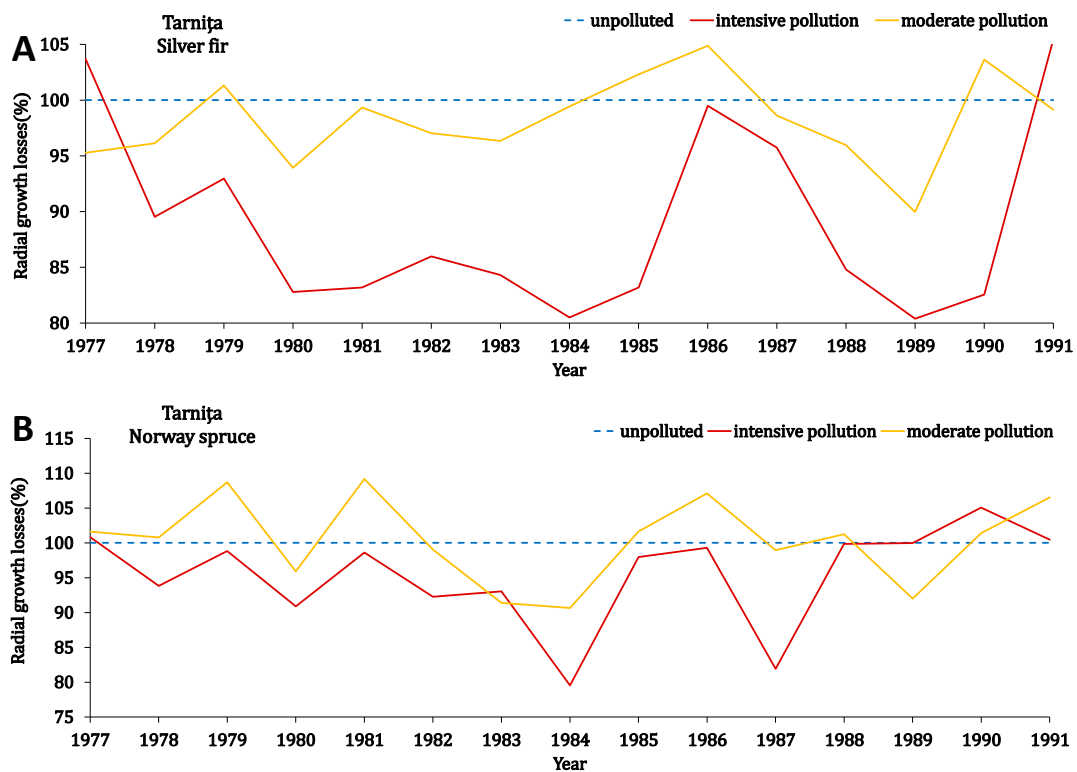
**Figure 3.** Resilience indices of the average radial growth series of silver fir (A) and Norway spruce (B).

For intensely polluted silver fir trees, after the cessation of the polluting activity, the resilience index values were significantly higher than those of trees in unpolluted areas (Figure 4A). From 1978 to 1990, for the silver fir trees from the intensely polluted area, the resilience indices (Figure 3A) and average radial growth indices for Norway spruce (Figure 4B) were much lower than those of the silver fir trees in the unpolluted areas.

The quantification of growth losses for silver fir reflects reductions of up to almost 20% (in 1984 and 1989) in heavily polluted areas (Figure 5A). The growth losses of trees located in moderately polluted areas were not as significant (up to 5–7% relative to normal). The average losses throughout the highlighted period for heavily polluted silver fir were approximately 14%. Compared to the silver fir tree, the Norway spruce suffered much smaller diameter growth losses (Figure 5B). The average loss of diameter growth of the intensely polluted Norway spruce during the entire period of pollution exposure was 5%, and the loss was only 2% for the moderately polluted.



**Figure 4.** Average resilience indices for each of the 3 categories of silver fir (A) and Norway spruce (B) stands in the Tarnița area (the dotted circle represents the time interval in which the trees were most affected by the influence of pollution).



**Figure 5.** Radial growth losses recorded by the trees ((A) silver fir; (B) Norway spruce) in the Tarnița area affected by moderate and intensive air pollution.



At the stand level, the most affected trees by the local industrial pollution were the silver fir trees, while Norway spruce trees were less affected.

#### 4. Discussion

As in the recent study developed in the same area [18], whose results confirmed that the frequency of growth events is determined by the distance from the sources of pollution, our results indicate that pollutant emissions near the local pollution zone significantly impacted the growth and development of coniferous trees in the Tarnița region, Suceava. In the studied area, the negative effect of pollution on the radial growth of coniferous trees (silver fir and Norway spruce) was greatest from 1978 to 1990. During this period, silver fir trees in the intensively polluted area experienced radial growth losses of up to almost 20% in 1984 and 1989. The growth losses of trees located in moderately polluted areas were not as significant, up to 5–7% compared to normal. The average loss throughout the highlighted period for heavily polluted silver fir was approximately 14%. In the case of Norway spruce, from 1978 to 1990, the trees were much less negatively affected by local pollution than the silver fir tree. The period in which the Norway spruce trees were most affected by local pollution was between 1978 and 1984, followed, except for 1987, by a period in which the trees did not experience as much growth reduction. Compared to the silver fir, the Norway spruce in this area showed much smaller radial growth losses. The average loss in radial growth of the intensively polluted and moderately polluted Norway spruce during the entire period of local pollution influence was 5 and 2%, respectively. After the 1990s, we observed a significant improvement in radial growth linked with the reduction in air pollution due to the closing of the mine. These results confirm those obtained in previous studies in this area [29]. Similarly, growth losses were registered as an effect of the long period of excessive drought [30]. The impact of air pollution on tree growth revealed by our results is slightly underestimated, because in the analysis were included only the trees that survived until the present. The growth decrease would likely be more evident in trees that did not survive the period of high industrial activity [18].

The effects of air pollution on forests are observed mainly as a direct impact on tree health, by crown damages and abnormal defoliation, favorable to losing tree vitality and in some cases, even death [31]. Coniferous trees are more sensitive to the effects of air pollution and acid rain than broad-leafed trees because of their greater capacity to intercept water from precipitation [32]. Similarly, in other regions of Europe with high air pollution, the silver fir is more pollution-sensitive than spruce [2,33].

Mihaljevič et al. [34] analyzed the annual tree rings relative to the mining period and a potential source of contamination, a high concentration of cobalt (Co) that corresponded to maximum mining production. Hojdová et al. [35] assessed contaminated soils and vegetation surrounding mining areas. The authors found a strong correlation between Hg concentration in beech and mining metal production and no correlation with spruce trees located closer to the source of pollution. Numerous studies on this topic have proposed that emissions of heavy metals cause imbalances in the forest ecosystem and beyond. Shparyk et al. [4] showed that the highest levels of defoliation of trees were close to sources of industrial emissions. Biochemical investigations performed on leaves and phloem in tree trunks revealed decreased assimilative pigments in trees in those areas affected by pollution [17]. Changes in the processes of photosynthesis, respiration and transpiration occur differently in intensity both at the individual and species levels [36,37]. Barium mining activities can affect the quality of sediments and soil water through pollution with Fe, Hg and Pb, indicating an unacceptable risk to human health and the environment. After assessing the degree of contamination and the risks due to barite mining, the authors showed that the average concentrations of Fe, Hg and Pb were above allowable levels [38].

The radial growth of trees is certainly influenced by the variation and capacity of mining production. In our study, differences found in the dynamics of radial growth indices between the two species analyzed were determined by the sensitivity and reaction

of each species to the imbalance of the ecophysiological process. The Norway spruce had much smaller diameter growth losses than the silver fir.

## 5. Conclusions

Air pollution from Tarnița mining operations induced strong growth reduction from 1978 to 1990, undoubtedly related to a decline in tree health and vitality due to airborne pollutants. The growth decline in stands further away (over 6 km) from the pollution source was weaker or absent, and the tree ring width variability was related to climate variation. Growth recovery of the intensively polluted stand was observed after the 1990s when the environmental condition improved because of a significant reduction in air pollution.

Of the two species analyzed (silver fir and Norway spruce), the silver fir demonstrated a higher sensitivity to local pollutants. Analyzing the dynamics of resilience indices and average radial growth indices in an integrated and comparative way allowed us to determine that the period in which the spruce trees suffered the most from the effect of local pollution was from 1978 to 1984, followed by, except for 1987, a period in which the trees experienced less growth reduction.

**Author Contributions:** Conceptualization, C.G.S.; methodology, C.G.S., R.V. and A.S.; software, C.G.S.; validation, C.G.S., O.B., I.P. and R.V.; formal analysis, C.G.S.; investigation, C.G.S.; resources, C.G.S.; data curation, C.G.S., R.V. and A.S.; writing—original draft preparation, C.G.S.; writing—review and editing, C.G.S., E.A., O.B. and I.P.; visualization, C.G.S., E.A., O.B. and I.P.; supervision, C.G.S., E.A., O.B. and I.P.; project administration, C.G.S. and O.B.; funding acquisition, C.G.S. and O.B. All authors have read and agreed to the published version of the manuscript.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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