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Impact of Industrial Pollution on Scots Pine (*Pinus sylvestris* L.) Radial Growth in the Areas of Mineral Fertilizer Factory "Achema"[1]

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Abstract—The aim of research was to assess the changes of annual radial increment of Scots pine forests in the vicinity of intensive and moderate industrial pollution of "Achema" factory producing mineral fertilizers. Our results indicate that the radial growth of the pine tree can be divided into three different periods: growth promotion, growth inhibition and growth recovery. Low levels of nitrogen emissions were beneficial for tree growth: the radial increment in the intensive and moderate pollution zones increased by 15–25% and 10%, respectively compared to control. During the growth inhibition period, the total annual industrial emission was 37–40 kt, and 40–45% radial growth loss was observed for the closest and 15–20% for the most distant stands. The radial growth decrease slowed down and the recovery of damaged stands began in 1990–1992 then air pollution was considerably reduced. The rate of forest recovery was high for the most damaged stands: their radial growth was close to control in 2000–2011.

Keywords: industrial emissions, Scots pine, radial growth, promotion, inhibition, recovery

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The extent of damage to trees is determined by the concentration of pollutants in the environment as well as the duration of its impact on trees. The annual radial increment is among the best indicators of forest health condition (Eckstein, 1990; Stravinskiene, 2002) and can serve as natural monitor (Schweingruber, 1996).

In the last decades of the 20th century, the significant damage in forest stands across Europe was determined (Santamaria and Martin, 1998; Dobbertin, 2005; Lorenz and Mues, 2007, Kask et al., 2008; Laubhann et al., 2009; Pridacha et al., 2011, etc.). Most scientists found that the decline in forest health was caused by a combination of various factors, but the main factor causing large-scale forest damage is environmental pollution (Serengil et al., 2011; Matyssek et al., 2012). Unfavourable climate conditions (Innes 1998; Laubhann et al., 2009) are often mentioned together with different pollutants as factor strengthening their impact. A variety of factors in addition to air pollution contribute to reduction in tree growth. It was pointed out that the impact of air pollution on forests appears only under very high concentrations of pollutants and is revealed only locally (Manion and Lachance, 1992). Polluted air severely limits the growth of coniferous. Some researchers agree that reduction in tree radial growth, decline in productivity of forests, forest damage and deterioration are caused by dry pollutants (Schulze, 1989; Wei et al., 2012).

The inherent information in tree radial growth provides unique possibilities for retrospective assessments of radial growth rates over long periods and the evaluation of the effect of long-term air pollution on forest vitality (Stravinskienė and Erlickytė-Marčiukaitienė, 2009).

The main goal of this research is to estimate the long-term impact of industrial pollution on the radial growth of Scots pine (*Pinus sylvestris* L.).

MATERIAL AND METHODS

The largest polluter in Lithuania, JV "Achema" factory producing the mineral fertilizers, is situated in Jonava district at the centre of Lithuania. "Achema" started production in 1965 and gradually expanded. The largest compound stream of total emissions was in 1979–1983: every year 37–40 kt of pollutants were emitted into the atmosphere. Annual sulphate emissions amounted to 100 kg/ha in the intensive pollution zone near the source of pollution and 87 kg/ha in the moderate pollution zone, which exceeded background concentrations by 2.5 and 2.2 times, respectively. Annual nitrate emissions exceeded the background by 5.9 times in the intensive pollution zone and by

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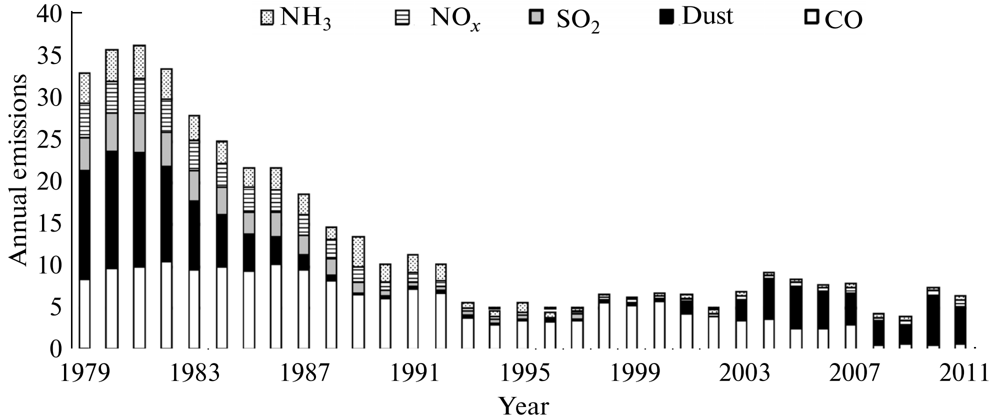

Fig. 1. The main annual emissions from "Achema" factory in 1979–2011 (according to the data from the Department of Environmental protection of "Achema").

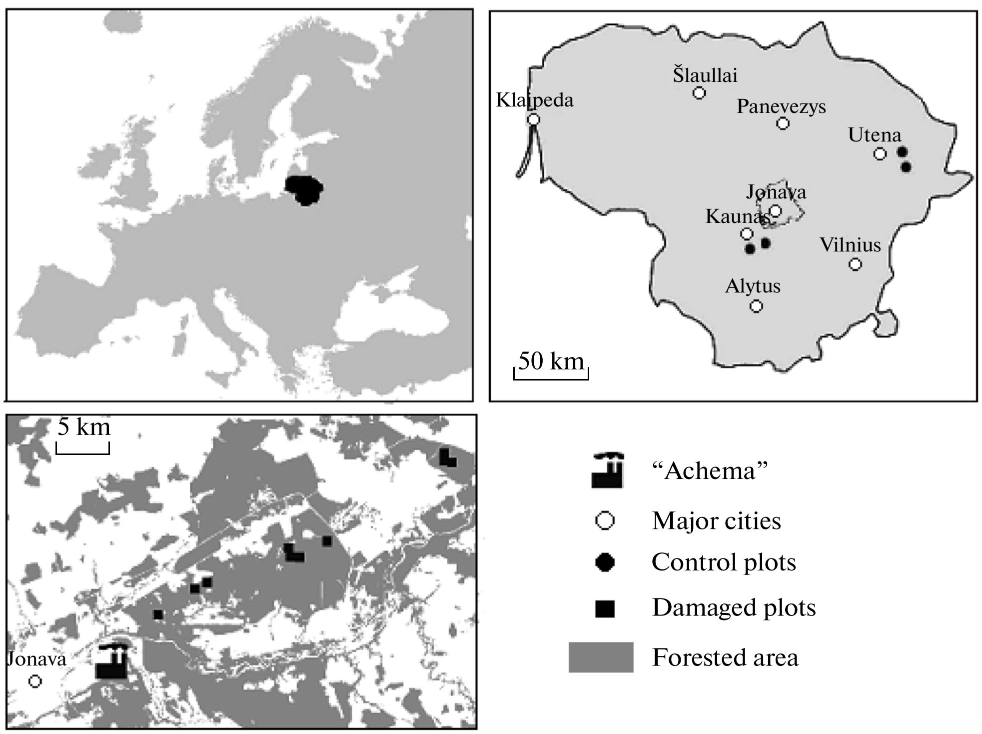

Fig. 2. Map of sampling sites.

4.8 times in the moderate pollution zone (Armolaitis, 1998). The major components of "Achema" emissions are NO_2, NH_3, CO, SO_2 compounds, and mineral dust. The main emissions have started to decrease since 1983, and until 1999 were reduced by more than 6 times, as well as until 2009—about 9 times (Fig. 1).

"Achema" factory is surrounded by the Scots pine forests of the Jonava forest enterprise. The prevailed sandy soils are classified as *Arenosols* (FAO/UN, 2006). An investigation was conducted in the vicinity of this factory. Ten damaged semimature (80–90 years old) Scots pine (*Pinus sylvestris* L.) stands growing in *Pinetum vaccinio–myrtillosum* forest types 3–10 and 11–20 km away from pollution source were chosen for this research. These forest stands were stocking levels of 0.7–0.9, mean diameter 33–35 cm, mean height 25–27 m, mean volume 200–250 m^3/ha, were either homogeneous or had up to 20% of other species, were located in the direction of prevailing south-westerly winds (Fig. 2). Four control stands were selected in the relatively clean environment in Utena district (Aukštaitija national park) and Kaunas district. Control forest stands by its age and all biometric parameters were similar to the stands growing nearby the "Achema".

Using the international monitoring methodology (UN/ECE, 1994), 25 Scots pine trees of 2 (codominant) class according to Kraft's classification were selected as samples in each investigated stand. 250 trees from damaged stands and 100 trees from relatively healthy stands were chosen as samples. Wood samples (cores) were taken by Pressler's borer at 1.3 m height from a root collar from two directions (1 from the southern and 1 from the northern side of the tree) to eliminate the effect of tree radial growth eccentricity (Schweingruber, 1988). 500 cores from the damaged stands and 200 cores from the relatively healthy stands were taken. The total number of cores used in this research is 700.

For tree radial increment measurement (accuracy ±0.01 mm), the tree-ring measurement system (LINTAB), and set of programs (TSAP) were used. Early and late tree ring parts were measured separately.

"Statistica" and "Microsoft Excel" software were used for data analysis. The chronologies of radial increment from the stands at different distances from the plant were statistically compared by using methods of regression analysis.

Annual radial increment losses and/or additional increment (%) were calculated by comparing radial increment data of control and polluted forest stands with the formula:

$$Z_{\mathrm{n}} = \frac{Z_{\mathrm{k}} - Z_{\mathrm{i}}}{Z_{\mathrm{k}}} \times 100, \qquad (1)$$

where Z_{n} is the reduction of annual radial increment (or an additional increment when the increment of the sampled stand is higher than that of the control stand); Z_{k} is the increment of the control stand; and Z_{t} is the increment of the sample stand (Stravinskiene, 2002).

In order to determine the start, duration of growth promotion and growth inhibition periods, as well as an additional (or loss) of radial increment at different distances from the pollution source a simple (logarithmic) regression analysis, expressed by the following equation was used:

$$y = a + b\ln x, \qquad (2)$$

where y is dependent variable (start, duration of growth promotion period, additional or losses of radial increment and the duration of the growth inhibition period); x is the distance from the pollution source; a and b are coefficients.

The dependence of radial increment on the quantity of polluting emissions has been estimated by using nonlinear regression analysis. The following model was applied:

$$y = a + b\exp(-cx), \qquad (3)$$

where y—dependent variable (annual radial increment, % of control) at different distances from the pollution source); x—independent variable (the amount of polluting emissions, kt); a, b and c—coefficients.

In order to determine the impact of different polluting emissions on radial increment Pearson correlation analysis was used, and the following model was applied:

$$y = a + bx, \qquad (4)$$

where y—annual radial increment (mm) at different distances from the pollution source; x—the amount of polluting emissions (kt); a and b are coefficients.

To assess the quality of the models and compare their coefficient of determination R2, the level of significance $p < 0.05$ was used.

The experimental material for this research was collected in 2011–2012. The data on the quantity of "Achema" polluting emission was received from the Department of environmental protection of this factory. Since the start of factory production in 1965 until 1978 industrial emissions were not measured, so the information on emissions was not received.

Some results of our study on health condition of forests growing in the areas of industrial pollution of "Achema" have already been published (Juknys et al., 2002; Stravinskiene, 2002; Stravinskiene et al., 2013). This paper describes the latest findings of our research in the vicinity of this factory.

RESULTS AND DISCUSSION

Monitoring of forest conditions in Europe has shown that high concentrations of nitrogen and sulphur are harmful to trees and causes their gradual degradation and decline (Lorenz et al., 2008; Laubhann et al., 2009).

Since 1965, the emitted pollutants (mostly NO_2 and NH_3) had a fertilizing effect and positive impact on tree radial growth, as nitrogen is an essential plant element and promotes tree growth as a fertilizer (Spiecker, 1991). The growth promotion period is defined as the years from the initiation of the plant operation in 1965. It was estimated that the tree growth promotion effect in the pine forest stands growing in the least distant (3–10 km) from the pollution source started 1–3 years after "Achema" started production. In the most distant stands growing 11–20 km away from the factory, the growth promotion period started only 4–5 years after the opening of "Achema". Differences between annual radial increment in Scots pine stands at different distances from the factory were revealed. The growth promotion (fertilisation) period was seen in our previous study (Stravinskiene, 2002), as well as in the findings of other investigators (Augustaitis et al. 2011).

The duration of the radial growth promotion period in stands growing at various distances from the plant was different. For Scots pine stands growing nearby (3–5 km) the plant, this period lasted approximately 7–8 years. In more distant (9–11 km away) forest stands, the period lasted on average approximately 6 years. In the most distant stands (18–20 km away), this period was around 5 years.

It was found that in the growth promotion period an additional growth of trees varies from 10% to 25% (compared to the control) at different distances from the pollution source. The greatest changes occurred in the stands closest to the pollution source. In the stands, growing in the distance 3–10 km from "Achema", the average additional radial increment 15–25% ($p < 0.05$) compared to the control was observed. The average additional annual radial increment in stands 11–20 km from the factory was close to 10% ($p < 0.05$) of the control.

The dependence of growth promotion start, duration, and additional radial increment of Scots pine trees on the distance from the pollution source can be expressed by the following equations:

$$S = 0.25-3.18\log(L), R^2 = 0.78, p < 0.05, \quad (5)$$

$$D = 0.98-3.47\log(L), R^2 = 0.72, p < 0.05, \quad (6)$$

$$Z_n = 30.31-14.11\log(L), R^2 = 0.71, p < 0.05, \quad (7)$$

where S—start of growth promotion period, years; D—duration of growth promotion period, years; Z_n—additional radial increment, %; L—distance from the pollution source, km.

Several years after the growth promotion period, the impact of the increased amount of pollutants became detrimental and resulted in the decrease of Scots pine radial growth. It should be noted that after the most polluting Nitrophosca department began operations in 1978, annual total emissions from the plant increased to 37 kt in 1979.

The extreme cold winter of 1979–1980 was an additional external factor causing a very rapid decrease of pine tree radial growth. As this has been proven by other researchers (Pederson et al., 2004), unfavourable climatic conditions such as very low winter temperatures can reinforce the negative impact of pollutants and limit tree radial growth. Our earlier findings have shown that temperatures in winter influences Scots pine growth in Lithuanian geographic latitudes most essentially (Juknys et al., 2002; Stravinskiene, 2002).

Growth inhibition in most of the investigated stands started in 1980. At that time, it was a very cold winter, and the obvious signs of forest damage in the direction of prevailing winds up to 10–12 km from the pollution source were observed. The coniferous forests subjected to these conditions died 2–3 km from the pollution source. The duration of the growth inhibition period in stands at various distances from the factory differed significantly: 10–12 years for the closest most damaged stands, and 4–6 years for the most distant least damaged stands.

The greatest damage to forests was caused in 1980–1983, when the annual radial increment of trees closest to the plant reached only 55–60% of the control. The average loss of annual radial increment during the growth inhibition period comprised 40–45% of the control for the most damaged stands growing nearby the source of pollution and 15–20% of the most distant ones.

The dependence of growth inhibition period duration, annual radial increment loss on the distance from the factory can be identified by the following logarithmic regression models:

$$D = 15.42-8.42\log(L), R^2 = 0.85, p < 0.05, \quad (8)$$

$$Z_n = 50.96-0.74\log(L), R^2 = 0.47, p < 0.05, \quad (9)$$

where D—duration of growth inhibition period, years); Z_n—loss of annual radial increment, %; L—distance from the pollution source, km.

Emissions from "Achema" were reduced after a serious accident in the Nitrophoska department and its subsequent closure in 1989. In the beginning of the 1990s due to the industrial decline and modernisation of technologies emissions decreased gradually (Armolaitis, 1998). Essential reductions of air pollution emissions started in 1992. According to the data from "Achema" Department of environmental protection, the yearly amount of main pollutants comprised 4–8 kt in 1993–2011 (see Fig. 1), i. e., a few times lower than in 1979–1983. Over time, the structure of pollutants had changed and at present mostly NO_x (36%), CO (32%) and NH_3 (18%) gases are emitted. Along with such a significant decrease in overall emissions, the deposition of acid compounds has also decreased.

The results of our research show the different intensity of pollution impact on a radial increment of trees growing in forest stands at different distances from the source of contamination. When annual emissions of pollutants were low, the radial growth of Scots pine trees growing in the zones of intensive and moderate pollution was similar. When the annual amount of emissions has exceeded 31 kt the radial growth of trees in the intensive pollution zone at 3–10 km distance from the factory was significantly ($p < 0.05$) less nor in the stands at 11–20 km distance. Similar events have been revealed in Estonia (Kask et al., 2008), where study of tree radial growth in the vicinity of the cement factory was performed. When the pollution was the highest, it was found that radial increment of pines growing nearby the cement factory was significantly lower than in 38 km distance from the contamination source.

Results of exponential regression analysis (Fig. 3) have indicated that the impact of total polluting emissions is different for stands growing at different distances from the factory of mineral fertilizers "Achema", and the impact decreases with the distance ($R^2 = 0.85$ and $R^2 = 0.83$, respectively; $p < 0.05$).

As a consequence of the considerable decrease in environmental pollution, the decrease in Scots pine radial growth stabilised. The radial growth decrease

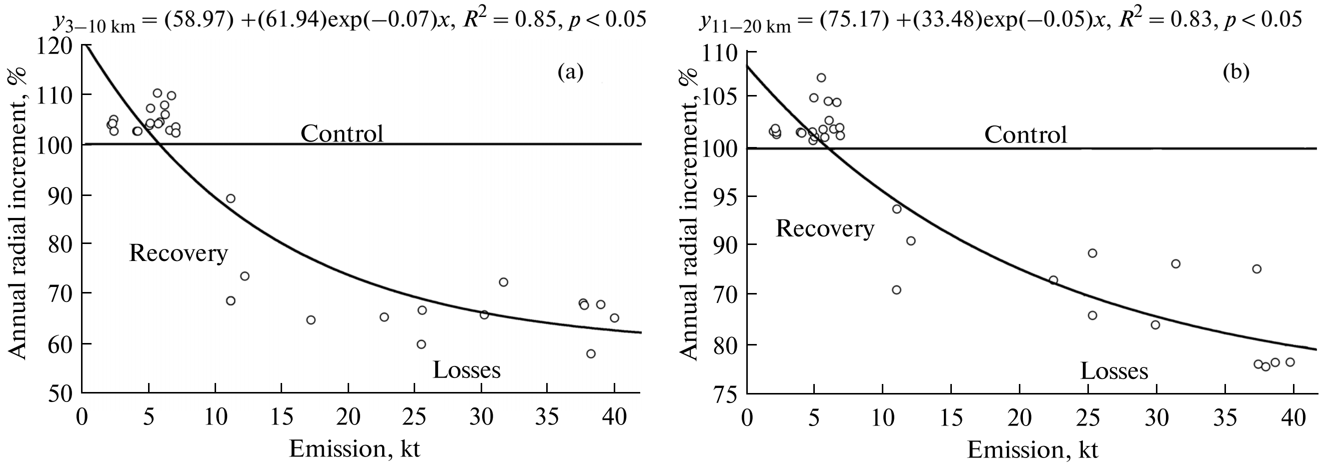

Fig. 3. Dependence of Scots pine (*Pinus sylvestris* L.) annual radial increment on amount of total emissions at 3–10 km (a) and at 11–20 km (b) distance from the pollution source in 1979–2011.

slowed down from in 1988–1989, even though climatic conditions were not favourable (rainy and cold growing seasons) for radial growth in trees (Stravinskiene, 2002).

The first obvious signs of recovery of damaged stands began in 1990–1992 when annual amounts of emissions decreased up to 11–12 kt. Our research has shown that Scots pine radial growth first began to increase in the stands more distant from the source of pollution. However, during the last 3–4 years, the most intensive recovery of actual growth rate and of the predicted normal annual increment has been discovered in the most damaged stands. Such positive changes could be considered a sign of decreased ecological risk for the surrounding forests.

Less damaged stands growing further from the factory recovered earlier. In 1995–1998, the health condition of Scots pines was restored. The annual radial growth in recovering stands was at the level of the first period (1965–1972) of pollution, in the growth promotion period.

During the period of pollution reduction, the pace of recovery was high for the most damaged stands. During the last decade (in 2001–2011), the most damaged stands showed the clearest recovery in radial increment and in compensatory growth. Presently, the radial growth of the previously strongly damaged and now recovering stands is stable and similar to the radial growth of the control stands.

In order to analyse in detail the impact of different pollutants (dust, SO_2, NO_x, NH_3), as well as the impact of overall emissions on radial increment, the period of pollution by "Achema" was divided into two stages according to the intensity of pollution (amount of pollutants):

1—the 1st stage—from the onset of pollution in 1979 till 1992, when the amount of annual emissions was 11–40 kt;

2—the 2nd stage (1993–2011)—reduction of emissions due to general industrial decline, modernization of technologies and effective use of resources; during this stage the amount annual emissions comprise 4–8 kt.

The results of Pearson correlation analysis have shown a statistically significant (r ranges from –0.66 to –0.75; $p < 0.05$) negative impact of each pollutant, as well as the impact of the total amount of main emissions on the radial increment of Scots pine trees growing in areas closest (3–10 km) to the pollution source during the 1st stage of pollution (table). Weaker, but statistically significant relations were found between radial increment of trees at a distance of 11–20 km from the factory and SO_2, NO_x emissions, as well as the total amount of polluting emissions (r ranges from –0.53 to –0.58; $p < 0.05$).

It was estimated that during the 2nd stage of pollution (1993–2011) in most cases the relationship between different components of pollutants, as well as the total amount of pollutants and radial growth of pine trees growing close (3–10 km) to the factory and 11–20 km away from it was statistically insignificant (table).

These results confirm that decrease in the amount of pollutants has led to positive changes in radial increment.

During the last 15 years, a number of studies have shown increased forest growth, the recovery of damaged forests, and improvement in forest health as the result of significantly reduced environmental pollution in central Europe (Solberg et al., 2009; Matissek et al., 2012, etc.) and the USA (McMahon et al., 2010). Our results confirm these trends and add to our knowledge of the local scale effects of air pollution on the radial growth and vitality of Scots pine forests.

Coefficients of correlations between the different pollutants and annual radial increment of trees during the different periods of pollution intensity (r—Pearson correlation coefficient, p—significance)

Pollutants	1979–1992				1993–2011			
	distance from the pollution source, km				distance from the pollution source, km			
	3–10		11–20		3–10		1–20	
	r value	p value	r value	p value	r value	p value	r value	p value
Dust	**–0.66**	0.01	–0.35	0.29	–0.42	0.07	0.25	0.28
SO_2	**–0.75**	0.02	**–0.57**	0.02	–0.39	0.06	–0.41	0.06
NO_x	**–0.74**	0.02	**–0.58**	0.03	0.32	0.08	0.43	0.06
NH_3	**–0.70**	0.03	–0.38	0.17	–0.49	0.06	–0.35	0.07
Overall emissions	**–0.69**	0.006	**–0.53**	0.05	–0.41	0.81	–0.18	0.44

Statistically significant r values ($p < 0.05$) are highlighted in Bold.

CONCLUSIONS

These generalised conclusions can be drawn from our research of radial growth of Scots pine in the areas affected by pollutants from the nitrogen fertilizer factory "Achema":

1—When the factory began polluting, the small amount of nitrogen compounds had a positive effect on the radial growth of Scots pine trees—in the zones of intensive (3–10 km) and moderate (11–20 km) industrial pollution, the radial growth increased approximately 15–25% and 10%, respectively, compared with the control ($p < 0.05$).

2—From 1974–1978, the increasing amounts of polluting emissions began to have a permanent negative impact on Scots pine radial growth. The greatest damages to forests were caused in 1980–1983 when 40–45% tree radial growth loss (compared to the control; $p < 0.05$) was observed in the closest forest stands and 15–20%—for the most distant from the pollution source stands has been established.

3—Radial growth of damaged Scots pine stands began to recover in 1990–1992 when the total annual amount of plant emissions decreased up to 11–12 kt and environmental pollution was considerably reduced.

4—The statistically significant negative impact (r ranges from –0.66 to –0.75; $p < 0.05$) of each pollutant, as well as the impact of the total amount of emissions on the radial increment of trees growing in areas closest (3–10 km) to the pollution source during the 1st stage of pollution was determined.

5—The rate of forest recovery was highest for the most damaged stands growing at the 3–10 km distance from the pollution source: their radial growth is stable and close to that of the control in 2001–2011. Less damaged stands growing further from the pollution source recovered earlier, and their radial growth was close to the control of 1995–1998.

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