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THE STATE OF THE FOREST ECOSYSTEM IN AN AREA OF OIL SHALE MINING AND PROCESSING

2. MORPHOLOGICAL CHARACTERISTICS OF NORWAY SPRUCE

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Air pollutants (oil shale fly ash, gases, organic compounds) emitted by the oil shale industry in Kohtla-Järve, North-East Estonia, have caused changes in the soil (pH = 4.7–7.4), subsoil water (pH = 5.9–7.2), rainwater (pH = 7.0–7.1) and snow melt (pH = 7.3–8.7) compared with an unpolluted control area in Lahemaa National Park (soil pH = 3.6, subsoil water pH = 5.1, rainwater pH = 6.9 and snow melt pH = 6.8). Compared with the period before 1990 the pollution load on the area investigated has fallen drastically; however, this has not resulted in an essential improvement in growth conditions of trees.

Morphological analysis of 80-year-old Norway spruces growing on sampling plots (six) in the polluted area and in the control area showed that air pollution has had temporally (1989–1990, 1994–1996) and spatially variable effect on the parameters characterising the state of trees: length growth, weight and dry matter content of needles and shoots, number and density of needles on shoots, radial increment of trees. The length growth of needles and shoots proved to be one of the most suitable parameters indicating the influence of air pollution, although not in all sample plots investigated. The results for fresh and dry weight of needles revealed great differences between sampling plots. The biomass of shoots was notably greater in the immediate vicinity of Kohtla-Järve than in the control area. The spruces whose shoots showed inhibited length growth had greater density of needles on shoots with difference from the control being up to 16 %. The effect on the radial increment of Norway spruces was especially strong in the immediate vicinity of pollution sources (<2 km) but it fell rapidly with distance from them.

Introduction

Air pollution problems at Kohtla-Järve date from the time when oil shale mining (1916) and its processing (1924) started in the area. The production was intensified and emissions increased notably since the 1950s when new oil shale fired power plants (Kohtla-Järve, Ahtme) were launched and several oil shale chemical enterprises were reconstructed or erected (oil shale chemical plants, later a plant producing nitrogen fertilisers, a shop for producing benzoic acid, etc.) [1]. From 1990 onward the amount of pollutants emitted into the atmosphere has fallen in almost all production units and a decrease in pollution can be observed in the towns of this area (Kohtla-Järve, Jõhvi) and in their surrounding territories.

Changes in the environment are reflected in the structure and state of vegetation. Conifers are regarded as sensitive to pollution. Air-borne pollutants have a strong effect on the life of trees, reducing their resistance to extreme temperatures and draught as well as to germs and pests [2]. Research by Karoles [3] shows that the state of Norway spruce crowns was the worst around the industrial centres in North-East Estonia and spruces near Kohtla-Järve have only two-year-old needles (in Estonia on average 5–8 years).

The aim of the present research was to estimate the state of Norway spruce, *Picea abies* (L.) Karst., which is one of the dominant tree species, in the region of oil shale mining and processing. Attention was focused on the analysis of the growth and development of spruce on the basis of morphological parameters (measurements and weight of needles and shoots, number and density of needles on shoots) and radial increment, which were treated together with data on the state of the growth environment (chemical composition of precipitation, soil and subsoil water).

Material and Methods

Sampling plots were established in the vicinity of Kohtla-Järve and Jõhvi, North-East Estonia, in 1995–1999. Morphological indices of Norway spruce, *Picea abies* (L.) Karst., and parameters of the environment affecting the growth of trees (pollution load; pH values and chemical composition of precipitation, soil and subsoil water) were determined in the course of comprehensive research. Changes caused by industrial load were compared in forest observation plots similar as to their natural conditions and in a control area not affected by industry (Lahemaa). A comparison of data obtained with earlier data allows drawing conclusions about the condition of trees after a sharp decline in emissions from industrial enterprises.

The study area is situated on the North-East Estonian plateau, where the main pollution sources are *Nitrofert* AS (production of nitrogen fertilisers), *Velsicol Eesti* AS (benzoic acid), Kiviõli, Kohtla-Järve and Ahtme power plants, *Viru Chemistry Group* AS (together with *Kiviõli* production unit), Ida-Viru Roads Board (asphalt concrete) and a number of smaller enterprises (boiler houses and enrichment plants of mines, a dairy, a furniture factory, etc.) (Fig. 1).

The sampling plots were established in different directions and at various distances from Kohtla-Järve with the control stands in a relatively unpolluted area of Lahemaa National Park (Table 1). The plots (spruce stands) were located 0.5 km north-east from a nitrogen fertiliser plant (Kohtla-Järve, plot *1*), 2 km north-east (Kohtla-Järve, plot *1a*), 7 km east (Kukruse, plot *2*), 15 km south-east (Kose, 3 km NE from Ahtme power plant, plot *3*), 6 km south (Sompa, plot *5*), 14 km south (Kalina, 0.5 km south from Viru enrichment plant and 6 km south-west from Ahtme power plant, plot *4*) and 6 km west (Aa, 6 km north-east from the Püssi plants of *Repo Plants* AS and 11 km north-east from Kiviõli power plant and oil shale chemistry plant, plot *6*).

In the selection of sample plots we proceeded from the principle of analogy of geographical and forestry characteristics. It was important that climatic and edaphic conditions as well as parameters of stand of the plots should be similar. The selected stands were 80-year old parts of (*Oxalis*–)*Myrtillus* site type forest of 0.7–0.8 density, quality class II with a medium-density or sparse understorey and no traces of sanitary felling. As in the Kohtla-Järve area no *Myrtillus* site type Norway spruce stands occur, the sampling plots in that area (*1* and *1a*) were established in *Aegopodium* site type stands, which are similar to the *Myrtillus* site type stands. At Kukruse (*2*) spruces growing in a Scots pine stand were used for this research.

Parameters characterising sample plots are presented in Table 1.

To find out changes in the environmental conditions of plots and to get a survey of the pollution load, we collected snow samples from the sample plots in Kohtla-Järve area and from the control area in 1996 when a permanent snow cover had formed. Rain, subsoil water and soil were collected during the vegetation period in 1996 and 1997. Chemical analyses of the samples were performed in the Central Laboratory of Environmental Research and in the Estonian Centre of Plant Material Control.

To characterise the state of trees in the area under study Norway spruces in the sample plots were evaluated with the help of parameters of morphometric evaluation system used in Central Europe [4]. The branches for analyses were collected from the southern side of the trees (10) in 1997. The length of 1- and 2-year old needles and shoots (cm), the fresh and dry weight of needles (g), the dry matter content of needles and shoots (%), density of needles on shoots (No. cm^{-1}) and thickness, expressed as the ratio of needle and shoot dry weight and their length (mg cm^{-1}) were determined and, if possible, the results were compared with earlier results (1989, 1990, 1994).

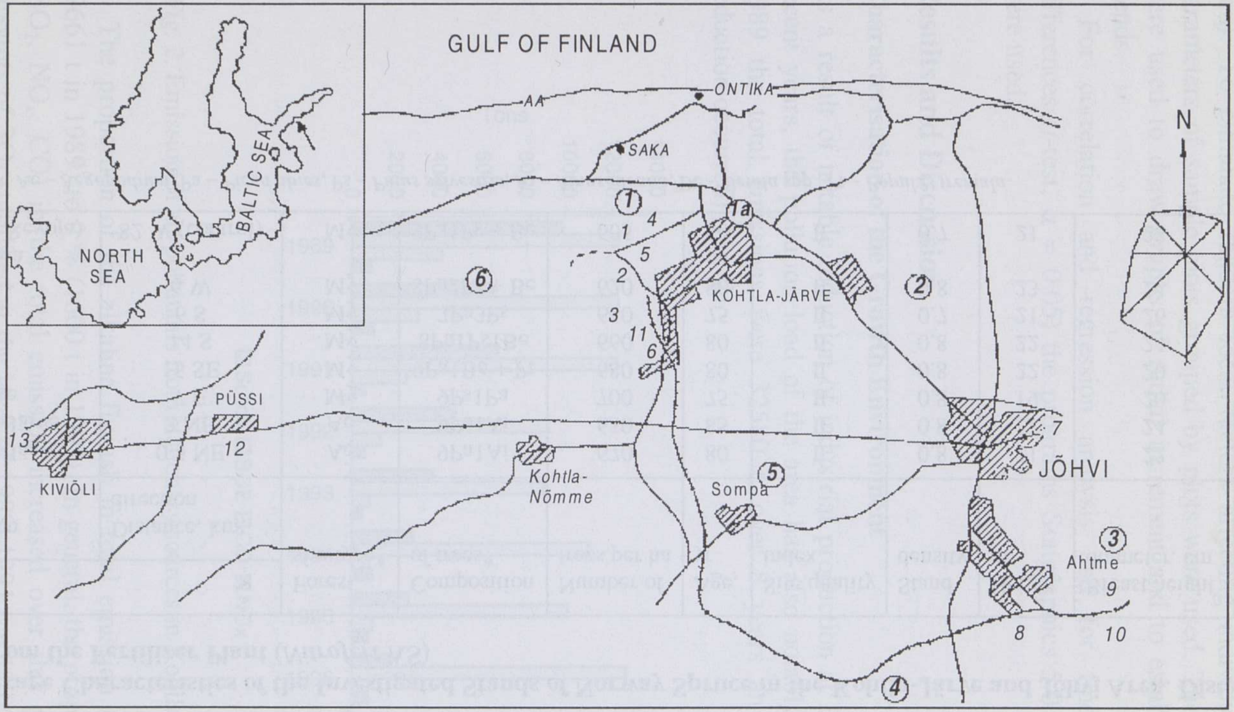

Fig. 1. The study area [6] with a wind rose. Location of sample plots (*1–6* in circles) and pollution sources: *1* – *Nitrofert* AS; *2* – Kohtla-Järve PP; *3* – *Viru Chemistry Group* AS; *4* – *Velsicol* AS; *5* – regional sewage purification plant; *6* – *Virko* AS (furniture production); *7* – *Dairy Plant* AS; *8* – Ida-Viru Roads Board (asphalt concrete); *9* – Ahtme Power Plant; *10* – *Silbet* AS; *11* – *Akte* AS; *12* – *Repo Plants* AS; *13* – *Viru Chemistry Group* AS (together with *Kiviõli* production unit)

Table 1. **Average Characteristics of the Investigated Stands of Norway Spruce in the Kohtla-Järve and Jõhvi Area. Distances Were Measured from the Fertilizer Plant (*Nitrofert* AS)**

Sample plot			Forest site type*	Composition of trees*	Number of trees per ha	Age, yr	Site quality index	Stand density	Height, m	Breast height diameter, cm	Density of understorey
No.	Location	Distance, km; direction									
1	Kohtla-Järve	0.5 NE	Ae	9Pa1Ai	670	80	II	0.8	21	22	Slight
1a	Kohtla-Järve	2 NE	Ae	9Pa1Ai	650	85	II	0.8	22	24	Moderate
2	Kukruse	7 E	My	9Ps1Pa	700	75	II	0.8	19	20	Slight
3	Kose	15 SE	My	9Pa1Be + Pt	680	80	II	0.8	22	20	Slight
4	Kalina	14 S	My	8Pa1Ps1Be	660	80	II	0.8	22	24	Moderate
5	Sompa	6 S	My	7Pa3Ps	640	75	II	0.7	21	26	Moderate
6	Aa	6 W	My	8Pa2Ps + Be	620	80	II	0.8	23	24	Moderate
7	Lahemaa (Eru, Revoja)	82 W (control)	My	9Pa1Ps + Be	605	80	II	0.7	21	28	Moderate

* My – *Myrtillus*, Ae – *Aegopodium*, Pa – *Picea abies*, Ps – *Pinus sylvestris*, Ai – *Alnus incana*, Be – *Betula* spp., Pt – *Populus tremula*.

To estimate the effect of the pollution complex on the radial increment on the trees increment cores were taken 1998 and 1999 at a height of 1.3 m from northern and southern sides of 15–20 dominant or co-dominant trees from all sample plots and from the control area and annual rings were measured (in mm). To eliminate random local factors affecting individual trees mean parameters of sample plots grouped by plots were used. The data obtained were used to draw graphs of radial increment and to establish increment trends.

For correlation and regression analysis and for finding statistical differences (*t*-test, $\alpha = 0.05$) the programs Statgraphics 5.0 and Excel 5.0 were used.

Results and Discussion

Characterisation of the Growth Environment

As a result of notable reduction of industrial production at Kohtla-Järve in recent years, the pollution load of the area has also notably decreased (in 1989 the total emissions were 32580 t, over 8 years an almost 2-fold reduction occurred) (Fig. 2).

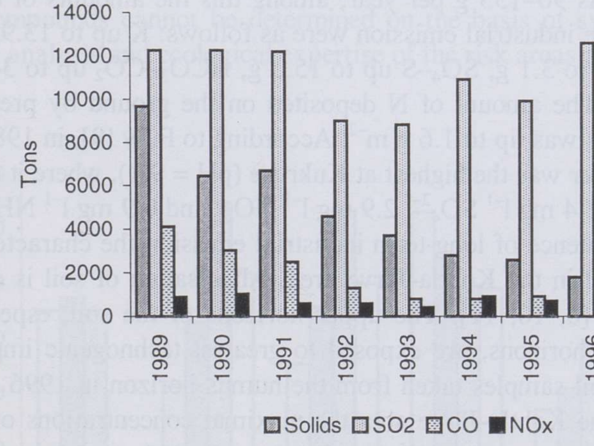

Fig. 2. Emission of air pollutants from stationary sources in Kohtla-Järve [27–34]

The proportion of the alkaline fly ash in total emission fell from 30 % (9661 t in 1989) to 9 % (1600 t in 1997). In general, the proportion of gases (SO_2, NO_x, CO) in the total emission increased over this period, but the amounts of SO_2 stabilised at the level of 1989–1991 (*ca* 12,000 t) in the last two years after a certain fall. No essential changes in the amount of NO_x were identified. Also large amounts of organic compounds, in which hydrocarbons (benzene, toluene, styrene), phenols, formaldehydes, etc. are represented, were emitted from the enterprises of Kohtla-Järve [5].

The pollution load in the influence zone of Kohtla-Järve and Jõhvi has a notable impact on the chemical composition of precipitation. The high electric conductivity of rainwater and snowmelt in the study area as compared with the control (in rainwater up to 2.4 and in snow water up to 2.6 times difference) indicates a larger amount of dissolved compounds in the precipitation of the industrial region of Kohtla-Järve [6]. As the concentration of sulfur compounds (SO_2, H_2S) in the air is still high, the rainwater falling on the ground and vegetation contains >14.2 mg l^{-1} of SO_4^{-2}. The rainwater in the study area has also elevated concentrations of Ca and K, elements that are present in the oil shale fly ash. The pH level of rainwater in the Kohtla-Järve area is somewhat higher (pH = 7.0–7.1) [6] and exceeds the level of normal for rainwater (5.6–6.6) [7].

The analysis of the snow samples collected in 1996 showed that the pH of snow in this area is higher than 7 (pH = 7.3–8.7), differing from the control (pH = 6.8) 1.1–1.3 times [6]. The concentrations of the elements dominating in the pollution complex (Ca, Mg, K, etc.) and nitrogen and sulfur compounds are also higher in the snow collected in the vicinity of Kohtla-Järve than in samples from Lahemaa (differences as maximum 2–4 times).

According to Petersell *et al.* [8], the amount of pollutants deposited in the Kohtla-Järve area per every square metre of ground surface by precipitation in 1992–1994 was 90–133 g per year; among this the amounts of the major constituents of the industrial emission were as follows: K up to 13.9 g, Ca up to 23.0 g, Mg up to 3.1 g, SO_4–S up to 15.0 g, HCO_3–CO_2 up to 34.0 g and Cl up to 5.0 g. The amount of N deposited on the ground by precipitation during these years was up to 1.6 g m^{-2}. According to Frey [9], in 1986 the pH level of snow water was the highest at Kukruse (pH = 9.1), where it contained 10 mg l^{-1} Ca^{2+}, 41.4 mg l^{-1} SO_4^{2-}, 2.9 mg l^{-1} NO_3^- and 0.9 mg l^{-1} NH_4^+.

Under the influence of long-term industrial emission the characteristics of soil have changed in the Kohtla-Järve area. Alkalisation of soil is especially clearly observed [6, 10, 11]. The upper horizons of the soil, especially the litter and humus horizons, are exposed to greatest technogenic impact. The analysis of the soil samples taken from the humus horizon in 1996 and 1997 showed that in the Kohtla-Järve area the maximal concentrations of Ca, Mg and K were 1533 ± 275, 127.6 ± 22.5 and 17.9 ± 5.8 mg 100 g^{-1}, respectively, being several times higher than in the control. Great differences from the control were observed in the total S content in soil (up to 4 times as high as the control). Also high values of electric conductivity of soil solutions indicate soil pollution in the study area [6].

Long-term deposition of fly ash has increased considerably the pH level of subsoil water in the Kohtla-Järve and Jõhvi area (pH = 5.9–7.2) as compared with Lahemaa (pH = 5.1) [6]. At Kose it is >2 times and in the other sampling areas 1.2–1.4 as high as the control. A steep rise in the concentration of compounds of the dominating elements of oil shale ash, Ca, K, Mg and

S, was observed in the samples from observation areas. The respective concentrations were up to 46.6 ± 7.8 (up to 4.1 times as high as the control), 24.4 ± 8.7 (5.3 times), 4.1 ± 0.4 (2.2 times) and 36.7 ± 26.6 mg 100 g^{-1} (3.9 times). In the subsoil water of the study area the contents of heavy metals occurring in the composition of oil shale (Mn, etc.) have fallen. It is well known that with the increasing pH of soil and subsoil water the mobility of heavy metals tends to be low; this was established also in the Kohtla-Järve area. The subsoil water of the study area showed elevated electric conductivity [6].

Morphology and Radial Increment of Trees

Environmental pollution with industrial emissions has great influence on the development of trees and on the formation of their biomass. The appearance of visible injuries in trees may be delayed for years [12]. Materna [13] is of the opinion that in the case of spruce, which is considered to be one of the most sensitive tree species towards sulfur dioxide [14], the appearance of symptoms of injury may take 4–6 years under unfavourable conditions and as long as 20 years under favourable conditions. When emissions stop, the injuries do not disappear immediately but remain until environmental conditions stabilise. The extent of chronic damage caused by low contents of sulfur compounds cannot be determined on the basis of symptoms; for this chemical analysis and ecological expertise of the risk areas are needed [15].

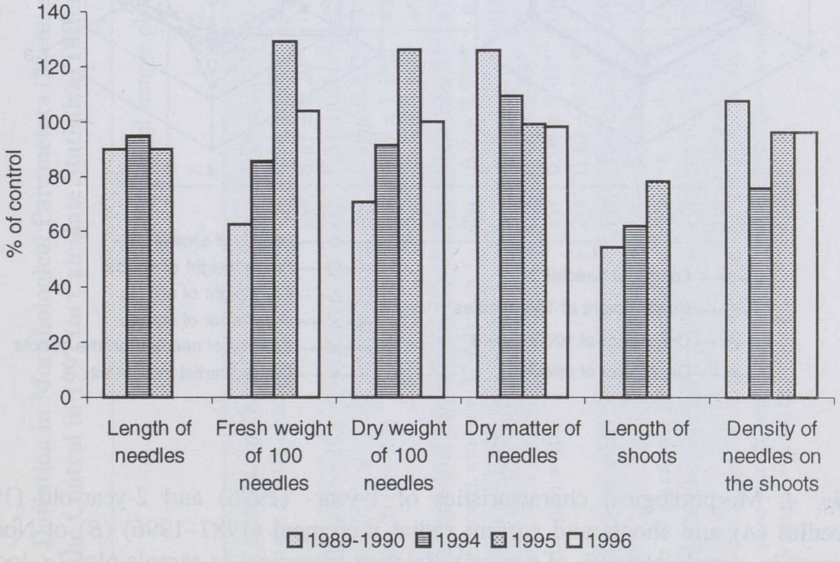

Fig. 3. Morphometrical characteristics of Norway spruce in northern areas at a distance of 0.5 km from Kohtla-Järve in different years (% of control) [16]

Length of needles. Earlier research has shown that during the period when the study area was characterised by high concentration of air pollutants, morphological parameters of trees were significantly affected [16]. When pollution loads are high, the length of needles usually diminishes while small loads may stimulate needle growth, especially in case the pollution complex contains nitrogen (NH_3, NO_x) or sulfur (SO_2) [17]. However, continuous and long-term air pollution represents a serious danger to the condition of forest massifs [13, 18]. The length growth of needles, which is one of the most widely used indicators of air pollution, was close to the control in the area of oil shale production and processing, but in the vicinity of Kohtla-Järve it differed notably from the control (Fig. 3; Table 2). In sample plots 1–5 the length of needles was 86–99 % of the length of the control needles; however, in sample plot 6 the length growth of needles formed in 1995 was higher than that of the control, making up 108 % of the control (Fig. 4; Table 2).

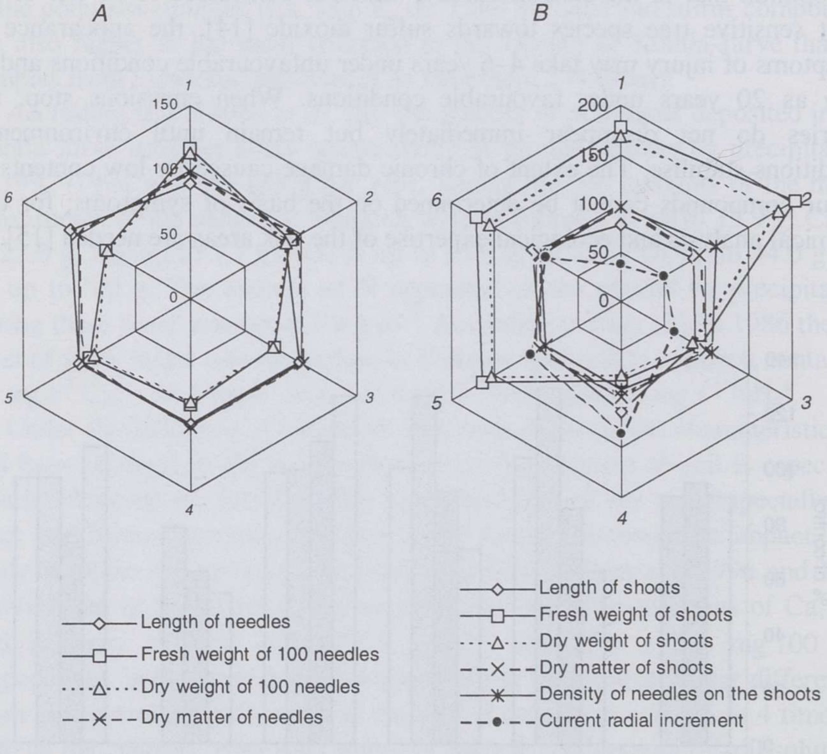

Fig. 4. Morphological characteristics of 1-year- (1996) and 2-year-old (1995) needles (*A*) and shoots and current radial increment (1987–1996) (*B*) of Norway spruce in sample plots (% of control). Relative increment in sample plot *1a*, located between plots *1* and 2, is shown in plot 2

Table 2. **Deviation in Morphological Parameters (Mean ± SD) of 60–80-Year-Old Norway Spruce in the Vicinity of Kohtla-Järve and at the Control in 1997 (to Calculate Statistical Differences the Two-sided *t*-test Was Used)**

	Age	No. of sample plot						
		1	*2*	*3*	*4*	*5*	*6*	*7* (control)
Length of needles, cm (*n* = 525-1125)	2yr	$1.29 \pm 0.21^{*3}$	$1.24 \pm 0.16^{*4}$	$1.40 \pm 0.19^{*2}$	$1.40 \pm 0.18^{*1}$	$1.43 \pm 0.19^{*1}$	$1.55 \pm 0.19^{*4}$	1.44 ± 0.22
Fresh weight of 100 needles, g (*n* = 7-10)	1yr	$0.71 \pm 0.23^{*1}$	$0.60 \pm 0.09^{*1}$	$0.51 \pm 0.08^{*3}$	$0.56 \pm 0.20^{*1}$	$0.66 \pm 0.15^{*1}$	$0.59 \pm 0.17^{*1}$	0.68 ± 0.14
	2yr	$0.81 \pm 0.20^{*1}$	$0.58 \pm 0.10^{*1}$	$0.48 \pm 0.13^{*2}$	$0.52 \pm 0.11^{*1}$	$0.71 \pm 0.27^{*1}$	$0.63 \pm 0.21^{*1}$	0.63 ± 0.18
Dry weight, g (*n* = 10)	1yr	$0.31 \pm 0.11^{*1}$	$0.26 \pm 0.04^{*1}$	$0.22 \pm 0.03^{*3}$	$0.24 \pm 0.06^{*2}$	$0.30 \pm 0.07^{*1}$	$0.25 \pm 0.07^{*1}$	0.31 ± 0.08
	2yr	$0.39 \pm 0.11^{*1}$	$0.27 \pm 0.04^{*1}$	$0.24 \pm 0.06^{*2}$	$0.26 \pm 0.06^{*1}$	$0.35 \pm 0.13^{*1}$	$0.30 \pm 0.10^{*1}$	0.31 ± 0.09
Dry matter, % (*n* = 10)	1yr	$43.9 \pm 1.18^{*1}$	$43.9 \pm 3.09^{*1}$	$44.9 \pm 7.42^{*1}$	$43.7 \pm 6.08^{*1}$	$45.2 \pm 1.90^{*1}$	$43.0 \pm 2.08^{*1}$	44.9 ± 2.56
	2yr	$48.3 \pm 2.19^{*1}$	$46.2 \pm 1.87^{*3}$	$49.7 \pm 1.75^{*1}$	$49.9 \pm 1.24^{*1}$	$49.0 \pm 0.70^{*1}$	$47.1 \pm 1.36^{*3}$	48.9 ± 1.29
Thickness of needle, mg cm^{-1}(*n* = 10)	2yr	$302.3 \pm 80.3^{*2}$	$217.7 \pm 121^{*1}$	$171.4 \pm 80.1^{*2}$	$185.7 \pm 60.5^{*1}$	$244.8 \pm 94.3^{*1}$	$193.6 \pm 112^{*1}$	215.3 ± 91.6
Length of shoots, cm (*n* = 32-64)	2yr	$6.2 \pm 0.55^{*4}$	$6.8 \pm 1.11^{*4}$	$7.0 \pm 1.89^{*2}$	$9.0 \pm 2.41^{*1}$	$7.5 \pm 1.49^{*1}$	$8.3 \pm 1.78^{*1}$	8.0 ± 2.32
Fresh weight of shoots, g (*n* = 10)	1yr	$0.15 \pm 0.05^{*2}$	$0.14 \pm 0.10^{*1}$	$0.09 \pm 0.06^{*1}$	$0.07 \pm 0.04^{*1}$	$0.14 \pm 0.09^{*1}$	$0.14 \pm 0.11^{*1}$	0.09 ± 0.05
	2yr	$0.18 \pm 0.09^{*2}$	$0.24 \pm 0.15^{*2}$	$0.08 \pm 0.07^{*1}$	$0.08 \pm 0.07^{*1}$	$0.16 \pm 0.08^{*1}$	$0.18 \pm 0.15^{*1}$	0.10 ± 0.06
Dry weight, g (*n* = 10)	1yr	$0.08 \pm 0.03^{*2}$	$0.07 \pm 0.04^{*1}$	$0.04 \pm 0.03^{*1}$	$0.04 \pm 0.01^{*1}$	$0.07 \pm 0.04^{*1}$	$0.07 \pm 0.05^{*1}$	0.05 ± 0.03
	2yr	$0.10 \pm 0.05^{*2}$	$0.13 \pm 0.07^{*2}$	$0.05 \pm 0.03^{*1}$	$0.05 \pm 0.03^{*1}$	$0.09 \pm 0.04^{*1}$	$0.10 \pm 0.07^{*1}$	0.06 ± 0.04
Dry matter, % (*n* = 10)	1yr	$50.8 \pm 2.8^{*1}$	$48.8 \pm 11.8^{*1}$	$51.0 \pm 7.8^{*1}$	$50.4 \pm 6.7^{*1}$	$52.4 \pm 6.4^{*1}$	$50.9 \pm 5.8^{*1}$	52.5 ± 4.3
	2yr	$57.3 \pm 3.5^{*1}$	$54.7 \pm 4.1^{*3}$	$60.0 \pm 8.1^{*1}$	$60.5 \pm 4.6^{*1}$	$56.0 \pm 3.5^{*1}$	$55.7 \pm 6.4^{*1}$	60.0 ± 2.0
Thickness of shoot, mg cm^{-1} (*n* = 10)	2yr	$16.1 \pm 1.12^{*4}$	$19.1 \pm 2.40^{*4}$	$7.1 \pm 0.18^{*1}$	$5.6 \pm 0.19^{*1}$	$12.0 \pm 1.12^{*1}$	$12.1 \pm 2.81^{*1}$	$7.5 \pm 1.08^{*1}$
Density of needles per cm (*n* = 10)	1yr	$15.6 \pm 2.5^{*1}$	$18.9 \pm 1.7^{*1}$	$17.6 \pm 1.7^{*1}$	$15.9 \pm 2.1^{*1}$	$14.4 \pm 3.6^{*1}$	$15.7 \pm 1.3^{*1}$	16.3 ± 4.1
	2yr	$15.0 \pm 1.7^{*1}$	$16.8 \pm 2.0^{*1}$	$16.0 \pm 1.6^{*1}$	$12.9 \pm 1.6^{*2}$	$13.8 \pm 3.6^{*1}$	$14.2 \pm 2.4^{*1}$	15.7 ± 3.5

Notes: 1yr – one-year-old needles and shoots formed in 1996; 2yr – two-year-old needles and shoots formed in 1995.
^{*1} p > 0.05; ^{*2} p ≤ 0.05; ^{*3} p ≤ 0.01; ^{*4} p ≤ 0.001.

Weight of needles. The analysis of needle weight collected from sample plots at different distances and directions around the town of Kohtla-Järve showed that, differently from earlier findings [16], in recent years the bioproduction of Norway spruces in the close vicinity of Kohtla-Järve has increased, being higher than the respective parameter in the control area. While in 1989–1990 and in 1994 the fresh weight of spruce needles collected near Kohtla-Järve (0.5 km NE) accounted respectively for 63 % and 86 % of the fresh weight of the control needles then the needles formed in 1995 were up to 29 % heavier than the needles from Lahemaa (Fig. 3; Table 2). Although the difference from the control was not so great in 1996 the fresh weight of needles in the near vicinity of Kohtla-Järve still surpassed the control. The difference between the fresh weight of needles formed in two consecutive years may have been caused by differences in weather and pollution loads (especially in SO_2).

Analogously to fresh weight the analysis of dry weight showed that in 1995 the dry weight of needles made up as much as 126 % of the control while no difference from the control was established in 1996. The fresh and dry weights of needles from sampling plots 2, 4, 5 and 6, situated at longer distances from Kohtla-Järve, were variable and as an average 83–113 % (1995) and 77–97 % (1996) of the control (Fig. 4). Both in 1995 and 1996 the fresh and dry weight of Norway spruce needles from sampling plot *3*, situated in the influence zone of Ahtme power plant were significantly lower than the control (fresh weight on average by 24 % and dry weight by 23–29 %) (Table 2). Needles from this sampling plot were longer but thinner than from other sampling plots.

Length and weight of shoots. Several studies have shown that changes in the characteristics of shoots are often one of the best indicators in investigating the impact of industrial emissions on conifers [16, 19, 20]. In the early 1990s high emissions from the industrial enterprises of Kohtla-Järve reduced significantly the length growth of shoots, which in a sampling plot 0.5 km to the north-east made up 54 % (1989–1990) and 62 % (1994) of the length of the control shoots (Fig. 3). In this area also the shoots formed in 1995 were by 22 % shorter than those from Lahemaa with differences between data obtained established at the level $p < 0.001$ (Fig. 4; Table 2). In sampling plots *2*, *3* and *5*, located farther from Kohtla-Järve, the length of the shoots formed in 1995 made up 85–94 % of the control, but in plots *4* and *6* stimulation of the length growth of shoots could be observed (respectively by 13 % and 4 % longer than from Lahemaa). In general the shoots from plots *3* and *4* were lighter than those from Lahemaa, in 1995–1996 their fresh and dry weights made up 78–100 % and 80–83 % of the control, respectively (Fig. 4; Table 2). The shoots from plots *1*, *2*, *5* and *6*, formed under the conditions of lower sulfur dioxide emissions, were 1.6–2.4 and 1.5–2.2 times heavier in fresh and dry weight in 1995 and a year later, 1.6–1.7 and 1.4–1.6 heavier than the control shoots from Lahemaa (Fig. 3). The shoots from these plots were also thicker than the control.

In general plants affected by air pollution are characterised by elevated contents of dry matter and disturbances in the water regime. In 1989–1990 the dry matter content in the area investigated was significantly higher than in the control area (Fig. 3). Together with the general fall in the pollution load, especially in the emission of solid pollutants, the dry matter content in needles has been decreasing from year to year. A significant difference in the dry matter content of needles and shoots from the control was observed only in a few cases (Fig. 4; Table 2).

Density of needles. Defoliation of trees is considerable due to air pollution. The defoliation of the crowns of conifers observed in heavily polluted industrial regions is caused by premature falling of needles as a result of which shoots are less densely covered with needles and the crown is thinned [21, 22]. The number of needles per 1 cm of shoot in its turn depends on the length of the shoot and thus the inhibited growth of shoots may be a reason why they are densely covered with needles.

According to forest monitoring data, the situation of the crowns of Norway spruces was in Estonia the worst in 1989 [23]; however, in the close vicinity of Kohtla-Järve (0.5 km NE) the spruce shoots were somewhat more densely covered with needles than the control. This may have been caused by the fact that the shoots in that region were nearly twice shorter than the control (Fig. 3). Karoles [24], based on forest monitoring results, observed since 1992 reduction in the defoliation of conifers, but in 1994, when the emission of gaseous pollutants SO_2 and NO_x from enterprises in Kohtla-Järve increased, the shoots of the trees growing at a distance of 0.5 km from this industrial town were by 24 % thinner than the shoots from Lahemaa (Fig. 3). On spruces whose shoot length growth was inhibited on plots *2* and *3* the needles were more densely located on shoots, with the respective indicator being by 7 % (1995) to 16 % (1996) higher than control (Fig. 4; Table 2). The shoots from other sampling plots were less densely covered with needles than the shoots from Lahemaa, making up 82–96 % (1995) and 88–98 % (1996) of the control. This indicates somewhat higher defoliation on those plots compared with Norway spruces from Lahemaa.

Radial increment. Air pollution impact is reflected also in the radial increment of trees (Fig. 4). Pollutants cause deviations in the metabolism of Norway spruce, which are reflected in changes in the bioproduction [25]. After a plant producing nitrogen fertilisers was launched in 1970 this impact has increased and it has caused decreasing vitality of spruces and their die-back (over half of the trees are dead) in the vicinity of the plant (0.5 km NE, plot *1*) [16]. Current radial increment in this area makes up only 37 % of the control and the state of the trees is continuously poor. At a distance of 2 km from the fertiliser plant (plot *1a*) the influence of air pollution is weaker and therefore the increment is greater (51 % of the control) and practically no dye-back of dominant trees was observed.

The situation is analogous in the area influenced by prevailing winds from Ahtme power plant (3 km NE from the plant, plot *3*), where the current radial increment is rather similar (62 % of the control). In more distant areas in the influence zones of Kohtla-Järve and Jõhvi industrial enterprises the effect of emissions is not strong enough to cause drastic changes in radial increment (plots 5 and 6, radial increment respectively 106 % and 89 % of the control). The increment on sample plot *4* is somewhat stimulated and differs significantly from the control (135 %); this can hardly be explained by air pollution alone. Results showed the clear trends in changes of radial increment in the studied area until 1996. To find out the reasons, additional studies of increment in this area are necessary.

The relationships between the current radial increment and the chemical composition of the needles and shoots of Norway spruce were complicated and non-linear. Regression analysis indicated significant polynomial dependence between the increment of spruces on sampling plots and the content of K and Ca in shoots and needles (for K $R^2 = 0.871$, for Ca $R^2 = 0.796$). These two are important nutrient elements with a significant effect on the increment of trees [26]. In the area investigated the content of K and Ca as well as their ratio in the shoots of Norway spruce were higher than in the control shoots [6]. The rise in the content of Ca was relatively much higher than in K; therefore we can expect that Ca had a stronger effect on increment in the area studied.

Conclusions

1. Our results show that changes in the length growth of needles and shoots are a suitable indicator of pollution emitted by the oil shale industry as these revealed a clear negative trend in sample plots closer to emission sources, although the total pollution load has fallen in recent years (in eight years since 1989 nearly two times). No clear trend was observed in the weight of needles and shoots (fresh and dry weight and dry matter content, which depends on these), the data are sometimes contradictory, requiring further research. As compared with earlier research (1989–1990 and 1994) the dry matter content of needles has fallen and a tendency towards improvement was observed in needle biomass, length growth of shoots and density of needles in the close vicinity of Kohtla-Järve (0.5 km from the nitrogen fertiliser plant).

2. Air pollution affects also radial increment of trees. The negative effect on the radial increment of Norway spruce is strong (radial increment more than 60 % less than of the control) in the vicinity of pollution sources

(<2 km); however, it decreases rapidly with increasing distance. Relationships between current radial increment and the chemical composition of needles and shoots were complicated and non-linear.

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