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IMPACT OF AIR POLLUTANTS EMITTED FROM THE OIL SHALE INDUSTRY ON CONIFERS

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Investigations on the emission quantities from chemical and power plants located in the town of Kohtla-Järve, and the results of atmospheric air quality calculations as well as monitoring data demonstrate that the role of acid deposition precursors among different air pollutants increased essentially during the period 1988-1995. Precipitation and soil analyses and morphometrical characteristics of Norway spruce and Scots pine on open land and in forest sites indicated serious changes in the morphology of conifers. A drastic aggravation of their state under the influence of the multicomponential air pollution complex was fixed in northern or northeastern areas at a distance of 2-3 km (Saka region) from Kohtla-Järve.

Introduction

The Estonian oil shale field is the largest commercially exploited oil shale deposit in the world. The Estonian oil shale deposit has been exploited since 1916 and its total yield tops 800 million tons of shale [1]. At present oil shale mining and processing are the most important industries in Estonia. However, these industries are responsible for serious problems of air pollution in North-East Estonia. Pollutants emitted from stationary sources of oil shale processing and chemical plants in Kohtla-Järve have a multicomponential chemical character as they consist of hazardous gaseous components, solid particles and aerosols [2, 3].

The influence of air pollution due to oil shale mining and processing on forest ecosystems is a topic which has economic and scientific implication, yet it is not completely understood and interpreted. Considering the disturbances in forest ecosystems in North-East Estonia [4–7] and lack of scientific consensus about them, the aim of the present work was to study a prolonged effect of an air pollution complex from oil shale industry rich in SO_2, NO_x, H_2S, fly ash, organic matter etc. on the forest ecosystem and on the morphological character of *Pinus sylvestris* L. and *Picea abies* L., the predominant conifers in Estonian forests.

Studied Area and Methods

The area located at a distance of up to 3 km to the north and north-east (to the direction of dominant winds) from the town of Kohtla-Järve (Fig. 1) - the centre of the Estonian oil shale industry in Ida-Viru County - suffers from serious environmental problems. Kohtla-Järve and its surroundings are affected by the emission of high amounts of multicomponential air pollutants (Table 1) due to intensive industrial activity and long-term influxes from a local oil shale processing plant (at present, RAS "Kiviter"), a mineral fertilizer plant (AS "Nitrofert"), benzoic acid production (AS "Velsicol Eesti") and Kohtla-Järve Thermal Power Plant (TPP).

Seven sample plots with 60-80-year-old Scots pine and Norway spruce trees located at different distances from the emission sources were chosen for the investigation of response reactions of conifers in both forest stands and in open areas (Fig. 1).

Table 1. **Emission of Pollutants into Atmosphere from Sources Determining* the Pollution Level in the Studied Area, tons yr^{-1} [12, 13]**

Pollutant	Year							
	1988	1989	1990	1991	1992	1993	1994	1995
Fly ash	36179	2318	1277	1464	1311	1073	498	337
SO_2	12951	10539	11179	10880	7939	8224	7931	7321
NO_x	1115	643	816	663	702	496	588	400
H_2S	-	27	19	18	16	13	11	16
NH_3	-	371	373	297	279	77	137	139
Organic**	-	-	7530	7454	8539	3377	2971	4116
Ratio of SO_2/fly ash	0.36	4.5	8.8	7.4	6.1	7.7	15.9	21.7

* In this Table the emissions from Kohtla-Järve oil shale processing plant (RAS "Kiviter"), mineral fertilizer plant (AS "Nitrofert"), Kohtla-Järve TPP and AS "Velsicol Eesti" were taken into account.

** Among this about 60 % of aliphatic and 30 % of aromatic hydrocarbons.

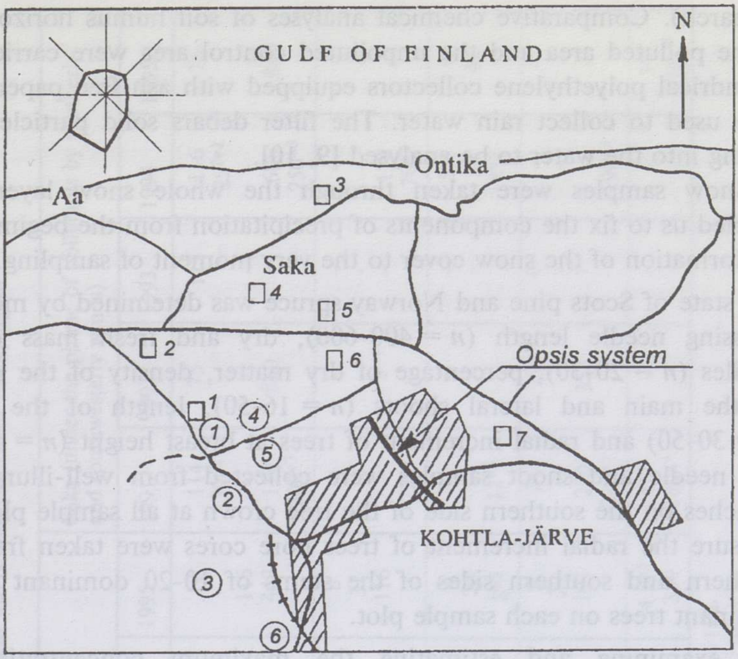

Fig. 1. Studied area.

Pollution sources (circles): *1* - AS "Nitrofert"; *2* - Kohtla-Järve TPP; *3* - RAS "Kiviter"; *4* - AS "Velsicol"; *5* - regional sewage purification plant; *6* - AS "Virko" (furniture production).

Observation points (squares): *1* - 0.2 km from AS "Nitrofert" (open land); *2* - 1.5 km from AS "Nitrofert" (open land); *3* - 3 km from AS "Nitrofert" (open land); *4* - 2 km from AS "Nitrofert" (open land); *5* - 2 km from AS "Nitrofert" (forest); *6* - 0.5 km from AS "Nitrofert" (forest); *7* - 2 km from AS "Nitrofert" (forest)

Three control sample plots were situated in a relatively unpolluted area in Lahemaa National Park, 78-84 km to the west from Kohtla-Järve. In order to be able to make certain comparisons of results received we selected sample plots having edaphic, meteorological and structural similarities.

In our integrated studies of the forest state some methods and parameters recommended by ICP on Integrated Monitoring on Air Pollution Effects [8] were used:

1. Physical and chemical properties of growth conditions were investigated to get information on deviations in the growth of trees. The accumulation of the dominant components of fly ash (SO_4^{2-}, NO_3^-, Ca^{2+}, K^+, Mg^{2+}, etc.) and gaseous ingredients (SO_2, NO_x, etc.) and the pH of the soil and precipitation were determined (the Estonian Centre of Agricultural Chemistry and Central Laboratory of

Research). Comparative chemical analyses of soil humus horizon (A_1) in the polluted area and the unpolluted control area were carried out. Cylindrical polyethylene collectors equipped with ash-free paper filters were used to collect rain water. The filter debars solid particles from getting into the water to be analysed [9, 10].

Snow samples were taken through the whole snow layer. This enabled us to fix the components of precipitation from the beginning of the formation of the snow cover to the very moment of sampling.

2. The state of Scots pine and Norway spruce was detemined by means of assessing needle length (n = 400-600), dry and fresh mass of 100 needles (n = 20-30), percentage of dry matter, density of the needles on the main and lateral shoots (n = 16-50), length of the shoots (n = 30-50) and radial increment of trees at breast height (n = 10-20). The needle and shoot samples were collected from well-illuminated branches on the southern side of the tree crown at all sample plots. To measure the radial increment of trees bore cores were taken from the northern and southern sides of the stems of 10-20 dominant or co-dominant trees on each sample plot.

For examining and estimating the maximum concentrations of pollutants in the atmospheric air the numerical semi-empirical computer-based modelling method OND-86 founded on Gaussian theory and elaborated by Main Geophysical Observatory (St. Petersburg, Russia), also the modified variant of those methods for the calculation of long-time mean concentrations has been used [3]. To study the factual air pollution level the data obtained by an automatic monitoring station "Opsis" located in the town of Kohtla-Järve at a distance of 2-3 km from the studied area (Fig. 1), and also the results of air sample analyses carried out by the Environmental Research Laboratory of Virumaa and the Sanitary Laboratory of RAS "Kiviter" using standardized methods were used [11].

Results and Discussion

1. Character of Air Pollution Complexes Affecting Ecosystems

The air pollution level in the studied area is continuously high because this region is situated at a distance of only 2-3 km from the above-mentioned chemical and power plants, in the leeward direction of the southerly (about 15 %) and south-westerly (up to 30 %) winds. Very light southerly winds (0.2-0.6 m s^{-1}) bring about a most hazardous situation and the inversion phenomenon, at which the concentrations of pollutants in the aboveground air layer rise twofold and more as compared with normal atmospheric conditions [3].

Table 2. **Concentrations of Pollutants in the Air of the Town of Kohtla-Järve and the Saka Area, μg m^{-3}**

Pollutants	Concentrations	Kohtla-Järve*1					Saka (under the plume of pollutants by southerly and southwesterly winds)				
		1991	1992	1993	1994	1995	1991	1992	1993	1994	1995
SO_2	Annual mean	19	16	17	12	12	114 30*2	75	117	116 252*2	76
	24-hour max.	108	106	113	73	116	-	-	-	-	-
	0.5-hour max.	442	342	348	271 250*3	443	110 300*3	860	-	600 250*3	840
NO_2	Annual mean	10	10	9.7	11	8	-	-	-	-	-
	24-hour max.	62	27	35	29	31	-	-	-	-	-
	0.5-hour max.	118	85 70*3	84	87 65*3	116	- 72*3	-	-	110 70*3	-
H_2S	Annual mean	-	-	1	1.6	2	-	-	-	5.6	9
	0.5-hour max.	-	-	13	46 7.2*3	20 8*3	- 12*3	-	-	38 8*3	48
NH_3	Annual mean	-	-	14	19	9	24	29	0	-	-
	24-hour max.	-	-	-	-	27	-	-	-	-	-
	0.5-hour max.	- 200*3	-	170	380 190*3	121	270 220*3	350	7	- 190*3	-
Fly ash	Annual mean	5*3	-	-	-	4.5*3	4*3	-	-	-	3*3
	0.5-hour max.	400*3	-	-	-	350	180*3	-	-	-	160*3

*1 For sulphur dioxide, nitrogen dioxide and ammonia the data by Opsis-system are given [13].
*2 The calculated annual mean by all wind directions: in 1991, 25-30 and in 1994, 18-25 μg m^{-3}.
*3 The calculated data.

Compared to 1988-1989, the pollution situation changed essentially (Tables 1 and 2) during the period of 1991-1995. Until the installation of the precipitators (in 1989) the amounts of oil shale fly ash emitted from Kohtla-Järve TPP were up to 30 times higher (up to 50 thousand t yr^{-1}) than at present. As a result the town of Kohtla-Järve and the areas up to Saka-Ontika were most heavily polluted with alkaline dust: the daily ash deposition intensity, according to snow-samples, was up to 0.4-1 g m^{-2} and near the TPP up to 8.4 g m^{-2} [14, 15] and the calculated half-hour maximum concentrations (C_{max}) of dust in the overground air layer in the Saka region were in the interval of 1500-4500 μg m^{-3}. Table 2 shows that in 1991-1995 the C_{max} values for solid particles (dust) did not exceed 160-400 μg m $^{-3}$.

However, the emissions of sulphur dioxide into the atmosphere from the sources affecting the studied area have diminished by only about 40 % during the period of 1988-1995. As a result the ratio of the emitted amounts of SO_2 /fly ash rose from 0.36 in 1988 to 15.9-21.7 in 1994-1995 (Table 1). In the atmosphere of the Kohtla-Järve region 44 % of the pollution level of SO_2 is due to the emissions from the boiler houses of RAS "Kiviter" and 38 % Kohtla-Järve TPP [15].

At the 5th International Conference on Acidic Deposition the following critical loads established (by the United Nations Economic Commission for Europe) for the atmospheric air in respect to forest trees were mentioned [16]:

- For SO_2 20 μg m^{-3} as annual mean or half-year mean (October to March) for winter conditions and 10 μg m^{-3} as annual mean for sensitive lichen (cyanobacterial) species
- For NO_x (as NO_2 equivalent) 30 μg m^{-3} as annual mean and 95 μg m^{-3} as 4 hour mean (it is assumed that SO_2 load is close to its critical level)
- For ammonia: annual mean - 8, monthly mean - 23, daily mean - 270 and 1-hour maximum 3300 μg m^{-3}

Table 2 shows that during the period of 1991-1995 the annual mean concentration of sulphur dioxide in the Kohtla-Järve region was within the range of 12-19 μg m^{-3}, which is near the critical level for forest trees. In the studied area affected by S and SW winds the annual mean values of SO_2 between 75-117 μg m^{-3} were determined. Short-term concentrations may be very high: the daily maximums - 73-116, half-hour maximums - 271-443 and immediately under the plume of pollutants from emission sources - up to 860 μg m^{-3}.

The relatively high mean concentrations (30-80 μg m^{-3}) of SO_2 occur in the Kohtla-Järve area during the periods of easterly winds (about 8 %) when for the northernmost part of NE Estonia (the coast of the Gulf of Finland) the level of 25 μg m^{-3} is exceeded [17].

The concentrations of NO_2 and ammonia are lower than the above-mentioned critical loads, but the simultaneous presence of those pollutants and SO_2 will increase the effect, so that SO_2 and NO_2 have a synergetic effect. It should be mentioned that in case of nitrogen deficiency nitrogen will act as a nutrient for forest ecosystems. This complicates the situation with respect to nitrogen (NO_x, NH_3). However, the uptake of nitrogen in the stemwood and bark will contribute to acidification [18]. The presence of hydrogen sulphide (H_2S) in the air will increase the influence of SO_2.

Relatively high concentrations of pollutants were registered in the air of Saka-Ontika area in the spring (March-May) of 1994 in case of SW winds (at wind speed 1-6 m s^{-1}), μg m^{-3}: SO_2 - 60-523; NO_2 - 30-110; H_2S - 2-18 and NH_3 - 30-290 [13]. In case of weaker winds (0.2-0.6 m s^{-1}) the air pollution level was about 2-2.5 times higher, as a rule. Moreower, when the winds are blowing from RAS "Kiviter" towards the Saka region (southerly winds) the plums of dicsharges from different chemical industries are summarized. As result, the total concentrations of SO_2, NO_x, H_2S, ammonia and organic contaminants, such as formaldehyde, phenol, aromatic and aliphatic hydrocarbons etc., may be up to 1500-2000 μg m^{-3}, under the conditions of very light winds and the inversion phenomenon.

Thus, the pollution situation in the studied area is very complicated. After the installation of electric precipitators in the Kohtla-Järve TPP (in 1989) the role of acid deposition precursors among the air pollutants has essentially increased. This conforms to the results of snow and rainwater analysis, which showed that the role of SO_4^{2-} ion had increased while pH had decreased correspondingly (see below).

2. Peculiarities of the Growth Condition of Trees on Sample Plots

To estimate the state of trees and to interprete the results received from the zones affected by air pollution it is necessary to estimate the changes in the growth conditions (soil, precipitation) of plants on the observation plots. Rain and snow investigations enable an approximate estimation of the pollution load upon the ecosystems.

Precipitation. High air pollution load in the Kohtla-Järve area is reflected in the chemical character of rainfall and snow. A comparison with the data from our control area in Lahemaa and Smidt scale [19] shows the summer precipitation in the studied area to have been moderately alkaline (annual average pH 6.8-7.0) during the period from 1990 to 1994. Earlier, from 1984 to 1989, the maximum average pH of rain water registered was 7.5 in 1985 and 1987 [20]. According to the scale of Smidt [19], the rainwater of this period was strongly basic with pH >7.10.

During the period of our investigations (1990-1994) the concentration of the sulphate ions exceeded that in the control area by 90 % and nitrate ions by 60 %. Also the content of elements occurring in fly ash was high: Ca^{2+} by 28 %, Mg^{2+} by 34 % and K^+ by 11 % higher than the control (Table 3). High sulphate contents of rainwater in Estonia were already noted in 1988 by Frey et al. [21]. According to the information of the Estonian Meteorological and Hydrological Institute in 1987-1989 air pollution with sulphuric compounds was the highest in North-East Estonia reaching in some areas 30-46 mg l^{-1} [22].

Table 3. **Chemical Characteristics of Rainwater in the Vicinity of Kohtla-Järve and in the Control Area (Lahemaa)**

Distance and direction from emission source	pH	Electric conductivity, mS cm^{-1}	Content of ions, mg l^{-1}					
			Ca^{2+}	Mg^{2+}	K^+	Na^+	SO_4^{2-}	NO_3^-
Average of 1990-94								
78-84 km W	6.4	0.09	9.4	4.1	2.5	3.0	10.8	0.7
3 km N	6.9	0.13	14.3	6.5	3.0	2.9	19.4	1.1
2 km NE	6.9	0.13	10.5	5.7	2.9	2.6	21.4	1.1
1.5 km N	7.0	0.20	12.0	5.1	2.7	3.2	20.3	1.7
0.5 km N	6.8	0.17	11.0	4.9	2.5	1.6	22.1	1.2
Average of 1984-89 [20]								
80 km W	6.6	0.10	9.3	5.1	3.0		17	
0.5 km SE	7.2	0.27	25.8	7.0	7.8		34	
3 km N	7.0	0.15	15.8	5.6	3.4		30.1	

The hydrochemical analyses of snow cover showed enrichment of environment with components of fly ash and gaseous pollutants in the area north from Kohtla-Järve. Compared with data collected in 1987, the average pH of snow melt in this area had decreased by 1.5 units in 1994. This indicates a decrease of alkaline components in the air pollution complex in recent years. High concentrations of NH_4^+ and SO_4^{2-} were also characteristic of snow melt in 1994 (Table 4).

Soil. Soil is a receptor of dry and wet deposits from air pollution. Analyses carried out showed higher concentrations of N and S as well as elements prevailing in the fly ash in the polluted areas. Studies in 1994 suggested a decreasing tendency of pH_{KCl} in A_1 horizons compared to previous years (Fig. 2); however, it is still about 2-3 units higher than in the control sample plots in Lahemaa. The soil parameters show very clearly the effect of atmospheric liming due to the industrial activity. Concentrations of CaO in the forest soil A_1 horizons at a distance of 0.5-3 km N from Kohtla-Järve were estimated at 1500 mg 100 g^{-1} and in Lahemaa at 84 mg 100 g^{-1}, the contents of MgO were respectively 160 and 10, K_2O 80 and 25 mg 100 g^{-1}.

Table 4. **Characteristics of Snow Melt at Different Distances from Kohtla-Järve in 1994**

Distance and direction from Kohtla-Järve	pH	Electric conductivity, mS cm^{-1}	Content of ions, mg l^{-1}							
			Ca^{2+}	Mg^{2+}	K^+	Na^+	NH_4^+	NO_3^-	SO_4^{2-}	Cl^-
78–84 km W	7.1	36	3.85	0.99	0.3	0.6	0.21	2.0	3.5	2.3
3 km N	6.8	58	7.17	1.48	0.6	0.6	0.77	1.7	12.5	2.5
2 km NE	7.1	60	8.58	1.39	0.3	0.6	0.67	1.3	13.0	2.6
1.5 km N	6.8	53	5.69	1.17	0.8	0.6	0.57	1.0	4.4	1.9
0.5 km N	7.0	77	9.70	1.51	0.4	0.6	1.46	2.5	19.8	2.9
0.5 km NE	6.9	54	7.90	0.61	0.3	0.5	0.57	1.5	11.1	1.9

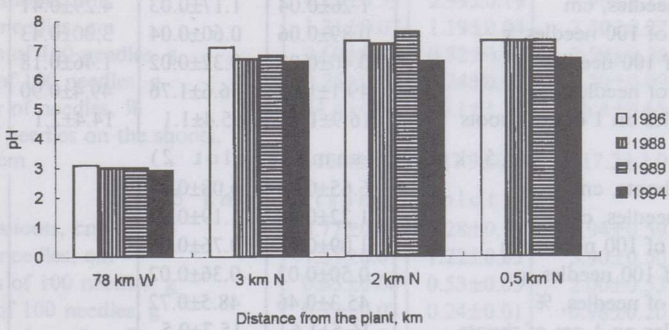

Fig. 2. Differences in pH of A_1 soil horizons of sample plots on the control and polluted areas

The concentrations of N, and especially S, in A_1 horizons of the soil in the polluted sample plots were estimated to be somewhat higher than in the control area. The value of N/S was lower than in the control area at all distances from the emission centre studied: 57 times at a distance of 0.5 km N, 29 times 1.5 km N and 15 times 3 km N.

The differences in the cation and anion accumulation in soil humus horizons in the vicinity of Kohtla-Järve might be explained by the intensive aerotechnogenic flow from the industry in this area.

3. Changes in the Morphology of Conifers

Morphological and structural alterations as well as deviations from normal growth and bioproduction are secondary consequences of functional changes in plants affected by several stressors. The sensitivity of conifers to air pollutants is generally known and reduction in the length of needles and suppressed increment of trees have been recorded by many authors [23-27].

Table 5. **Average Morphometrical Characteristics of Norway Spruce and Scots Pine on Open Land (1989, 1990, 1994)**

Distance and direction of sample plots from emission centre, parameters	Norway spruce		Scots pine	
	Main shoots	Lateral shoots	Main shoots	Lateral shoots
78 km W (control area)				
Length of shoots, cm	8.12±0.60	7.61±0.19	14.90±1.34	13.66±0.49
Length of needles, cm	1.32±0.04	1.37±0.07	4.90±0.14	4.52±0.08
Fresh mass of 100 needles, g	1.37±0.05	1.03±0.03	4.62±0.28	4.66±0.29
Dry mass of 100 needles, g	0.59±0.04	0.43±0.01	2.09±0.19	2.07±0.13
Dry matter of needles, %	41.7±0.65	41.4±0.32	46.9±0.52	44.5±0.44
No of needles on 1 cm of shoots	16.1±0.5	14.0±0.16	10.6±0.91	10.6±1.10
0.2 km N (sample plot *1*)				
Length of shoots, cm	4.78±0.66	3.72±0.36	7.28±1.54	3.71±0.51
Length of needles, cm	1.26±0.04	1.17±0.03	4.29±0.41	3.63±0.19
Fresh mass of 100 needles, g	0.89±0.06	0.60±0.04	3.00±0.43	2.41±0.25
Dry mass of 100 needles, g	0.40±0.03	0.32±0.02	1.46±0.18	1.07±0.07
Dry matter of needles, %	49.1±1.02	56.6±1.76	49.4±0.90	48.2±0.67
No of needles on 1 cm of shoots	16.9±1.7	15.4±1.1	14.4±2.1	13.8±1.5
1.5 km N (sample plot *2*)				
Length of shoots, cm	6.65±0.86	4.08±0.23		
Length of needles, cm	1.22±0.08	1.19±0.03		
Fresh mass of 100 needles, g	1.09±0.05	0.76±0.04		
Dry mass of 100 needles, g	0.50±0.02	0.36±0.02		
Dry matter of needles, %	45.3±0.46	48.5±0.72		
No of needles on 1 cm of shoots	16.5±1.6	15.7±0.5		
2 km N (sample plot *4*)				
Length of shoots, cm	4.34±0.75	4.08±0.28		
Length of needles, cm	1.43±0.12	1.22±0.05		
Fresh mass of 100 needles, g	0.58±0.08	0.57±0.06		
Dry mass of 100 needles, g	0.28±0.04	0.26±0.03		
Dry matter of needles, %	47.4±0.90	46.5±0.69		
No of needles on 1 cm of shoots	14.5±1.4	12.2±0.5		
3 km N (saple plot *3*)				
Length of shoots, cm	5.80±0.76	4.28±0.24	5.37±0.65	3.29±0.20
Length of needles, cm	1.23±0.05	1.29±0.03	4.80±0.21	4.69±0.09
Fresh mass of 100 needles, g	0.98±0.05	0.78±0.04	3.75±0.20	3.09±0.11
Dry mass of 100 needles, g	0.45±0.02	0.36±0.02	1.78±0.07	1.43±0.05
Dry matter of needles, %	46.1±1.00	46.4±0.61	47.5±0.63	46.5±0.48
No of needles on 1 cm of shoots	17.1±1.5	14.9±0.65	12.7±1.3	10.9±0.8

However, there is relatively little information about changes in the morphology of conifers caused by air pollution from oil shale industry in Estonia. In general, the visual observations of forest state by the Forest Protection Service show the most severely damaged forests to be located in North-East Estonia, especially in the vicinity of Kohtla-Järve, including the area studied by us [4–7, 28].

Table 6. **Morphometrical Characteristics of Norway Spruce and Scots Pine in Forest Sites in 1994**

Distance and direction of sample plots from emission centre, parameters	Norway spruce		Scots pine	
	Main shoots	Lateral shoots	Main shoots	Lateral shoots
78 km W (control area)				
Length of shoots, cm	8.39±0.82	6.29±0.34	17.38±3.13	8.55±0.63
Length of needles, cm	1.25±0.06	1.38±0.11	5.33±0.18	4.80±0.12
Fresh mass of 100 needles, g	0.73±0.03	0.65±0.03	4.30±0.30	3.87±0.33
Dry mass of 100 needles, g	0.31±0.01	0.27±0.01	1.88±0.13	1.71±0.14
Dry matter of needles, %	41.1±0.51	41.0±0.33	43.9±0.51	44.2±0.83
Density of needles on the shoots, No per cm	16.3±0.1	17.1±0.5	14.4±1.6	15.2±0.8
2 km N (sample plot *5*)				
Length of shoots, cm	4.22±0.59	2.99±0.19	7.96±1.13	4.46±0.83
Length of needles, cm	1.31±0.07	1.19±0.04	3.50±0.27	2.96±0.12
Fresh mass of 100 needles, g	0.60±0.08	0.52±0.03	2.59±0.20	1.75±0.10
Dry mass of 100 needles, g	0.26±0.04	0.24±0.02	1.20±0.09	0.82±0.05
Dry matter of needles, %	44.4±0.65	46.1±1.19	46.4±0.65	47.6±1.12
Density of needles on the shoots, No per cm	13.4±1.2	13.9±0.5	17.2±2.0	22.2±1.5
0.5 km NE (sample plot *6*)				
Length of shoots, cm	4.77±0.44	4.28±0.19	3.94±0.59	3.14±0.25
Length of needles, cm	1.35±0.07	1.22±0.03	3.90±0.50	3.85±0.18
Fresh mass of 100 needles, g	0.65±0.06	0.53±0.03	2.00±0.53	1.96±0.14
Dry mass of 100 needles, g	0.29±0.03	0.24±0.01	0.98±0.20	0.90±0.06
Dry matter of needles, %	45.1±0.75	44.7±0.34	46.2±2.17	45.7±0.64
Density of needles on the shoots, No per cm	12.0±0.9	13.2±0.4	17.2±4.6	16.7±1.7

Detailed investigations of the morphology and increment of Norway spruce and Scots pine growing in both forest sites and in open land showed a drastic aggravation in their state under the influence of the polycomponental air pollution complex in northern or northeastern areas at distances to 3 km from Kohtla-Järve (Tables 5 and 6).

Reduced increment of the needles accompanied with a decrease in their mass is most notable in case of trees growing in forest sites and on lateral shoots. Compared with needles, the growth of shoots seems to be more sensitive to air pollution. A comparison of the sensitivity of spruce and pine to the air pollution impact in the studied area shows that no essential differences exist in the depression of the growth of their shoots under high pollution loads.

The growth of the shoots seems to be an essential factor determining the development of needles. Regression trendlines and *R*-squared values showed a strong relationship between the length of shoots and the parameters of needles (Fig. 3).

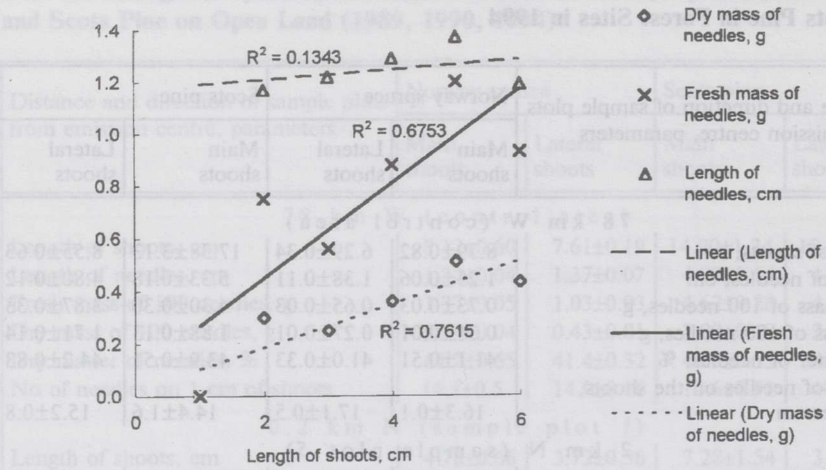

Fig. 3. Relationship between the length of shoots and needle development of Norway spruce. Regression trendlines and *R* -squared values were calculated using MS EXCEL 5.0

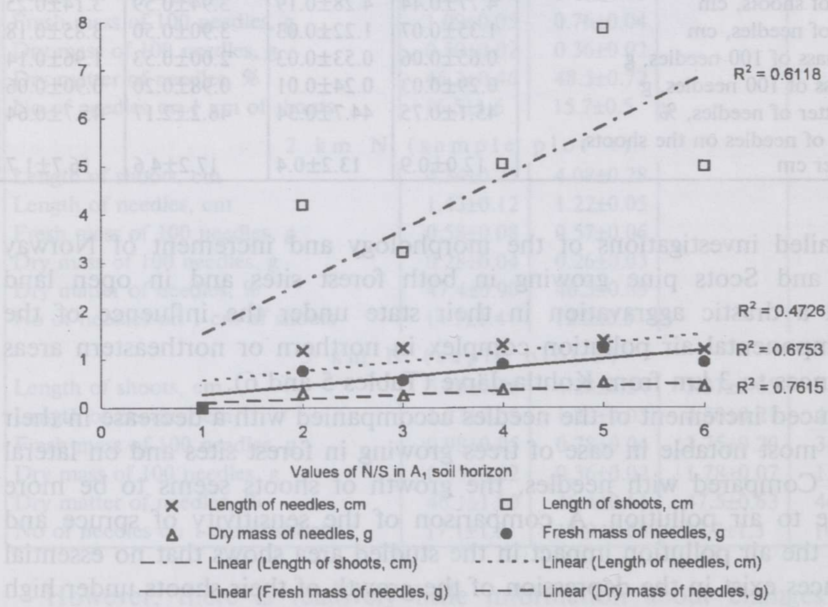

Fig. 4. Relationship between the ratio N/S in soil and parameters of Norway spruce needles and shoots. Regression trendlines and *R* -squared values were calculated using MS EXCEL 5.0

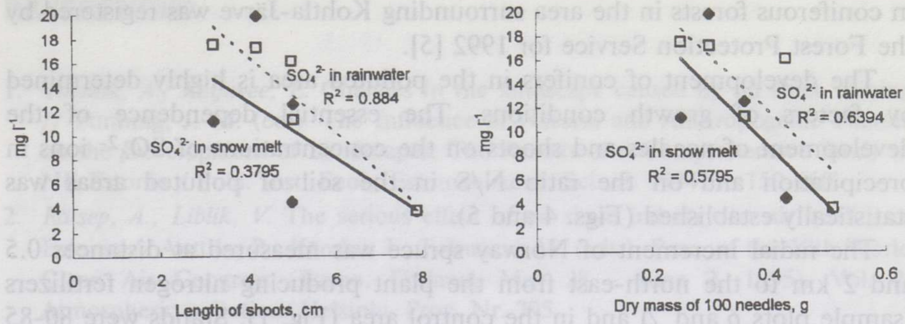

Fig. 5. Dependence of the length of shoots and dry mass of 100 needles of Norway spruce in studied areas. Regression trend-lines and *R*-squared values were calculated using MS EXCEL 5.0

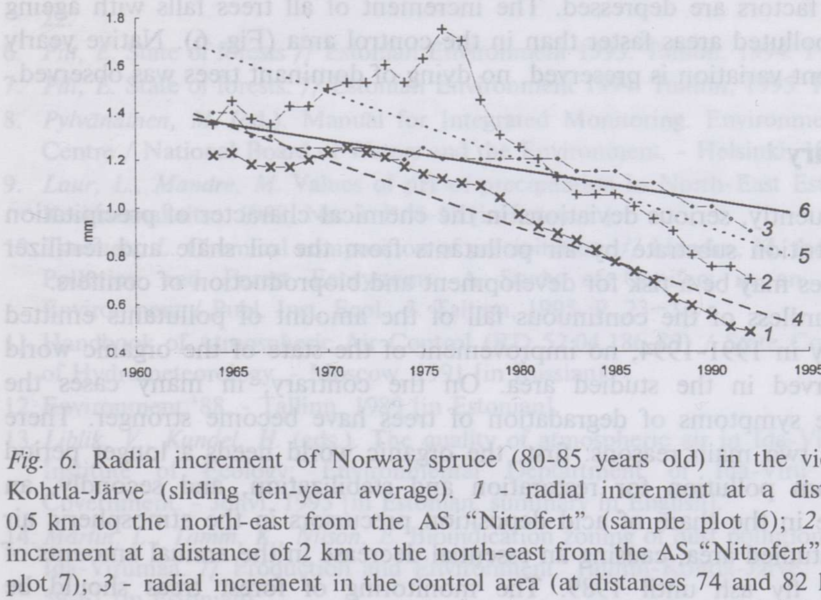

Fig. 6. Radial increment of Norway spruce (80-85 years old) in the vicinity of Kohtla-Järve (sliding ten-year average). *1* - radial increment at a distance of 0.5 km to the north-east from the AS "Nitrofert" (sample plot 6); *2* - radial increment at a distance of 2 km to the north-east from the AS "Nitrofert" (sample plot 7); *3* - radial increment in the control area (at distances 74 and 82 km from Kohtla-Järve); *4* - trend of the radial increment at a distance of 0.5 km to the north-east from the AS "Nitrofert" (sample plot 6); *5* - trend of the radial increment at a distance of 2 km to the north-east from from the AS "Nitrofert" (sample plot 7); *6* - trend of the radial increment in the control area

The number of needles on shoots is important as regards the photosynthetic potential of the trees and it is a suitable parameter for assessing forest vitality. Because of the rapid decrease of shoots in length (Scots pine to about 60 %, Norway spruce 43 %) the number of needles counted per cm of shoots may be higher than that of the control trees (Tables 5 and 6). Predominantly only needles on current-year and on one-year-old shoots have been found. In general, a 26-60 % defoliation degree in coniferous forests in the area surrounding Kohtla-Järve was registered by the Forest Protection Service for 1992 [5].

The development of conifers in the polluted area is highly determined by factors of growth conditions. The essential dependence of the development of needles and shoots on the concentrations of SO_4^{2-} ions in precipitation and on the ratio N/S in the soil of polluted areas was statistically established (Figs. 4 and 5).

The radial increment of Norway spruce was measured at distances 0.5 and 2 km to the north-east from the plant producing nitrogen fertilizers (sample plots *6* and *7*) and in the control area (Fig. 1). Stands were 80-85 years old with quality index II and density 0.7. It appeared that after the launching of the fertilizer plant in 1970 the pollution effect has increased and has caused a steep decrease in the vitality of spruces. Current annual radial increment constitutes only 30 % of the control, differences of increments between the successive years are small and reactions to the natural factors are depressed. The increment of all trees falls with ageing but in polluted areas faster than in the control area (Fig. 6). Native yearly increment variation is preserved, no dying of dominant trees was observed.

Summary

Consequently, serious deviations in the chemical character of precipitation and nutrition substrate by air pollutants from the oil shale and fertilizer industries may be a risk for development and bioproduction of conifers.

Regardless of the continuous fall of the amount of pollutants emitted annually in 1991-1994, no improvement of the state of the organic world is observed in the studied area. On the contrary, in many cases the negative symptoms of degradation of trees have become stronger. There may be two main reasons: first, the organic world needs a longer period free from pollution for restoration and stabilization, and secondly, an increase in the share of acid deposition precursors in the atmospheric air of the studied area, causing an essential decrease in the annual amounts of alkaline fly ash until 1989. The monitoring of forest trees should be continued to establish the real factors. A complex of criteria for the assessment and investigation of changes in ecosystems conditioned by multicomponential air pollution level should be developed.

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