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

1. CHEMICAL COMPOSITION OF TREES AND ENVIRONMENT

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The state of Norway spruce stands on a territory influenced by air pollution from oil shale mining and processing was investigated in 1996–1998. Sample plots were selected in different directions from the industrial centre of Kohtla-Järve (Ida-Viru County, North-East Estonia). After a decrease of the emission of oil shale fly ash and a lower total concentration of pollution from industrial enterprises, the chemical composition of needles and shoots still showed latent injuries and deviations in the biochemical state of trees. Also the soil humus horizon, subsoil water and precipitation were still notably alkaline.

Introduction

Oil shale mining and processing are the most important industries in Estonia. Before 1990 the whole economic activity was characterized by intensive and unbalanced production and serious social-economic and ecological problems. Negligent attitude towards environmental protection has led to the pollution of ecosystems in many areas of North-East Estonia. The state of the atmosphere depends first of all on prolonged emissions from the industrial enterprises. Over 40 years oil shale mining and processing in the area of Kohtla-Järve, the main industrial centre in Ida-Viru County, has brought about several ecological problems like changes in the state of forests [1], soil, surface and ground water [2].

Pollutants emitted from stationary sources of oil shale processing and chemical plants in Kohtla-Järve have a multicomponental chemical character. They consist of hazardous gaseous components, soil particles and aerosols. The proportion of alkaline ash in the pollution complex has changed from year to year, making up 16–30 %. Gaseous pollutants have predominated in air pollution complexes: SO_2 has made up 42–56 %, CO 8–10 % and NO_x 2 %. From organic pollutants 30 % of aromatic and 60 % of aliphatic hydrocarbons have been established [3, 4].

Ida-Viru County is one of the most forested areas in Estonia, making up more than 10 % of all forest land [5]. Essential forest damages from human pressure were observed during the period of intensive industrial activity in Ida-Viru County [3, 6, 7]. Estonian forest monitoring showed that 14 % of young spruce had only 1–2-year-old needles [6, 8] and in autumn 1992 only current year needles were found on numerous pines [9].

The reduction in the emission of pollutants has led to a decrease in air pollution in North-East Estonia. Nevertheless, several problems such as alkalization of the environment, high concentrations of accumulated pollutants in the substrate, starting neutralization processes in the environment caused by diminishing concentrations of dust and ash in pollution complexes, should be studied thoroughly. Special attention should be paid to estimating changes taking place in forest ecosystems as several forests are commercial forests or protected forests and parks in this region.

The main aim of integrated monitoring in the forests is to determine the state of forest today. A detailed investigation of forest sample plots in different directions from the industrial town of Kohtla-Järve was carried out by the authors of this paper. The aim of our investigations was to estimate the state of the forest ecosystem and trees in 1996–1998, the period of rapid decrease in air pollution in an area that used to be highly polluted. The object of our study was Norway spruce (*Picea abies* L.), one of the prevailing tree species in Estonian forests.

Study Area and Methods

The research was carried out in the neighbourhood of Kohtla-Järve and Jõhvi towns, North-East Estonia, in 1996–1998. It was financed by the Ida-Viru County Subfund of the Estonian Environmental Fund and Estonian Science Foundation (Grant No. 3234). The chemical composition of Norway spruce, *Picea abies* (L.) Karst., and its relationships with environmental parameters (pollution load, pH and chemical composition of precipitation) were determined.

The study area is situated on the Northeast Estonian plateau, where the main pollution sources are AS Nitrofert (production of ammonium fertilizers), *Velsicol Eesti* AS (production of benzoic acid), Kohtla-Järve and Ahtme power plants, AS *Kiviter* (*Viru Keemia Grupp* AS now), AS *Metra Ehitused* (asphalt concrete), AS *Silbet* and a number of smaller enterprises (boiler houses and enrichment plants of mines, a dairy, a furniture factory etc.) (Fig. 1). Sample plots (*1–6*) were established in different directions and at different distances from Kohtla-Järve town. The control plots were located in relatively unpolluted stands on the territory of Lahemaa National Park (plots *7.1* and *7.2*) (Table 1). In selecting sample plots we proceeded from the principle of analogy of geographic and forestry parameters, with similarity of climatic and edaphic conditions as well as parameters of stand regarded as the main principle.

Parameters characterizing sample plots are presented in Table 1.

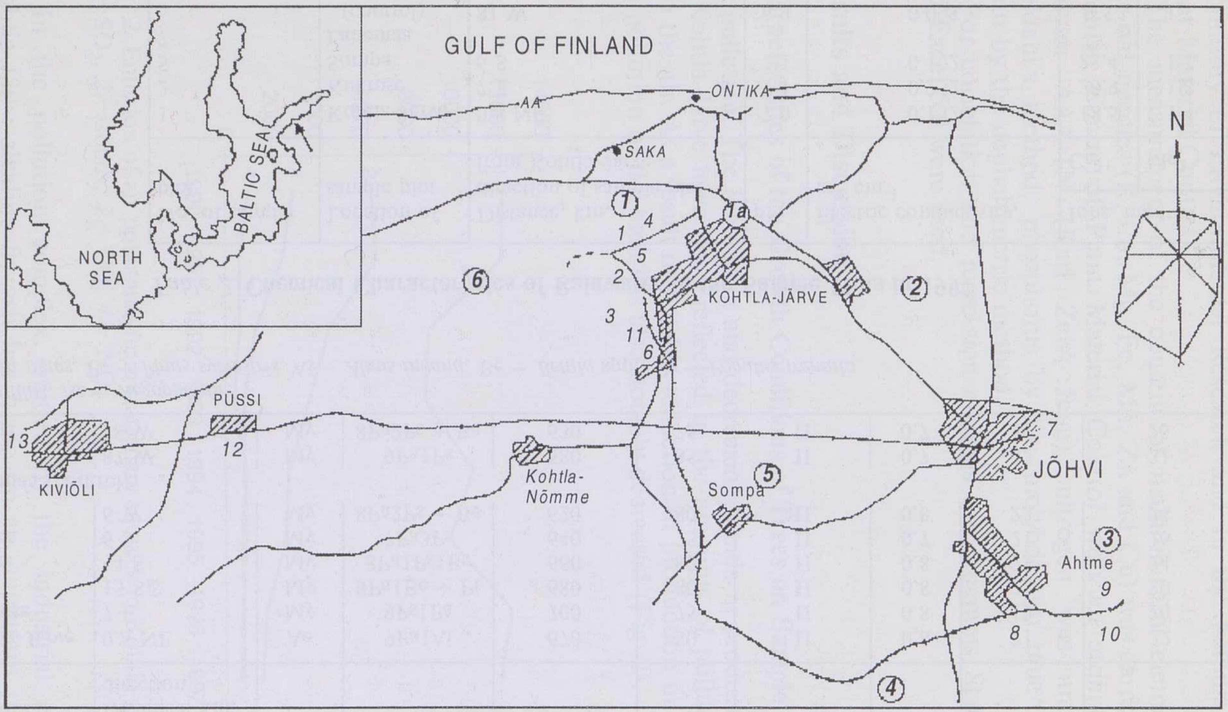

Fig. 1. The study area. Location of sample plots (*1–6* in circles) and pollution sources: *1* – AS *Nitrofert*; *2* – Kohtla-Järve PP; *3* – AS *Kiviter*; *4* – *Velsicol Eesti* AS; *5* – regional sewage purification plant; *6* – AS *Virko* (furniture production); *7* – OÜ *Jõhvi Piim* (dairy produce); *8* – AS *Metra Ehitused* (asphalt concrete); *9* – Ahtme PP; *10* – AS *Silbet*; *11* – AS *Viru Teed* (asphalt concrete); *12* – AS *Repo Püssi Vabrikud*; *13* – Kiviõli PP and Kiviõli Production Unit of *Kiviter* AS

Table 1. **Average Characteristics of the Investigated Stands of Norway Spruce at Different Distances from Kohtla-Järve [10]**

Sample plot			Forest type*	Composition of trees**	No. 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	Low
2	Kukruse	7 E	My	9Ps1Pa	700	75	II	0.8	19	20	Low
3	Kose	15 SE	My	9Pa1Be + Pt	680	80	II	0.8	22	20	Low
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 (control)										
7.1	Eru	87 W	My	9Pa1Ps	580	85	II	0.7	22	31	Moderate
7.2	Revoja	78 W	My	8Pa2Ps + Be	630	75	II	0.7	20	26	Moderate

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

Table 2. **Chemical Characteristics of Rainwater in the Sample Plots in 1997**

No. of sample plot	Location of sample plot	Distance, km, direction of sample plot from Kohtla-Järve	pH	Electric conductivity, mS cm^{-1}	Ions, mg l^{-1}			
					Ca^{2+}	Mg^{2+}	K^{+}	SO_4^{2-}
1	Kohtla-Järve	0.5 NE	7.0	0.132	18.5	1.4	3.8	14.2
2	Kukruse	7 E	7.1	0.232	30.9	1.8	8.5	16.9
5	Sompa	6 S	7.1	0.102	22.4	1.7	11.0	15.7
7	Lahemaa (control)	81 W	6.9	0.098	9.5	1.7	2.1	8.1

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 and from the control area when a permanent snow cover had formed. Rain, subsoil water and soil were collected during the vegetation period. Chemical analyses of the samples were performed in the Central Laboratory of Environmental Research and in the Estonian Centre of Plant Material Control.

The measurement of the content of main mineral elements in one-year-old needles (K, Ca, Mg, Fe, Mn, Zn and Cu) was carried out at the Estonian Centre of Plant Material Control with an atomic absorption analyser AAA-1N (Karl Zeiss Jena), nitrogen was analysed using Kjeldahl's method, phosphorus by the molybdenum blue method and sulfur by the nephelometric method.

For correlation and regression analysis the programs Statgraphics 5.0 and Excel 5.0 were used.

Results and Discussion

Characteristics of the Growth Conditions of Trees on Sample Plots

Air pollution. The increasing and decreasing trends in production intensity at Kohtla-Järve have been reflected in the amounts of pollutants emitted into the air. As a result of notable reduction of production in recent years, the pollution of the area has also notably decreased (Fig. 2).

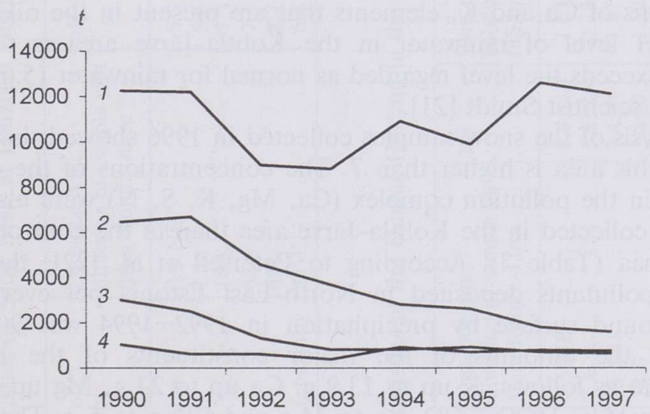

Fig. 2. Emission of air pollutants from stationary sources in Kohtla-Järve [11–18]. *1* – SO_2, *2* – solids, *3* – CO, *4* – NO_x

In the pollution complex emitted by the industrial enterprises of Kohtla-Järve alkaline fly ash has made up an important percentage. Its proportion in total emission has fallen from 30 % (in 1989) to 9 % (in 1997). A sharp decline has occurred in the amount and share of solid matter among the air pollutants in the last five years. The proportion of SO_2 in the total emission has increased by today after a certain fall in

1992–1994. Although a tendency of decrease of amounts of H_2S emitted from AS *Kiviter* was established: 16 t yr^{-1} in 1992 and decreasing to 8 t yr^{-1} in 1998 [19]. No major reduction in the amounts of NO_x and CO was registered. In addition to oil shale ash and gases the pollution complex emitted from the enterprises of Kohtla-Järve contains also large amounts of organic compounds, in which hydrocarbons (benzene, toluene, styrene), phenols, formaldehydes etc. predominate [4].

The distribution of pollutants depends strongly on meteorological conditions – the direction and strength of winds, precipitation amounts, etc. In case winds blow from north and north-west the pollution load will be carried towards Sompa–Kalina. However, in case of the prevailing southwesterlies (30–40 %) the air of Kohtla-Järve, Kukruse and also of the Kose area (the last one largely due to pollutants from Ahtme power plant) will be polluted most heavily [20]. Winds both from east and west carry pollutants to the Aa area (which is affected by several pollution sources from different directions) [3].

Precipitation. The precipitation samples collected in 1997 show significant differences from the control area. The high electric conductivity of rain and snow water in the study area as compared to the control (in rainwater up to 2.4 and in snow water up to 2.6 times difference) indicate a bigger amount of dissolved compounds in the precipitation of the industrial region of Kohtla-Järve (Tables 2 and 3). As the concentration of sulfur compounds (SO_2, H_2S) in the air is high and they are easily soluble, the rainwater falling on the ground and vegetation contains large amounts of SO_4^{-2} (Table 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 and exceeds the level regarded as normal for rainwater (5.6–6.6) by the Austrian scientist Smidt [21].

The analysis of the snow samples collected in 1996 showed that the pH of snow in this area is higher than 7. The concentrations of the elements dominating in the pollution complex (Ca, Mg, K, S, N) were also higher in the snow collected in the Kohtla-Järve area than in the control samples from Lahemaa (Table 3). According to Petersell et al. [22], the annual amount of pollutants deposited in North-East Estonia per every square metre of ground surface by precipitation in 1992–1994 was 90–133 g. 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 g, Mg up to 3.1 g, SO_4–S up to 15 g, HCO_3–CO_2 up to 34 g and Cl up to 5 g. The amount of N deposited on the ground by precipitation during these years was up to 1.6 g m^{-2}. The high pH levels and contamination of precipitation in the Kohtla-Järve area was earlier pointed out by Frey [23].

Soil. The chemical composition of soils in the Kohtla-Järve area also indicates long-term industrial pollution. Alkalization of soils is especially clearly observed (Table 4). This phenomenon was also described by Kokk [24] in mapping forest soils in North-East Estonia. Alkalization has been found also in case of bog soils in this region [25].

Table 3. **Chemical Characteristics of Snow Melt at Different Distances from Kohtla-Järve in 1996**

No. of sample plot	Location of sample plot	Distance, km; direction of sample plot from Kohtla-Järve	pH	Electric conductivity, mS cm^{-1}	Ions, mg l^{-1}						
					Ca^{2+}	Mg^{2+}	K^+	Na^+	NH_4^+	NO_3^-	SO_4^{2-}
1	Kohtla-Järve	0.5 NE	8.3	47.2	8.2	0.36	0.30	0.6	0.35	0.56	7.1
2	Kukruse	7 E	8.6	58.9	9,2	0.40	0.20	2.4	0.25	0.56	7.4
4	Kalina	14 S	7.3	43.7	6.3	0.52	0.30	0.5	0.21	0.43	9.3
5	Sompa	6S	7.4	47.7	8.9	0.43	0.20	0.5	0.27	0.48	7.8
6	Aa	6W	8.7	40.1	13.7	0.25	0.30	0.3	0.12	0.50	4.2
7	Lahemaa (control)	81W	6.8	22.5	4.3	0.14	0.15	0.6	0.17	0.30	2.4

Table 4. **Chemical Characteristics of the Humus Horizon in the Sample Plots in 1996–1997**

No. of sample plot	Location of sample plot	Distance, km; direction of sample plot from Kohtla-Järve	pH	Electric conductivity, mS cm^{-1}	Elements available by plants, mg $100g^{-1}$					
					S (total)	Ca	Mg	K	P	Fe
1	Kohtla-Järve	0.5 NE	6.8±0.1	1.09±0.06	55±5.2	659±163	29.3±1.5	8.9±3.06	1.32±0.06	2.00±0.21
2	Kukruse	7 E	5.8±1.0	0.78±0.35	60±4.4	484±55	40.0±43.3	11.0±4.9	0.40±0.01	3.3±0.09
3	Kose	15 SE	7.4±0.1	1.47±0.15	80±5.3	1533±275	127.6±22.5	15.2±1.5	1.01±0.10	5.3±0.98
4	Kalina	14 S	5.8±0.5	1.59±0.17	165±18.1	983±238	60.6±1.1	17.9±5.8	1.19±0.10	5.7±1.41
5	Sompa	6 S	4.7±0.1	0.59±0.03	50±1.9	173±35	13.0±4.3	4.3±0.3	0.66±0.09	8.6±2.11
6	Aa	6 W	5.9±0.9	1.52±0.36	135±10.2	1258±181	73.7±26.8	14.7±3.2	1.01±0.10	5.5±0.12
7	Lahemaa (control)	81 W	3.6±0.3	0.48±0.03	43±8.9	85±38	9.2±3.9	4.7±2.0	1.85±0.14	0.6±0.01

The upper horizons of the soil, especially the litter and humus horizons, are under the 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 concentrations of Ca, Mg and K, which dominate in the solid fraction of the pollution complex, are high as compared to Lahemaa samples, being respectively 18, 14 and 4 times as high as the control (Table 4). Big differences from the control were observed in the total S content in soil (up to 4 times as high as the control). In 1986–1990 the mean N content in humus horizons of soils in the Kohtla-Järve region was several times higher than in the soils of the same type in Lahemaa (0.4 mg $100g^{-1}$) [3] being 1.9 mg $100g^{-1}$ at the distance of 0.5 km N, 2.57 at 2 km W, and 1.58 at the 3 km N from Kohtla-Järve. Studies in 1997 suggested a decreasing tendency of N in humus horizons compared to previous years. Also high values of electric conductivity of soil solutions indicate soil pollution in the study area.

Subsoil water. The analysis of the subsoil water samples taken from the study plots showed analogously to soil samples changes in the chemical composition and electric conductivity (Table 5). Like the pH of soil also the pH of subsoil water in the Kohtla-Järve is higher than the control. The subsoil water in the vicinity of the Ahtme power plant at Kose sample plot is highly alkalized (pH by more than 2 units higher than the control), but also elsewhere the pH levels are higher than the control. As a rule, samples showed higher amounts of Ca, K, Mg and S, dominant elements of oil shale fly ash. As a result of reduced solubility in an alkaline environment, the content of heavy metals in the subsoil water of Kohtla-Järve area was somewhat lower than the control. If the soil pH is within the range 5.5–7.0, the mobility of heavy metals tends to be low; however, acidification of soil may trigger their higher mobility. The subsoil water of the study area showed as a rule elevated electric conductivity (Table 5).

Chemical Composition of Trees

Foliar analyses provide a means for assessing changes in the content of nutrients and contaminants allowing for assessment of nutrient fluxes and nutritional status of forest trees or are an effective means of recognizing emission-related stress in trees.

Although up to 1998 the level of air pollution emitted from Kohtla-Järve industrial enterprises decreased essentially (Fig. 2), the foliar diagnosis of the accumulation of pollution components demonstrated differences between the spruces in the industrial territory and in the unpolluted control area.

Essential changes in the chemical composition of forest soil during the prolonged air pollution impact in the studied area can be interpreted as a real cause affecting the chemical composition and nutrient balance in trees. The investigated stands are located in industrial regions with moderately alkalized soils [24, 26], where the pH of the soil litter horizon has increased by 2.0–2.9 units compared to the control.

Table 5. **Chemical Characteristics of Subsoil Water in the Sample Plots in 1996–1997**

No. of sample plot	Location of sample plot	Distance, km; direction of sample plot from Kohtla-Järve	pH	Electric conductivity, mS cm^{-1}	Ions, mg $100g^{-1}$					
					Ca^{2+}	Mg^{2+}	K^{+}	Mn^{2+}	Fe^{3+}	SO_4^{2-}
1	Kohtla-Järve	0.5 NE	6.9±0.4	0.12±0.01	19.0±3.3	2.2±1.5	7.0±0.2	0.005±0.002	0.081±0.04	10.6±8.3
2	Kukruse	7 E	6.4±0.3	0.07±0.02	9.6±2.1	1.3±0.6	1.8±0.8	0.009±0.004	0.182±0.129	8.4±3.7
3	Kose	15 SE	7.2±0.5	0.21±0.03	46.6±7.8	3.1±0.3	4.4±2.3	0.018±0.011	0.110±0.024	18.7±6.7
4	Kalina	14 S	6.4±0.4	0.16±0.07	26.9±7.9	3.7±1.6	7.5±3.2	0.005±0.001	0.266±0.076	13.3±6.4
5	Sompa	6 S	6.1±0.7	0.22±013	17.1±4.7	4.1±0.4	24.4±8.7	0.032±0.002	0.294±0.021	36.7±26.6
6	Aa	6 W	5.9±0.4	0.21±0.11	28.2±13.4	4.0±2.1	9.5±2.2	0.010±0.005	0.269±0.094	16.5±8.4
7	Lahemaa (control)	81 W	5.1±0.3	0.08±0.01	11.4±3.3	1.8±0.5	4.6±1.0	0.041±0.011	0.882±0.194	9.5±5.2

Obviously the soil pH and its changes vary with the distance from the pollution source, characteristics of the emitted pollutants, landscape peculiarities and other factors. As a rule the pH level of precipitation, snow and subsoil water had increased in our forest sample plots (Tables 2–4). As known, the assimilation of nutrients by plants depends essentially on the pH of the growth substrate.

It has been determined that plants assimilate easily Ca and K from the growth substrate with pH over 6.5 [27, 28], so their concentrations in needles and shoots of Norway spruce compared to the control area were higher in the Kohtla-Järve, Kukruse, Sompa, Aa sample plots (Fig. 3; Table 6). In several sample plots the concentration of Mg, an essential element in photosynthesis, has decreased in needles and especially in shoots (Sompa) (Fig. 3; Table 6).

The regression analysis of the content of different elements in Norway spruce needles and soil pH in the humus horizon showed that soil pH is an important factor regulating Ca^{2+} ($R^2 = 0.617$), K^+ ($R^2 = 0.568$) and Mg^{2+} ($R^2 = 0.528$) accumulation in needles.

The content of heavy metals (Cu, Zn, Mn) in the needles and shoots from the industrial area was much lower than the control. However, Fe concentration in trees showed high values in the needles and shoots in the Kohtla-Järve region. This variability is in direct correlation with the pH of the growth substrate. The regression analysis showed a linear relationship between the subsoil water pH and the concentration of Fe^{3+} ($R^2 = 0.671$), Cu^+ ($R^2 = 0.656$) and Mn^{2+} ($R^2 = 0.512$) in needles.

The drastic decrease in the Mn concentration in plant tissues is also connected with soil alkalization. In alkaline conditions Mn^{2+} compounds turn into Mn^{3+} or Mn^{4+} compounds, which have low solubility and are practically not available to plants [27]. Mn deficit has developed in spruces in the surroundings of Kohtla-Järve (Aa, Kalina, Sompa): its concentration in the investigated needles and shoots was respectively only 19 % and 30 % of the control. A similar situation was observed earlier in coniferous trees in alkalized areas in the vicinity of a cement plant emitting alkaline (pH 12.7) dust pollution [29]. The assimilation of Cu and Zn compounds is better in acidic conditions of growth at pH 3.5–5.5 [28], so their availability to Norway spruce in the Kohtla-Järve region has decreased respectively by 28 % and 37 % in the needles and 15 % and 4 % in the shoots compared to the control trees.

The territory surrounding Kohtla-Järve has been influenced by a high level of air pollution emitted from the local industrial enterprises for over 40 years. High concentrations of SO_2 in the atmosphere have caused an increase in SO_4^{2-} concentration in precipitation, snow and soil (Tables 2–4). The comparatively high level of S in soil and subsoil water has led to increasing S concentrations in spruce needles, which make up about 45 % of the S content in the control trees. Regression analysis showed a linear relationship between the S concentrations in needles and the SO_4^{2-} content in soil ($R^2 = 0.564$), subsoil water ($R^2 = 0.697$), and also with the electric conductivity of subsoil water ($R^2 = 0.608$).

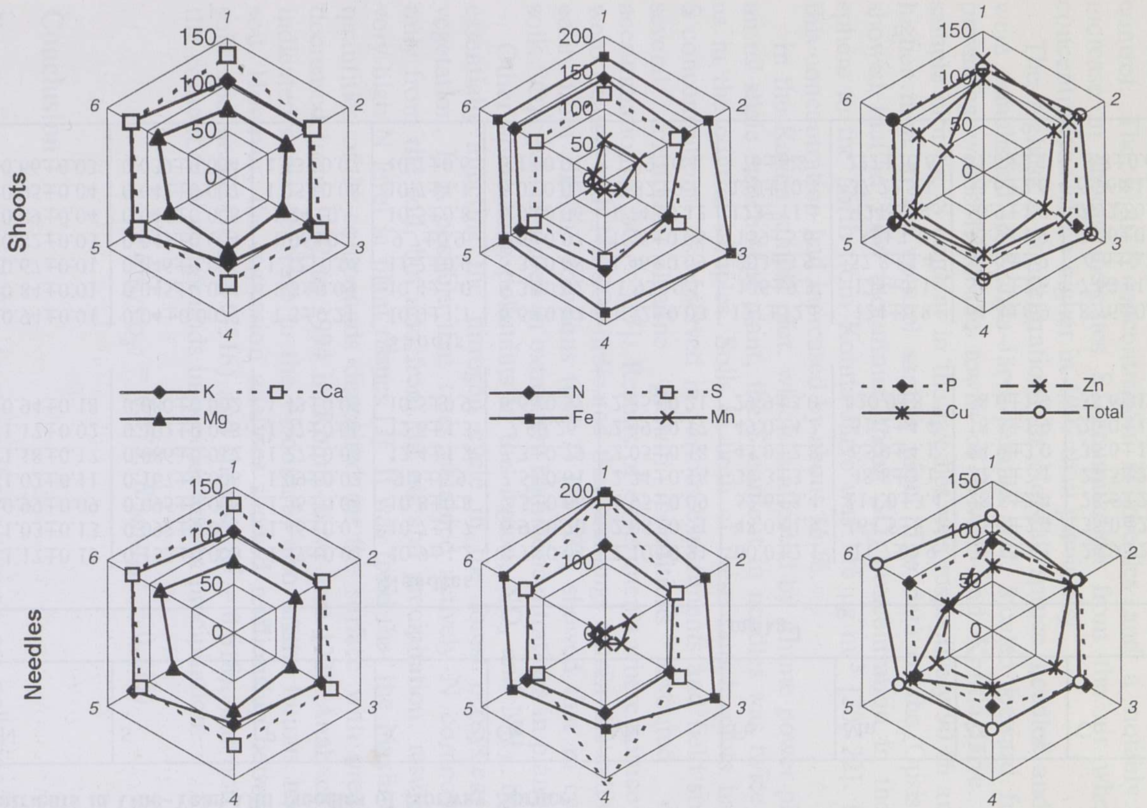

Fig. 3. Content of mineral elements (% of control) in one-year-old needles of Norway spruce growing different sample plots. *1–6* – numbers of sample plots

Table 6. **Content (±SD) of Nutrients in One-Year-Old Needles of Norway Spruce**

No. and location of sample plot	Total content of mineral elements	N	S	P	K	Ca	Mg	Fe	Mn	Zn	Cu
	% dw			g kg⁻¹				mg kg⁻¹			
					Needles						
1 Kohtla-Järve	4.06±0.9	1.17±0.13	0.152±0.006	1.27±0.02	10.9±1.2	8.7±0.04	2.10±0.21	60.0±2.1	11.7±0.9	36.1±3.1	24.9±2.1
2 Kukruse	3.50±0.8	1.03±0.13	0.092±0.001	1.46±0.02	10.7±1.7	6.9±0.20	2.08±0.31	48.0±1.9	164.5±8.2	35.0±2.4	35.0±2.7
3 Kose	3.64±0.8	0.99±0.09	0.095±0.003	1.56±0.04	10.8±0.8	7.5±0.68	1.95±0.09	52.6±3.4	114.0±3.4	28.5±2.4	28.5±2.2
4 Kalina	3.50±0.1	1.02±0.11	0.167±0.008	1.09±0.02	9.8±0.9	7.5±0.61	2.24±0.18	39.3±3.1	48.8±3.1	22.5±2.1	22.5±2.4
5 Sompa	3.82±0.2	1.18±0.17	0.086±0.012	1.27±0.03	12.4±1.2	7.3±0.79	2.05±0.18	45.0±2.8	65.9±4.1	24.9±3.0	36.6±1.9
6 Aa	4.68±0.2	1.17±0.02	0.102±0.013	1.37±0.06	12.6±1.3	7.90.26	2.49±0.17	49.0±4.2	61.2±4.4	18.5±1.9	20.0±1.0
7 Lahemaa (control)	3.53±0.3	0.94±0.18	0.080±0.002	1.43±0.05	10.5±0.9	6.6±0.37	2.85±0.21	29.9±3.0	420.0±8.7	38.6±1.9	38.6±1.7
					Shoots						
1 Kohtla-Järve	2.73±0.08	0.94±0.01	0.04±0.0077	1.3±0.21	10.0±1.1	6.6±0.02	1.52±0.03	131±12.1	124±6.8	61.8±8.9	8.76±0.9
2 Kukruse	2.98±0.12	0.84±0.01	0.045±0.004	1.3±0.09	10.4±1.0	6.3±0.02	1.95±0.1	126±9.3	125±7.1	54.5±2.6	7.63±1.9
3 Kose	3.34±0.11	0.67±0.01	0.046±0.006	1.32±0.08	11.2±0.9	6.3±0.08	1.46±0.09	302±3.9	57.8±3.4	51.0±2.2	6.6±2.1
4 Kalina	2.86±0.09	0.82±0.03	0.043±0.004	1.06±0.1	9.7±0.9	6.6±0.07	1.46±0.06	139±5.6	32±3.1	45.9±1.9	7.66±0.8
5 Sompa	2.69±0.08	0.89±0.04	0.044±0.008	1.24±0.1	10.5±0.8	6.0±0.06	1.74±0.12	123±11.1	424±2.9	50.9±1.9	7.32±0.9
6 Aa	2.73±0.12	0.95±0.04	0.042±0.007	1.25±0.08	10.7±1.1	6.0±0.02	1.82±0.1	130±10.1	37.2±5.1	33.6±2.6	6.96±1.1
7 Lahemaa (control)	2.49±0.15	0.66±0.03	0.039±0.004	1.13±0.07	10.1±0.6	5.1±0.01	1.49±0.1	76±6.8	222±10.6	51.6±2.1	8.8±0.6

The increase in the S content in shoots was estimated at 14 % of the control. The high S concentration in needles and a notably smaller increase in shoots indicates S accumulation from the air where SO_2 concentrations where higher in 1997 than previously.

The highest S concentrations in Norway spruce needles and shoots were established in Kohtla-Järve sample plot, located leeward from the prevailing winds from the towns of Kiviõli and Kohtla-Järve. In this sample plot the S content in the needles was respectively 90 % and 21 % higher than in the control sample plots. The data of the Opsis system showed that in 1994 the annual mean SO_2 concentration in the atmosphere to the north from Kohtla-Järve was 116 $\mu g\ m^{-3}$ [20, 22]. By 1997 this concentration had increased by about 16 %.

In the Kalina sample plot, which is affected by Ahtme power plant and an oil shale enrichment plant, the S content in needles was twice as high as in the control samples. Soils in both of these sample plots have high S concentrations. As observed in various experiments and field studies by several scientists alkaline growth conditions favoured intensive accumulation of SO_4^{2-} [27]. Results of experiments where spruce needles were treated with Na_2SO_4 indicated that seedlings obtain the sulfate more easily from alkaline solutions (pH 7) [30, 31] showed that in calcinated soils SO_4^{2-} was absorbed in extremely high concentrations in plants.

Other dominating pollutants in this region, NO_x and NH_3, have been essentially affecting the nitrogen cycle in the forest ecosystem and in vegetation. It is possible that trees obtained actively N compounds not only from the soil, but also from the air and precipitation, resulting in a very high N level in plant tissues [3]. It was fixed that the maximum total quantities of N compounds deposited on the surface with precipitation decreased from 1992 to 1994 in Kohtla-Järve area [22]. Analyses in 1997 indicated some decrease in the N content also in the humus horizon of soil. However, the regression analyses showed relationship between N in soils and needles ($R^2 = 0.516$) and shoots ($R^2 = 0.618$), but not between the content of N compounds in needles and in precipitation

$$(R^2_{N/NO_3^-} = 0.475;\ R^2_{N/NH_4^+} = 0.111).$$

Conclusion

To sum up we would like to stress that although air pollution has been decreasing in the area investigated, this decrease is not significantly reflected in the chemical composition of Norway spruce. Obviously the composition of the needles and shoots of conifers is affected in addition to the current air pollution also by pollutants accumulated in the soil during previous years.

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