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NEEDLE BIOMASS ACROSS A POLLUTION GRADIENT IN ESTONIA

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The research was carried out in the region of oil shale industry (Kohtla-Järve) and a cement factory (Kunda) in Northeast Estonia in 1996–1999. Large amounts of solid wastes emitted into the atmosphere in this region for decades have brought about alkalisation of the environment, especially in the vicinity of Kunda. Needle biomass analysis of 75–85-year-old Scots pine and Norway spruce growing on sample plots in the polluted area and in the control area showed that air pollution has had variable effect on the parameters characterising the state of trees: fresh and dry mass of needles and dry matter content. Changes in the fresh and dry mass of the needles of conifers revealed a clear negative trend in sample plots closer to emission sources. With the fallen total air pollution load, some improvement of the state of conifers was estimated in recent years. A significant difference in the dry matter content of needles from the control was observed only in a few cases.

Introduction

Depending on air pollution, the duration of its effect and chemical composition of pollution may cause negative or positive responses in forest productivity [1–3]. The problem of forest damages caused by dust pollution emitted from different industrial enterprises is not new, but it is not completely understood and interpreted on the level of impact mechanisms, the character of responses and tolerance of forest types. Several studies describe negative changes in the morphology and physiology of plants under high levels of alkaline cement dust [4, 5].

Investigations have shown that adverse effects on coniferous species occur within 2-3 km from a cement plant, with mature pine forests damaged to a moderate degree, and spruce forests to a high degree [6]. Outside this zone, the state of pine forests and their growth improves toward the plant. A clear fall can be observed in the radial increment of Scots pine and Norway spruce

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at a distance of 2-3 km W and up to 5 km E from the Kunda cement plant where the emission load was especially high [7, 8].

Zubareva *et al.* [9] investigated the effect of dust pollution on the components of birch (*Betula* spp.) phytocenoses in the impact zone of limestone quarries and a cement plant in Russia. It was shown that cement dust causes a reduction of the leaf size and alters the optical density and physiological parameters of the leaves of woody plants [10]. The amount of photosynthesis, respiration and transpiration of the needles of *Thuja orientalis* polluted by cement dust decreased to various extents [11]. However, cement plant emissions may also have a positive effect on the length growth of needles and shoots when the dust deposition is low [2, 12].

Emission of air pollution from the enterprises of oil shale mining and processing in the area of Kohtla-Järve, the main industrial centre in Ida-Viru County, Estonia, has brought about several ecological problems due to high level of fly ash in air pollution complexes [3, 13]. Around this industrial centre, the level of defoliation of conifers is relatively high and only one- and two-year-old needles were registered on the shoots of spruces in the vicinity of Kohtla-Järve (the average needle age in Estonia is 5–8 years) [13]. Changes in the concentrations of neither fly ash nor SO_2 caused variations in the growth of trees and needle retention at the distance of 15 km SE from Kohtla-Järve [14].

Our previous results [15] showed that changes in the length growth of needles and shoots are a suitable indicator of pollution emitted by oil shale industry as these revealed a clear negative trend in sample plots closer to emission sources. Pollution fluxes in the vicinity of industrial enterprises in Kohtla-Järve have brought about a major fall in the vitality of trees, their large-scale dying out and an about 50-% reduction in their radial increment [3, 15].

The main objective of the current study was to estimate the state of Scots pine (*Pinus sylvestris* L.) and Norway spruce (*Picea abies* (L.) Karst.), which are the dominant tree species in Northeast Estonia, in the area influenced by alkaline oil-shale fly ash and cement dust emitted from industrial enterprises of this region by using biomass and dry matter content of needles of conifers and environmental analyses. Our results will hopefully contribute to a better understanding of forest health dynamics in Estonian as well as in other industrial areas.

Material and Methods

Study Sites

Problems connected with environmental pollution in Estonia are most urgent in Ida-Viru and Lääne-Viru counties, where the relationship between nature and human society has in places aggravated beyond limits. In these counties several major sources of air pollution are situated: the cement plant *Kunda*

Nordic Cement, Estonian and Baltic thermal power plants, a number of smaller power stations, oil shale mining and processing and chemical enterprises. Main pollution sources in the surroundings of Kohtla-Järve are *Nitrofert AS* (production of nitrogen fertilizers), *Velsicol Eesti AS* (benzoic acid), Kiviõli, Kohtla-Järve and Ahtme power plants, *Kiviter 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.).

Six sample plots (pine and spruce stands) were established in different directions and at various distances from Kohtla-Järve. The plots were located: 0.5 km NE from the nitrogen fertilizer plant (Kohtla-Järve); 7 km E (Kukruse); 15 km SE (Kose, 3 km NE from Ahtme power plant); 6 km S (Sompa); 14 km S (Kalina, 0.5 km S from Viru enrichment plant and 6 km SW from Ahtme power plant); and 6 km W (Aa, 6 km NE from the Püssi plants of *AS REPO* and 11 km NE from Kiviõli power plant and oil shale chemistry plant) from the plant.

Five sample plots were established around the Kunda Cement Plant: 2 km (Tigapõllu) and 3 km (Toolse) to the west, and 2.5 km (Lontova 2), 5 km (Malla) and 12 km (Rannametsa) to the east.

Control stands were situated in a relatively unpolluted territory of Lahemaa National Park: about 34 km W from the cement plant and 82 km W from the fertilizer plant (*Nitrofert AS*) in Kohtla-Järve.

In selection of sample plots (0.05 ha each) we proceeded from the principle of analogy of geographical and forestry characteristics. It was important for comparison that climatic and edaphic conditions as well as parameters of stands on the plots should be similar. The selected stands were 75–85-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. The soils of the forest sample plots were Gleyic Podzols on sands [16]. As in the Kohtla-Järve area no *Myrtillus*-site type Norway spruce stands occur, the sampling plot (0.5 km NE) in that area was established in *Aegopodium*-site type stands, which are quite similar to the *Myrtillus*-site type stands.

Usually the *Myrtillus*-site type stands consist of pine or spruce, or both, often including also birch and aspen [17, 18]. A typical feature is abundant natural renewal of spruce. The understorey is sparse or absent [18]. It may include black alder, rowan and juniper. A characteristic feature of the plant cover is occurrence of dense moss and blueberry [17, 18].

Morphological Measurements

Attention was focused on the analysis of the morphological parameters of pine and spruce: fresh and dry mass (dried at 65 °C to a constant mass) of needles (g) and dry matter content (dry mass : fresh mass × 100, %) of needles. Branches for analyses were collected at the height of 6 m and from the southern side of trees (10) of similar size in summer 1996–1999. Soil samples were taken from the humus horizon of the sample plots during the vegetation period in 1996, 1997 and 1999. The concentrations of Ca, Mg and K in the soil were determined in the Estonian Control Center of Plant Production. For finding statistical differences (t-test, $\alpha = 0.05$) the program Excel 5.0 was used.

Results and Discussion

Character of the Growth Environment

Oil shale mining and processing in the area of Kohtla-Järve and cement production in Kunda are relatively important industrial activities in Estonia.

Pollutants emitted from stationary sources of oil shale processing and chemical plants in Kohtla-Järve have a multicomponent chemical character as they consist of hazardous gaseous components (SO_2, NO_x, CO), fly ash, solid particles and aerosols. The total emission was 32,580 t in 1989; since then the total pollution load has fallen nearly two times [19]. The proportion of alkaline ash in the pollution complex has changed from year to year, falling from 30 % (1989) to 9 % (1997). The proportion of gases in the total emission increased over this period. Gaseous pollutants have predominated in air pollution complexes: SO_2 has made up 42–56 %, CO 8–10 % and NO_x 2 % (Fig. 1).

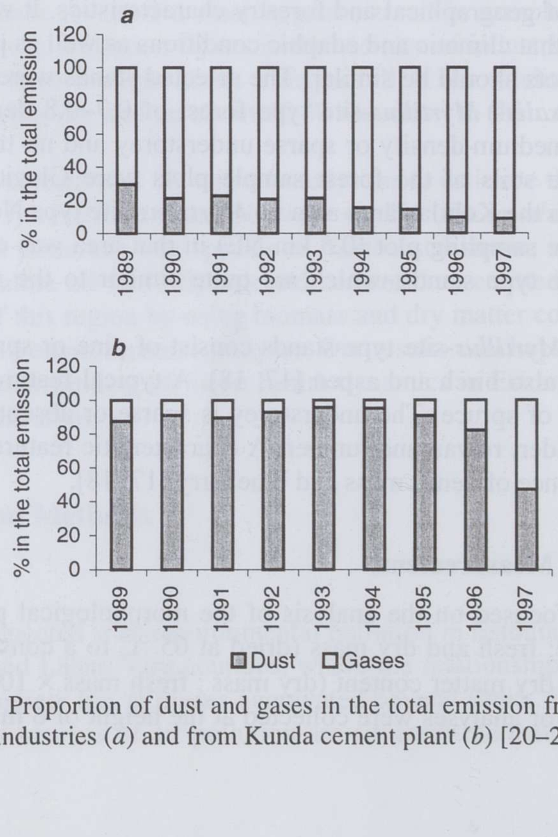


Fig. 1. Proportion of dust and gases in the total emission from Kohtla-Järve industries (*a*) and from Kunda cement plant (*b*) [20–28]

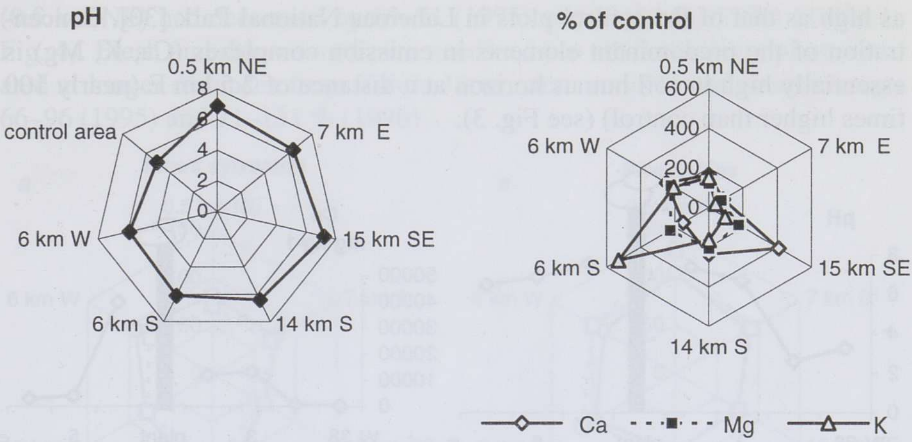
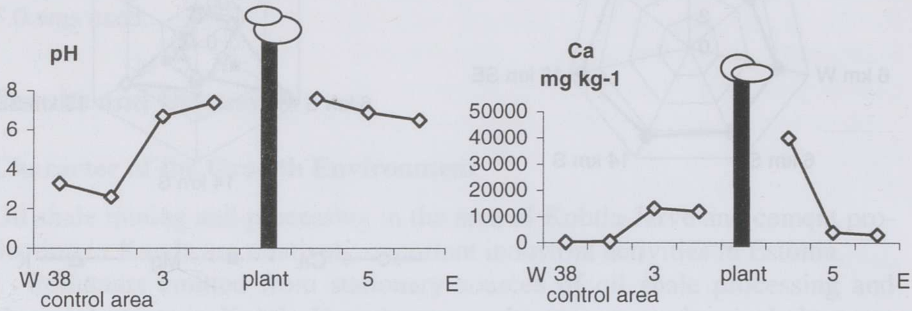


Fig. 2. Chemical characteristics of the humus horizon at different distances and directions from Kohtla-Järve in 1996–1997

Over 40 years of oil shale mining and processing in the area of Kohtla-Järve has brought about several ecological problems such as changes in 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 (subsoil water pH 5.1, rainwater pH 6.9 and snow melt 6.8) [19]. The influence of air pollution is observed also in chemical composition of the soil. The average pH is 6.1 versus 3.6 in the control samples (Fig. 2). 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.

Long time the main damaging factor for trees in the area of Kunda Cement Plant (established in 1871) was apparently dust. The total amount of dust emitted by the plant varied depending on the condition of equipment and production intensity. In 1989–1996, dust made up 82–96 % of the total emission, but only 47 % in 1997 [20–28] (see Fig. 1). According to the information provided by the plant, maximum emission of dust (98,900 tons) occurred in 1991 [22]. In the last years, dust pollution fell significantly: from 31,400 tons in 1995 to 521 tons in 1999 [26, 29].

The amount of various exhaust gases (SO_2, NO_x, CO, etc.) has been smaller. Dust load on the sample plots was approximately 100–2700 g m^{-2} $year^{-1}$ depending on the velocity and direction of winds [2]. High deposition of alkaline dust (pH 12.3–12.6) has brought about significant alkalisation of the soil (pH of the humus horizon is 6.5–7.6) (Fig. 3). Although emission of dust had decreased in the last years, analyses of soil samples collected to characterise the growth substrate of trees showed that within the 3-km radius from the cement plant the pH value of the humus horizon of weakly podzolised temporarily overmoist sandy soils was still 2.0–2.9 times as high as that of the control plots in Lahemaa National Park [30]. Concentration of the predominant elements in emission complexes (Ca, K, Mg) is essentially high in soil humus horizon at a distance of 2.5 km E (nearly 100 times higher than control) (see Fig. 3).



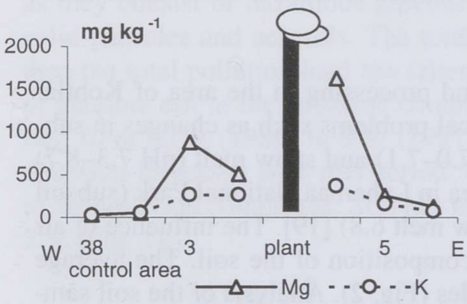


Fig. 3. Values of soil pH and content of some mobile forms of chemical elements in humic horizons of forest sample plots in Kunda area in 1999

Biomass of Needles

Biomass of needles and total foliage are recommended parameters for estimation of tree quality and production. The dust layer covering the needles creates a higher temperature, and thus more active respiration and transpiration in the tissues of needles [31, 32]. This results in a greater water content variability in polluted needles than in unpolluted ones, reflected in the fresh mass of needles.

Impact of industrial dusts on the dry mass of needles may differ depending on chemical composition and pH of dust from various sources. So, dust emitted from an aluminium factory decreased the dry mass of pine needles by about 26 %; however, the same amount of dust from a power station had no effect on the dry mass of needles while cement dust may be stimulative [1].

Our results showed that the complex effect of the changed growth conditions and continuing air pollution still decreased the biomass of needles of 75–85-year-old Scots pine and Norway spruce in the industrial area of Northeast Estonia. The data for fresh and dry mass of needles revealed great differences between sample plots. As compared with the control trees, the average biomass of 100 needles of Scots pine in the vicinity of Kohtla-Järve

(0.5 km NE) had decreased by 69–71 (1995) and 79–81 % (1996) (Table 1, Fig. 4). Fresh and dry mass of pine needles from other sampling plots, situated at longer distances from Kohtla-Järve, were variable and as an average 66–96 (1995) and 71–131 % (1996).

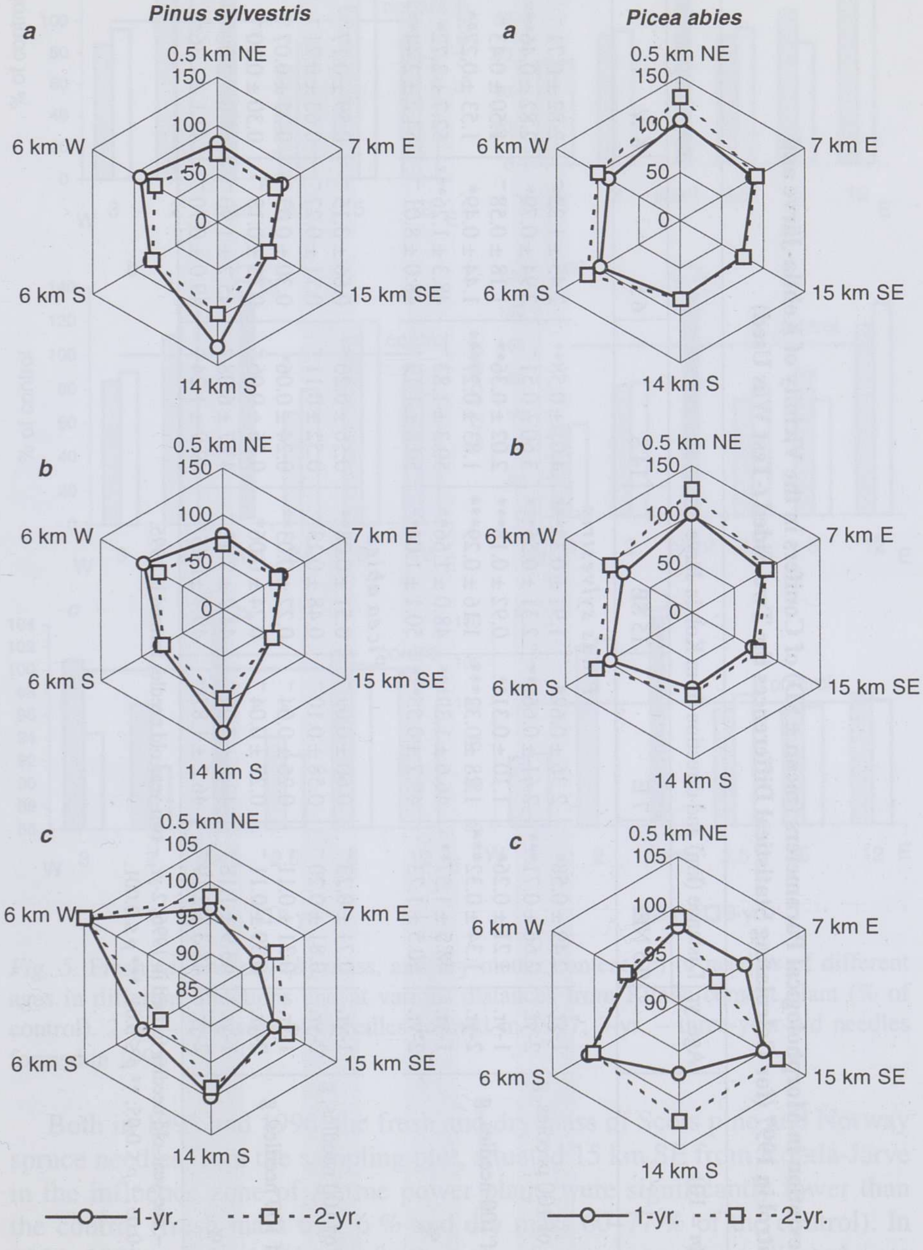


Fig. 4. Fresh (*a*) and dry (*b*) mass, and dry matter content (*c*) of needles of different ages in sample plots around Kohtla-Järve (% of control). 1-yr. – one-year-old needles formed in 1996; 2-yr. – two-year-old needles formed in 1995

Table 1. **Deviation in Morphological Parameters (mean ± SD) of Conifers in the Vicinity of Kohtla-Järve and at the Control in 1997 (for Calculating Statistical Differences the Two-Sided *t*-Test Was Used)**

Parameter (n = 10)	Age, yr	Distance (km) and direction from Kohtla-Järve						
		0.5 NE	7 E	15 SE	14 S	6 S	6 W	82 W (control)
Pinus sylvestris								
Fresh mass of 100 needles, g	1-yr.	2.49 ± 0.58*	2.36 ± 0.62*	1.91 ± 0.33***	4.01 ± 0.58**	2.45 ± 1.22$^-$	2.82 ± 0.71$^-$	3.06 ± 0.61
	2-yr.	2.66 ± 0.71***	2.61 ± 0.66***	2.31 ± 0.55***	3.60 ± 0.51$^-$	2.94 ± 0.79*	2.82 ± 0.46***	3.76 ± 0.45
Dry mass of 100 needles, g	1-yr.	1.21 ± 0.26*	1.10 ± 0.31**	0.92 ± 0.18***	2.02 ± 0.36**	1.18 ± 0.58$^-$	1.50 ± 0.43$^-$	1.54 ± 0.30
	2-yr.	1.34 ± 0.32***	1.28 ± 0.32***	1.16 ± 0.29***	1.83 ± 0.27***	1.44 ± 0.49*	1.53 ± 0.27**	1.94 ± 0.23
Dry matter, %	1-yr.	48.5 ± 1.57**	46.6 ± 1.39***	48.0 ± 1.66**	50.3 ± 1.83$^-$	48.3 ± 1.26***	52.7 ± 2.72*	50.4 ± 0.90
	2-yr.	50.5 ± 1.73$^-$	49.2 ± 0.55***	50.1 ± 1.04**	50.8 ± 1.19$^-$	48.0 ± 8.01$^-$	54.2 ± 2.44**	51.6 ± 0.43
Picea abies								
Fresh mass of 100 needles, g	1-yr.	0.71 ± 0.23$^-$	0.60 ± 0.09$^-$	0.51 ± 0.08**	0.56 ± 0.20$^-$	0.66 ± 0.15$^-$	0.59 ± 0.17$^-$	0.68 ± 0.14
	2-yr.	0.81 ± 0.20$^-$	0.58 ± 0.10$^-$	0.48 ± 0.13*	0.52 ± 0.11$^-$	0.71 ± 0.27$^-$	0.63 ± 0.21$^-$	0.63 ± 0.18
Dry mass of 100 needles, g	1-yr.	0.31 ± 0.11$^-$	0.26 ± 0.04$^-$	0.22 ± 0.03**	0.24 ± 0.06*	0.30 ± 0.07$^-$	0.25 ± 0.07$^-$	0.31 ± 0.08
	2-yr.	0.39 ± 0.11$^-$	0.27 ± 0.04$^-$	0.24 ± 0.06*	0.26 ± 0.06$^-$	0.35 ± 0.13$^-$	0.30 ± 0.10$^-$	0.31 ± 0.09
Dry matter, %	1-yr.	43.9 ± 1.18$^-$	43.9 ± 3.09$^-$	44.9 ± 7.42$^-$	43.7 ± 6.08$^-$	45.2 ± 1.90$^-$	43.0 ± 2.08$^-$	44.9 ± 2.56
	2-yr.	48.3 ± 2.19$^-$	46.2 ± 1.87**	49.7 ±1.75$^-$	49.9 ± 1.24$^-$	49.0 ± 0.70$^-$	47.1 ± 1.36**	48.9 ± 1.29

Notes: 1-yr. – one-year-old needles formed in 1996; 2-yr. – two-year-old needles formed in 1995.
$^-$ $p > 0.05$; * $p \le 0.05$; ** $p \le 0.01$; *** $p \le 0.001$.

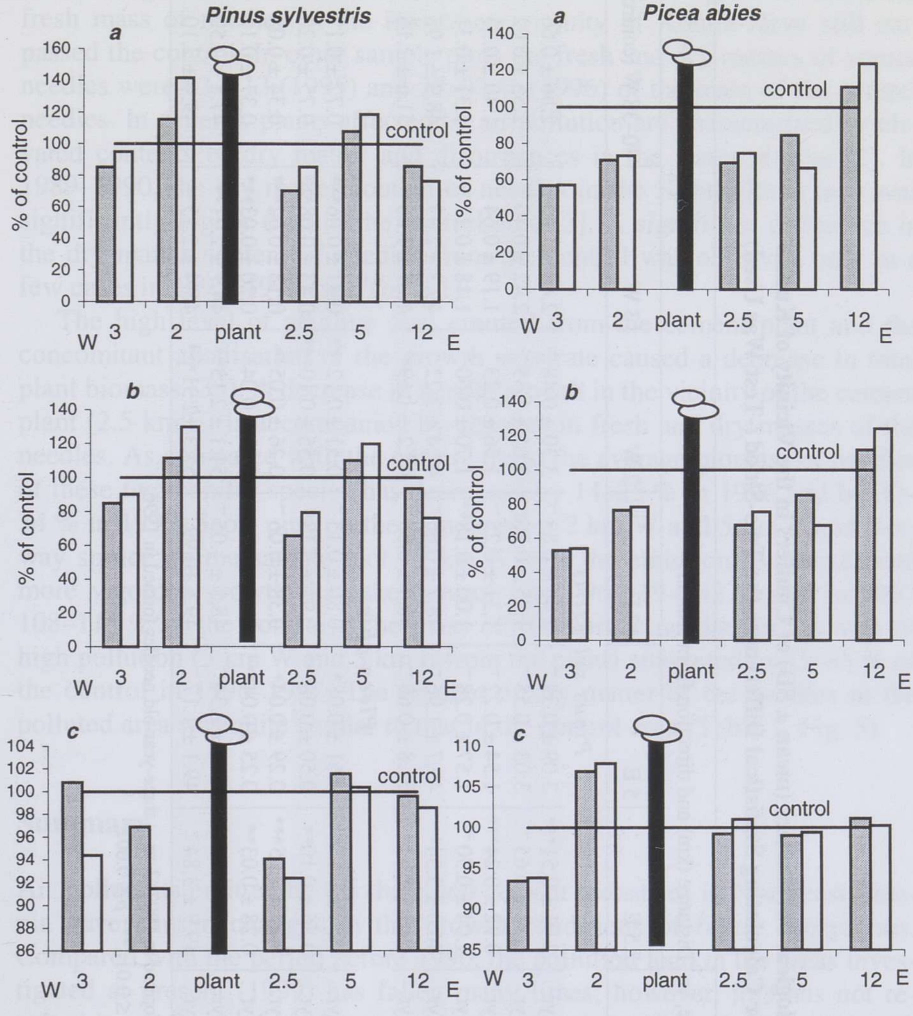


Fig. 5. Fresh (*a*) and dry (*b*) mass, and dry matter content (*c*) of needles of different ages in different directions and at various distances from Kunda cement plant (% of control). 2-yr. – two-year-old needles formed in 1997; 3-yr. – three-year-old needles formed in 1996

Both in 1995 and 1996, the fresh and dry mass of Scots pine and Norway spruce needles from the sampling plot, situated 15 km SE from Kohtla-Järve in the influence zone of Ahtme power plant, were significantly lower than the control (fresh mass 61–76 % and dry mass 60–77 % of the control). In 1989–1990 and in 1994, the fresh mass of spruce needles collected near Kohtla-Järve (0.5 km NE) accounted for 63 and 86 % of the fresh mass of the control needles, respectively [3]. The needles formed in 1995 were up to 29 % heavier than those from Lahemaa (see Table 1, Fig. 4).

Table 2. **Deviation in Morphological Parameters (mean ± SD) of Conifers in the Vicinity of Kunda and at the Control in 1999 (for Calculating Statistical Differences the Two-Sided *t*-Test Was Used)**

Parameter ($n = 10$)	Age, yr	Distance (km) and direction from Kunda					
		2.5 E	5 E	12 E	2 W	3 W	30–38 W (control)
Pinus sylvestris							
Fresh mass of 100 needles, g	2-yr.	2.01 ± 0.51***	3.09 ± 0.39^{-}	2.46 ± 0.83^{-}	3.30 ± 0.48^{-}	2.44 ± 1.40^{-}	2.86 ± 0.66
	3-yr.	2.18 ± 0.65^{-}	3.03 ± 0.55^{-}	1.95 ± 0.36*	3.61 ± 1,35^{-}	2.42 ± 0.40^{-}	2.55 ± 0.62
Dry mass of 100 needles, g	2-yr.	0.92 ± 0.24***	1.54 ± 0.22^{-}	1,31 ± 0.25^{-}	1.56 ± 0.23^{-}	1.19 ± 0.64^{-}	1.40 ± 0.33
	3-yr.	1.04 ± 0.30^{-}	1.57 ± 0.28^{-}	1.00 ± 0.18**	1.70 ± 0.60^{-}	1.18 ± 0.21^{-}	1.31 ± 0.31
Dry matter, %	2-yr.	46.0 ± 3.34*	49.7 ± 1.96^{-}	48.7 ± 0.66^{-}	47.4 ± 2.22^{-}	49.3 ± 2.39^{-}	48.9 ± 1.98
	3-yr.	47.7 ± 3.73*	51.8 ± 0.69^{-}	50.9 ± 0.55^{-}	47.2 ± 1.69*	48.7 ± 1.26**	51.6 ± 2.16
Picea abies							
Fresh mass of 100 needles, g	2-yr.	0.50 ± 0.13***	0.61 ± 0.08*	0.80 ± 0.13^{-}	0.51 ± 0.12***	0.41 ± 0.06***	0.72 ± 0.17
	3-yr.	0.56 ± 0.10**	0.50 ± 0.08***	0.93 ± 0.18*	0.55 ± 0.10***	0.43 ± 0.08***	0.75 ± 0.20
Dry mass of 100 needles, g	2-yr.	0.23 ± 0.05***	0.29 ± 0.04*	0.38 ± 0.06^{-}	0.26 ± 0.05**	0.18 ± 0.02***	0.34 ± 0.08
	3-yr.	0.28 ± 0.03**	0.25 ± 0.04***	0.46 ± 0.09*	0.29 ± 0.04*	0.20 ± 0.03***	0.37 ± 0.11
Dry matter, %	2-yr.	47.4 ± 3.07^{-}	47.3 ± 2.34^{-}	48.3 ± 1.05^{-}	51.1 ± 2.13**	44.7 ± 2.21**	47.8 ± 2.50
	3-yr.	49.9 ± 3.84^{-}	49.1 ± 1.17^{-}	49.5 ±0.99^{-}	53.3 ±2.31***	46.4 ± 2.35**	49.4 ± 2.10

Notes: 2-yr. – two-year-old needles formed in 1997; 3-yr. – three-year-old needles formed in 1996.
$^{-}$ $p > 0.05$; * $p \leq 0.05$; ** $p \leq 0.01$; *** $p \leq 0.001$.

Although the difference from the control was not so great in 1996, the fresh mass of needles in the immediate vicinity of Kohtla-Järve still surpassed the control. In other sample plots the fresh and dry masses of spruce needles were 83–113 (1995) and 77–97 % (1996) of the mass of the control needles. In general, plants affected by air pollution are characterised by elevated contents of dry matter and disturbances in the water regime [2]. In 1989–1990, the dry matter content of needles in the Kohtla-Järve area was significantly higher than in the control area [3]. A significant difference in the dry matter content of needles from the control was observed only in a few cases in 1995–1996 (see Table 1).

The high level of alkaline dust emitted from the cement plant and the concomitant alkalisation of the growth substrate caused a decrease in total plant biomass [33]. A decrease in needle growth in the vicinity of the cement plant (2.5 km E) is accompanied by changes in fresh and dry masses of the needles. As compared with the control trees, the average biomass of needles of these two conifer species has decreased by 14–25 % in 1996 and by 30–34 % in 1997. Scots pine on the sample plots 2 km W and 5 km E and Norway spruce on the sample plot 12 km E from the emission source showed more vigorous growth than the control (in 1996 119–142 % and in 1997 108–115 % of the control). The mass of 100 spruce needles in the area of high pollution (3 km W and 5 km E from the plant) amounted to 53–85 % of the control in 1996–1997. The content of dry matter of the needles in the polluted area was quite similar to that in the control area (Table 2, Fig. 5).

Summary

Air pollutants emitted by oil shale and cement industries in Northeast Estonia have caused changes in the growth conditions of forest ecosystems. Compared with the period before 1990, the pollution load in the areas investigated at present (1999) has fallen many times; however, this has not resulted in an essential improvement in the growth conditions of trees. Application of morphometric parameters gives versatile information on damage due to pollution.

The Scots pine and Norway spruce trees studied were found to be sensitive towards oil-shale fly ash and cement dust and alkalised environment; however, the impact on different morphological parameters is different. Our results showed that changes in the fresh and dry mass of needles of Norway spruce are a suitable indicator of pollution emitted by the cement industry as these revealed a clear negative trend in sample plots closer to emission sources.

No clear trend was observed in the mass of the needles of Norway spruce around Kohtla-Järve; the data were sometimes contradictory, requiring further monitoring. As compared to the control, the strongest inhibition of needle biomass was revealed in the sample plots situated 15 km SE from

Kohtla-Järve fertiliser plant and 2.5 km E from the Kunda cement plant. Needles of Scots pine on the sample plot 2 km W and Norway spruce 12 km E from the cement factory showed more vigorous biomass than the control. Differently from the pine, the fresh and dry mass of spruce needles were significantly reduced within up to 2-3 km W and 5 km E from the cement plant.

Together with 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. Compared with the control, in spruce needles it decreased with distance from the emission sources.

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