Changes in the Allocation of Nutrients and Biomass in Scots Pine (*Pinus sylvestris* L.) Canopy in an Area of Cement Industry in Northeast Estonia

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Abstract

The study was carried out in selected stands in the Northeast Estonian industrial region (cement dust pollution). A total of 4 *Pinus sylvestris* stands with similar natural characteristics were studied in 2005. Emissions from the cement plant have decreased several times during the last ten years. The long-term impact of cement dust and alkalisation of the environment complicate mineral nutrition, disbalance the mineral composition and influence the growth of trees. Vertical gradients of nutrients and their ratios in canopies varied depending on the alkalisation of soil and the level of dust pollution in the past. Reduced N and increased K, Ca and Mg concentrations in needles were characteristic of pines in the heavily polluted areas. The stimulation growth of pine needles 2.5 and 5 km E of the cement plant may be a sign of a positive effect of reduced doses of cement dust in soil.

Key words: alkaline dust pollution, canopy layer, needles, nutrients, *Pinus sylvestris*, soil

Introduction

Numerous investigations describe damages of large forest areas in many industrial countries in Europe and Northern America by acidic air pollution at the end of the 20th century (Smith 1990, Staaf and Tyler 1995, Hartling and Schulz 1998). Research into the impact of different alkaline types of air pollution on forests has never drawn so much attention as that of SO_2, NO_x or O_3. In general the production of building materials, open-cast mining and quarrying, metallurgical engineering and chemical industry are among the important emitters of solid pollutants in the form of alkaline dusts or ashes. Deposition of alkaline solid air pollutants on the ground results in serious alkalisation of the environment.

Problems of forest damages caused by alkaline types of pollution, however, are not completely understood and interpreted. Still, many authors have found deviations in plant metabolism and physiology (Manning 1971, Auclair 1977, Manning and Feder 1980, Lal and Ambasht 1982, Mandre 1995, Klõšeiko 2003, Skuodiene 2005) and in the growth and bioproduction (Gluch 1980, Jäger and Klein 1980, Rauk 1995, Ots 2002) caused by alkaline dust pollution.

Cement dust emitted from the cement and building materials production is one of the characteristic alkaline solid industrial pollutants. Yet, responses of plants to cement dust impacts described in the literature are contradictory. Lichen communities were shown to be very sensitive to dust pollution (Farmer 1993, Nilson 1995, Loppi and Pirintsos 2000, Branquinho et al. 2008). An increase in the amount of certain pollutants and changes of growing conditions in the polluted surroundings affect forests directly, cause various morphological, physiological and chemical changes, the results of which is the slow down of tree growth, decline of its condition or even death (Petraš et al. 1993, Ots 2002, Juknys et al. 2003, Erlickyte and Vitas 2008). Migahid and El-Darier (1995) are of the opinion that cement dust has little influence on the growth of trees while Ots and Rauk (1999) argued that the inhibition of the radial increment and height growth of *Pinus sylvestris* and *Picea abies* is significant. In any case the differences in the reactions of tree species to cement dust influence depend on several species-specific peculiarities, pollution load and its chemical composition, climatic conditions etc. (Mandre et al. 1999, Pärn 2002, Erlickyte and Vitas 2008).

One of the major producers of industrial alkaline dust pollution in Estonia is the cement plant in Kunda, Lääne-Viru County. During 1987–93 the dust fall-out at a distance of 1–1.5 km from the emission source reached yearly 1600–2700 g m^{-2}, depending on the direction and velocity of winds, on the size of dust particles and on the intensity of industrial production (Mandre 1995). Areas surrounding the cement plant have suffered alkalisation of the soil and subsoil water, snow melt and rain water for a long time. In the vicinity of the pollution source the pH of upper layers of soils may be extremely high (pH 7.8–8.1) (Annuka and Mandre 1995, Mandre and Ots 1999, Ots 2002).

From the ecophysiological point of view cement dust has relatively large amounts of mineral elements (K, Mg, Ca and Mn), which have entered the environment and have followed biogeochemical cycles and may cause important deviations in the growth and physiology of trees.

The aim of the present studies was to clarify the status of the forest soil and *Pinus sylvestris* in the area affected by cement dust pollution for over 40 years. Analysis of the chemical composition of different soil horizons and morphological and chemical characteristics of assimilative organs of trees will serve as a foundation for understanding the persistence of alkaline soil reaction. Although nutrient uptake and translocation mechanisms in conifers have not been explored extensively, the limited evidence suggests that partitioning of nutrients among various tree compartments is of importance in the formation of canopy architecture and biomass production (Rook 1991, Schaedle 1991). Nutrient allocation in trees can be influenced by a variety of internal and external factors and the question whether a particular organ is adequately supplied with assimilates, and can therefore fully realize its growth potential, may not have a simple answer (Ingestad and Ågren 1988, Ingestad 1991). Integrated evaluation of chemical characteristics and morphological parameters of the soil and trees can show the state of and individual tree or a forest site.

The findings help us understand the dynamics in the system tree-growth environment and prognosticate possible trends in the territory of cement industry and Northeast Estonian forests in general.

Material and methods

Study area

Investigations were carried out on a territory affected for over 40 years by a cement plant in the town of Kunda (59º30´ N, 26º32´ E), Northeast Estonia. Considering the direction of prevailing winds (from south-west and south) and spreading of pollutants, the sample plots were located on the transect parallel to the North Estonian coastline in an area rich in *Pinus sylvestris* stands. In the selection of sample plots we proceeded from the principle of analogy of geographical and silvicultural characteristics. It was important that climatic and edaphic conditions as well as the stand parameters of the plots should be similar. The investigation was performed on a mixed *Pinus sylvestris* stand of *Myrtillus–Oxalis* site type according to the system presented by Lõhmus (2004) in the vicinity of a cement plant and in the control area. The selected stands (0.05 ha) were similar as to their crop density, quality class, age, site type and species composition of stands (0.7–0.8 crop density, II quality class, average density or sparse understorey, 75–85-year-old pine stands). This made comparison of the results possible as the effect of numerous external (climatic, phytocoenotic, anthropogenic etc.) and internal (metabolism, hereditary properties etc.) factors affecting the growth of conifers in addition to pollution could be reduced. Following the FAO System (1990) the soils in the investigated area are Gleyic Podzols (Lkg) on sand, one of the most acidic soils in Estonia.

For comparison, the control sample plot was selected in similar climatic and edaphic conditions on a relatively unpolluted area in Lahemaa National Park (59º31´ N, 26º00´ E) at a distance of about 38 km W (plot 1) of the cement plant, opposite to prevailing winds (Figure 1). This area is considered relatively free of pollution on the basis of analyses of precipitation, bioindications by mosses and lichens as well as studies on biochemical indicators of air pollution (Kannukene 1995, Mandre and Tuulmets 1995, Nilson 1995). The sample plots were situated at distances of 3 km W (plot 2) and in the direction of prevailing winds – 2.5 km E (plot 3) and 5 km E (plot 4) of the emission centre (Figure 1).

Climatically the areas investigated belong to the mixed-forest subregion of the Atlantic-continental re-

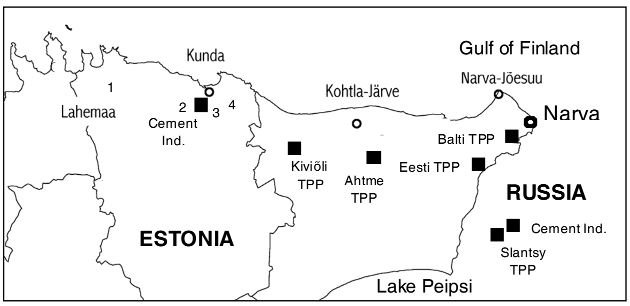

Figure 1. Schematic map of the location of sample plots (numbers 1–4) and pollution sources in the study area

gion, where the influence of the Baltic Sea is strongly felt. The mean annual temperature is 4.9°C, annual amount of precipitation 550 mm and dominating winds blow from the southwest at a mean velocity of 5.2 m s^{-1} (data from the Estonian Meteorological and Hydrological Institute, http://www.emhi.ee).

The production of cement in Kunda started in 1871. After the last reconstruction in 1960, cement production has rapidly increased, fluctuating between 0.5 and 1.2 million t yr^{-1}. The emission from the cement plant in 1987–1993 contained 87–91% technological dust and 9–13% gaseous pollutants (SO_2, NO_x, CO etc.). For a long time the main damaging factor to trees in the investigation area was apparently a high level of alkaline dust emission from cement plant. It contains many components, among which the following are predominant: 40–50% CaO; 12–17% SiO_2; 6–9% K_2O; 4–8% SO_3; 3–5% Al_2O_3; 2–4% MgO; but also Fe, Mn, Zn, Cu, B, etc. occur. The water solution of dust from electric filters had pH values from 12.3 to 12.7 (Mandre 1995).

The dust emission from the cement plant was extremely high in 1990–1992 being 80–100 kt per year (Keskkond '90 1991, Estonian Environment 1991 1991, Estonian Environment 1995 1996) (Figure 2). In 1993–1996 the emission of cement dust from the plant decreased notably thanks to the installation of efficient filters and it was 224 t per year in 2005 which is lower than the permitted quantity.

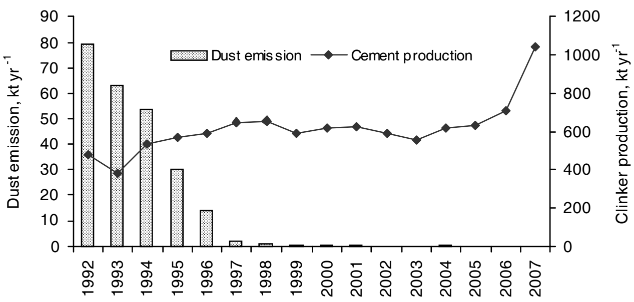

Figure 2. Emission of dust into the atmosphere of the cement plant and cement production in Kunda, Northeast Estonia

However, the high level of dust pollution for over 40 years has brought about alkalisation and serious changes in the chemical composition of the soil, groundwater and precipitation in this area (Annuka 1994, Kört and Truuts 1997, Mandre et al. 1999). In the vicinity of the cement plant the concentrations of Ca, K, Mg, S and other elements predominating in the dust are extremely high in the upper layers of the soil and in precipitation. The decrease in the average pH of precipitation in Kunda in 1996 was very small compared to the previous years (7.6 in 1996, 7.7 in 1995, 7.3 in 1994); however, several components of pollution (Ca, K, Mg, SO_4) decreased significantly (Kört and Truuts 1997). According to the Kunda station, in the vegetation period of 1999–2004 the average pH of precipitation was pH 6.8, varying from 4.8 to 7.9 (weakly acidic to strongly alkaline precipitation water) (http://www.stat.ee). The all-Estonian monitoring by the Estonian Environmental Research Centre shows that precipitation is most heavily polluted in Kunda (Otsa et al. 2005). According to the Centre's data the research conducted in the 1st quarter of 2003 showed that in Kunda (precipitation pH = 6.2) the concentration of Ca was 56 times higher than in the precipitation at Lahemaa (pH = 4.4) while that of K was 30 times higher.

At present, the amounts of dust emitted are very small as the data by the plant show; however, because of the large amounts of pollutants emitted for decades alkalisation of the environment is still an important factor affecting tree growth in the influence zone of the cement plant.

Chemical analyses of soil

For the collection of the soil samples soil pits were dug and described according to FAO (1990). Different soil horizons were distinguished to the depth of 1 m from the surface and chemical analyses of different soil horizons were carried out. Representative samples for chemical analyses (n = 36) were taken from each genetic horizon, air dried, crushed and sieved on a 2 mm sieve in September 2005.

The nutrient status of the soil was determined in the Laboratory of Plant Biochemistry of the Estonian University of Life Sciences and Laboratory of the Environmental Chemistry Department of the Estonian Environmental Research Centre. Standard methods of soil analysis were used: the content of P and K was determined by the Egner–Riehm double lactate method and that of Ca and Mg by Egner–Riehm–Domingo ammonium acetate–lactate method (ISO/11260 1995) with an atomic absorption analyser AAA-1N (Carl Zeiss, Jena). Total N was determined by the Kjeldahl method (ISO/11261 1995) and the pH of the soil was measured as the acidity in H_2O (ISO/10390 1994).

Samples from analyzed trees

In September 2005 three dominant pine trees with a similar habitus of canopy were selected from each sample plot for analysis so that they would represent evenly the diameter classes (25–27 cm at breast height) of the stand in the sample plots and the average trees within each sample plot. Trees were felled and the canopy of each tree was divided into three equal horizontal layers. Altogether, 12 model trees were felled and samples were collected from each tree for analysis of the chemical composition and morphological measurements of needles at upper, middle and lower layers of canopy (Figure 3). Taking into consideration individual variation of trees, samples of the current year and one-year-old pine needles were taken from all sides of the trees to get an average sample for analysis.

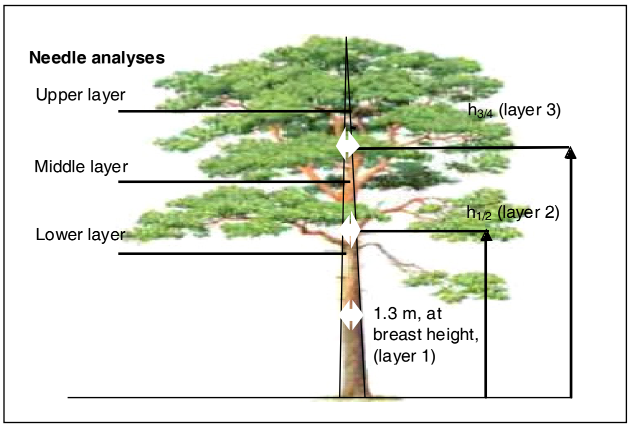

Figure 3. Sampling of the material for analysis of the chemical composition and morphological measurements of needles

Sample branches for morphological analyses were collected from the different layers of pine canopies and from sides in September 2005 that is after the needles and shoots had attained full length. The length (cm) of the one-year-old needles (n = 28–300) was measured; the dry mass (g) of 100 needles (n = 28–30) was weighed and the density of needles on shoots (number of needles on a shoot : length of shoot, No. cm^{-1}) (n = 28–30) were calculated and the results were compared with the earlier results (length of needles, dry mass of 100 needles and density of needles on shoots) from 1998 (Ots 2002). For dry mass determination the needles were stored in a thermostat at 70ºC until they reached a constant mass.

Chemical analysis of trees

Carefully cleaned needles were oven-dried at 70 ºC to stop metabolic activity (Wilde et al. 1979, Landis 1985) and ground in September–October 2005. After grinding, macronutrients (N, P, K, Ca, Mg) were analysed in 1–2 g of dried plant material. The methods used for analyses are certified by international ring-tests including FATAS 2002, AACC, European Grain Network, etc. Concentrations of metals (Ca, K, Mg) were determined using an atom-adsorption analyser AAA-1N (Carl Zeiss, Jena). The method of Kjeldahl was used to measure N, P was extracted with vanadium molybdate yellow complex and C was measured by dry combustion to CO_2 according to ISO 10694 (1995).

Statistical tests

Averages of analyses and standard errors of the mean (mean ± SE) for all results were calculated. All statistical effects are considered significant at $P < 0.05$. Correlation analysis (r, Pearson correlation coefficient) was used to estimate the statistical significance of the dependence between mineral elements in trees and in the upper 30 cm horizon of soil. Analysis of variance was used to test for the general differences between the mean mineral elements in the soil and parameters of trees from different sample plots (ANOVA, two-way analysis of variance). When the data did not follow the normal distribution, or when there occurred an inhomogeneity of group variance, the non-parametric analysis of variance was used. Statistical calculations were performed with Systat 10 (SPSS Inc., USA), Statistica 7 and Excel (Microsoft Corp., USA).

Results

Soils in the area of cement dust pollution and nutrients partitioning in soil

In the area under the strongest dust load sample plots 3 km W, 2.5 km E and 5 km E, the total contents Ca, Mg and K in soil litter (O) and humus (A) horizons exceed considerably those in the soils of the control sample plot (Figure 4). The Ca contents was higher in the humus horizons with the highest level detected in the sample plot 2.5 km E. The mean total content of Ca in the Estonian soils is 8,600 mg kg^{-1} (Petersell et al. 1997). The Ca concentrations in the mineral horizons of soils were low, increasing slightly in the illuvial (B) horizon.

In the soils of the control plot (38 km W), the contents of Ca, Mg and K in the humus horizon were smaller compared to the litter horizon. Large amounts of these elements are still bound in soils that formerly received high loads of cement dust deposition. Elevated levels of Ca and Mg, less in the case of K, in the humus horizon of soils on the dust polluted sample plots, especially on the 2.5 km E sample plot, was detected. In natural, unpolluted forest podzols (i.e. control sample plot), the content of basic cations was low. In our case this was expressed as a high P content of the litter, podzol and illuvial horizons on this sample plot (Figure 4). The exceptionally high P content in the illuvial horizon on the 3 km W sample plot may be connected with the above-mentioned general variability in the P distribution in soils.

Because of the negligible content of N in cement dust, its concentration in the soil of heavily polluted areas did not differ much from the N concentration in the soil of the control sample plot. A certain variability in the N content between the sample plots was visible in the litter horizon (Figure 4). On the polluted sample plots the N concentrations in this horizon were lower compared to the unpolluted control, however. In the mineral horizons with a small content of organic matter, the N content was close to zero on all sample plots.

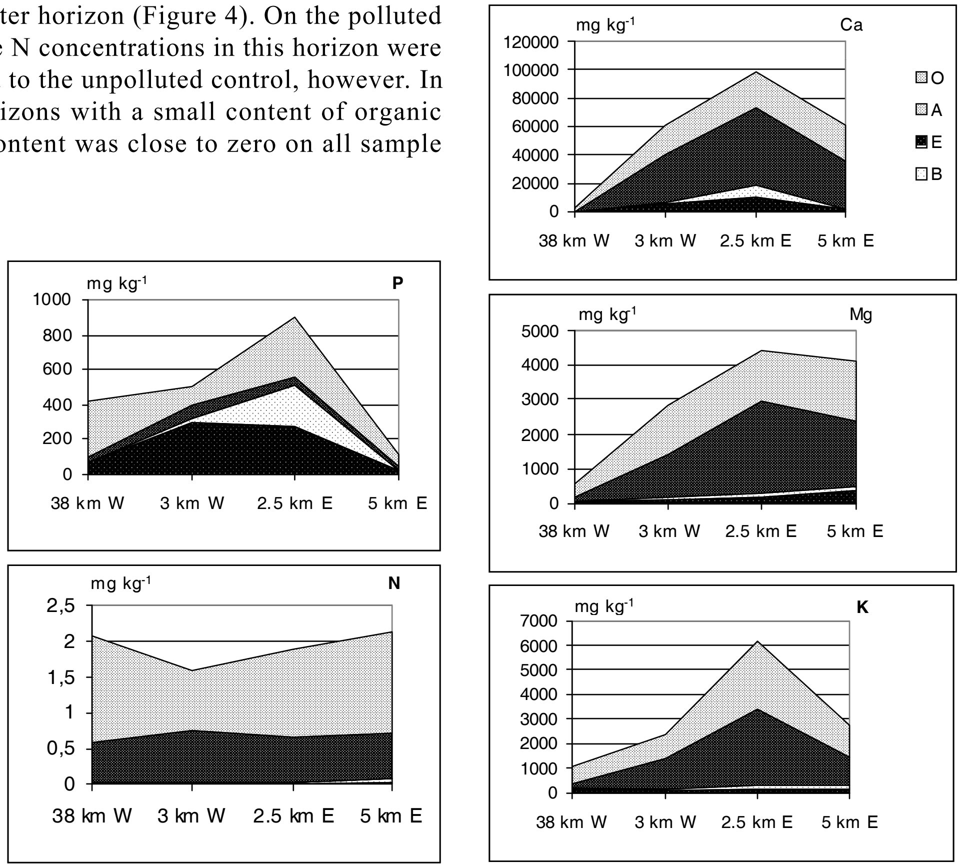

Figure 4. The content of some chemical elements in soil horizons in different zones of cement dust pollution

Nutrients in needles

The concentration of N in the *Pinus sylvestris* needles from the previously heavily polluted area (2.5 km E) was on average 12% and that of P 20% lower, while those of Ca, K and Mg were respectively by 25%, 24% and 11% higher as compared with the control trees (Table 1). Needle C concentrations in this territory did not differ from the control (Table 1).

Results of our study showed that lower-canopy needles had a higher N in the unpolluted control area, but in the alkalised soils N concentrations were higher in the upper layer of the canopy (Table 1). Apart from slight differences of N concentrations between canopy layers, we found that N scaled positively also with Mg (r = 0.51), Ca (r = 0.52) and K (r = 0.68) in the needles from the polluted areas. The allocation of P in different parts of the canopy showed trends to higher concentrations in the sample plot 2.5 km E and to lower concentrations in the moderately polluted sample plots (3 km W, 5 km E) (Table 1).

Comparison of trends of Ca, Mg and K allocation in different layers of *Pinus sylvestris* canopies showed, that in the polluted areas Ca was more abundant in the needles of the upper canopy layer, while needles in the unpolluted area had the highest concentration of Ca in the lowermost layer. Magnesium was found in excess in all polluted sample plots and in the lower part of canopies if compared to the control. Potassium was accumulated predominantly in the needles of the lower part of the canopy. However, in the most polluted area (2.5 km E) excess of Ca, K and Mg in pine needles was detected (Table 1).

The ratio P/N in polluted areas did not differ from control, only on the sample plot 2.5 km E it was higher than in control (Figure 5). The ratios N/Ca, N/Mg and N/K were significantly lower (by 20%, 28% and 20% respectively) than control, while the ratios Ca/Mg, K/Mg and K/Ca were higher (by 7–9%).

Needle morphology in different canopy layers

The balance disturbed of nutrients in plants, inhibits the growth of the trees. The length growth of needles has been proved to be one of the best indicators of the effect of cement dust pollution (Manning and Feder 1980, Braniewski and Chrzanowska 1988, Ots 2002). The length growth of needles is greater in the

Table 1. Percentages of mineral elements in needles of *Pinus sylvestris* in sample plots at different distances from the emission centre (n = 18). Lower layer – 1, middle layer – 2, upper layer – 3; DM – dry mass

Sample plot, distance and direction from the cement plant	Layer of canopy	Ca		K		Mg		P		N		C	
		% of DM	% of control	% of DM	% of control	% of DM	% of control	% of DM	% of control	% of DM	% of control	% of DM	% of control
Revoja, 38 km W control	1	0.36	100.0	0.62	100.0	0.11	100.0	0.17	100.0	1.67	100.0	49.51	100.0
	2	0.38	100.0	0.57	100.0	0.12	100.0	0.16	100.0	1.67	100.0	49.81	100.0
	3	0.30	100.0	0.50	100.0	0.16	100.0	0.16	100.0	1.59	100.0	50.02	100.0
Toolse 3 km W	1	0.25	68.5	0.58	94.3	0.12	107.3	0.15	95.8	1.41	85.9	49.53	100.0
	2	0.30	79.4	0.49	86.9	0.13	110.5	0.14	88.8	1.43	87.1	50.01	100.4
	3	0.32	109.1	0.40	79.5	0.16	101.1	0.15	95.0	1.51	94.8	49.88	99.7
Kunda 2.5 km W	1	0.40	113.4	0.72	116.3	0.13	118.0	0.18	109.0	1.43	85.6	49.46	99.9
	2	0.44	118.9	0.79	139.8	0.15	123.3	0.18	112.0	1.52	85.0	49.95	100.3
	3	0.46	160.2	0.59	115.5	0.16	96.6	0.17	108.0	1.46	92.0	50.34	100.6
Malla 5 km E	1	0.25	70.1	0.59	97.0	0.14	121.9	0.16	93.0	1.52	91.0	50.23	101.5
	2	0.27	72.0	0.42	74.7	0.16	137.2	0.14	86.0	1.60	95.8	50.88	102.1
	3	0.29	97.8	0.39	77.2	0.18	111.8	0.14	88.0	1.58	99.4	50.07	100.1

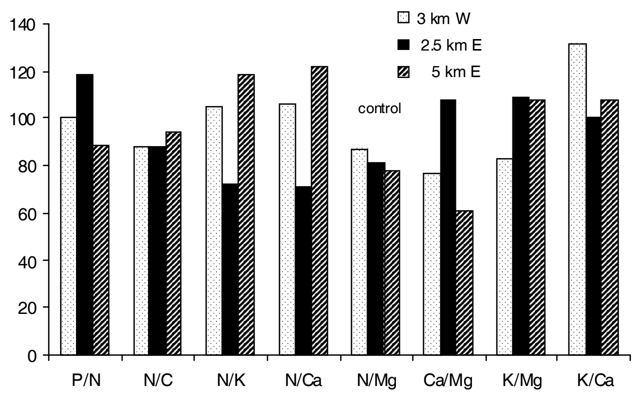

Figure 5. Differences from the control (100%) of nutrient ratios in current-year needles of *Pinus sylvestris* growing at different distances from the emission centre

highest layer of pine canopies, where illumination conditions are better than in the lower layers (Figure 6). Pines growing 5 km E of the pollution source had the longest needles. The greatest differences in needle length in pine canopies were observed in trees growing 3 km W of the cement plant. The difference between the length growth of needles in the upper layer and the lowest layer was 1.1–1.4 times. The average length of needles measured in the lowest layer in the canopy in the influence zone of the cement plant was 4.8–5.5 cm (in the control area 5.3 cm), in the middle layer 5.2–5.7 cm (control 5.6 cm) and in the upper layer 6.1–6.8 cm (control 6.5 cm) (Figure 6). After the significant fall in the pollution load since 1996 the length growth of pine needles intensified around the Kunda cement plant, at the same time no changes occurred in the length growth of needles in the control area (Ots 2002). As compared to the data from 1998, the length growth of pine needles had improved, especially 2.5 and 5 km E from the cement plant, needles being respectively 1.5 cm and 1.1 cm longer than 6 years ago.

It has been established that changes in needle dry mass are one of the secondary consequences of technogenic dust pollution (Lim and Cousens 1986, Braniewski and Chrzanowska 1988, Gigauri et al. 1992). The dry mass of 100 needles was statistically significantly larger ($P < 0.001$) in the upper layer of the canopy than in the lower layers, being 1.5–2.3 greater than in the lowest layer. The most homogeneous needles over the whole canopy were established in the sample plot 2.5 km E of the cement plant (Figure 6). A statistically significant difference from the dry mass of needles from the control plot was found in the case of needles from the upper layer of trees growing 2.5 km E of the cement plant ($P < 0.05$). As compared to the data from 1998, the dry mass of pine needles had increased, especially 2.5 and 5 km E of the cement plant, being respectively 1.8 and 1.6 times greater than 6 years ago.

Data collected from mature pine stands in the influence zone of the cement plant indicate that in the sample plots located east of the cement plant the density of needles on shoots is highest in the middle layer of the canopy but on the sample plots west of the plant the needle density is highest in the lowest canopy layer and lowest in the upper layer (Figure 6). Comparison between the data from all plots in 1998 and 2004 showed that the decrease in the pollution load in the influence zone of the cement plant was accompanied with an increasing density of needles on pine shoots in mature stands.

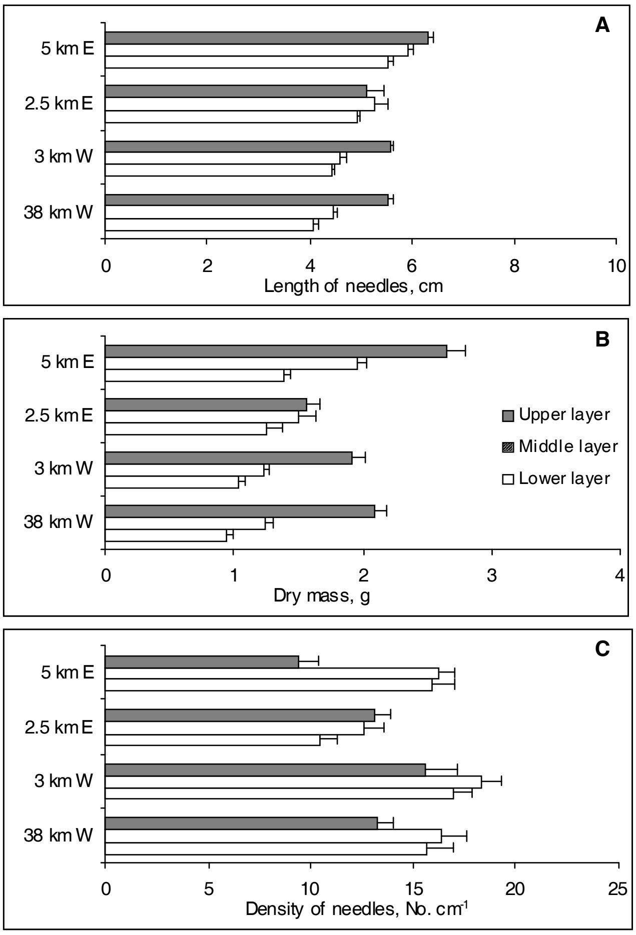

Figure 6. Length growth (A), dry mass of 100 needles (B) and density of needles on the shoot (C) of one-year-old needles (mean±SE) in different layers of *Pinus sylvestris* crowns at different distances (km) and directions from the cement plant and in the control area in 2005

Discussion and conclusions

Significant changes in the content of different elements in soils have been reported in regions under the influence of the cement industry (Asubiojo et al. 1991, Klose and Makeschin 2004, Świercz 2006, Zerrouqi et al. 2008). The present study revealed that also Gleyic Podzols in mixed pine–spruce stands were enriched with Ca, Mg, K and P in the vicinity of the cement plant in Kunda. As compared to the data from 1991 (Annuka and Mandre 1995), a decline in the pH of the upper 10 cm layer by about 0.4 units and in Ca, Mg, and K pools in soils of the sample plots closest to the plant had occurred.

Commonly dust and fly-ash emissions cause an accumulation of mineral constituents in surface horizons (Armolaitis et al. 1996, Klose and Makeschin 2004). Our results showed that the influence of atmospheric pollutants is not limited to upper horizons. Although the content of Ca, Mg and K decreases rapidly towards the lower horizons of soils on polluted sample plots, their contents in these soil horizons are somewhat higher than in the control sample plot, especially for Ca. Similarly to soils in the vicinity of Kunda, a significant impact of dust emitted by a cement plant on soil was observed in India, especially on the two upper horizons of soil, at a distance of 2–4 km E of the plant, where the concentration of mobile $CaCO_3$ in the soil had notably increased (Chitralekha and Dhakshinamoorthy 1997, Chitralekha et al. 1997).

Limestone, clay and oil shale, used for cement production, contain very small amounts of biogenic elements such as P and N (Pets 1997). During the process of cement production they volatilise or are removed with waste waters. This may be the main reason for the low concentrations of P in soils of all sample plots with some exceptional cases. The P distribution in the organic horizons of Estonian soils is rather variable being higher and more irregular in the upper layers that contain peat fractions (Petersell et al. 1997). Immediately south of an outcrop of a phosphorite layer in northern Estonia a belt enriched with P occurs. The vicinity of the phosphorite area to the 38 km W control sample plot may have some influence on the P content in the litter horizon of this plot. The sample plot 2.5 km E is situated in the narrow area near the Kunda cement plant characterised by the high P content in soil (Annuka 1994).

Alkalisation of soil inhibits the availability of several nutrients, causing serious deviations in the mineral composition of plants (Marschner 2002). It is known that in alkaline soils of increasing pH, decreasing soil organic matter and increasing Ca concentration, the mobility of P is limited and its deficiency becomes evident due to the formation of calcium phosphates of low solubility as was found in the sample plots 3 km W and 5 km E. Branquinho and colleagues (2008) found that the increase in distance from a cement plant was significantly correlated with the exponential decrease in+ the total concentration of Ca, Fe and Mg.

Our study showed variation in the allocation of nutrients between upper and lower layers of the canopy. The strength and direction of nutrient transport and partitioning in different canopy compartments have been demonstrated to be regulated by source–sink relationships (Helmisaari 1992, Billow et al. 1994, Nieminen and Helmisaari 1996).

In the unpolluted and relatively optimal growth conditions of the control pines, the partitioning of higher amounts of N in the lower layer of the canopy may be explained by higher concentrations of K and

Ca. In the lower canopy layer, the concentration of Ca and Mg in the needles of pines growing 2.5 km E of the cement plant made up respectively 113% and 118% of the control while in the upper layer the respective figures were 160% and 97%.

Trends in the needle P concentration in different layers of the *Pinus sylvestris* canopy in alkaline growth substrate were similar to the control trees and there were no essential differences between needles in different canopy layers. Our results showed, that alkalisation of the environment did not affect C distribution in the canopy of *Pinus sylvestris* and no differences were revealed between different sample plots either.

Many authors stress, that the interaction of different mineral elements in plant tissues and the balance of mineral elements are of great importance in tree growth and survival under stress conditions (Ingestad and Ågren 1988, Shuman 1994, Marschner 2002, Portsmuth et al. 2005). Optimum nutrient ratios for maximum relative growth rates have been found for a range of species (Ingestad 1987, Ericsson 1994, Knecht Billberger 2006). It was ascertained that in optimal conditions the ratios N/P and N/Mg in the needles of conifers are relatively stable all year round (Mandre and Tuulmets 1995). Deviation of the ratio N/P from the optimum may cause drastic changes in plant metabolism (Marschner 2002). Our results show that in the sample plot most heavily affected by cement dust pollution (2.5 km E) the ratios of N in needles to K, Ca and Mg concentrations were significantly smaller than in the control trees while the ratios P/N, Ca/Mg and K/Mg were significantly larger than the control.

Earlier studies showed that changes in primary metabolism and mineral nutrition of trees are accompanied by changes in growth processes (Mandre and Klõšeiko 1997). In recent years a tendency towards a slow recovery of the state of crowns has occurred particularly in the pine and mostly in the less damaged area (Ots 2002). The needles of *Pinus sylvestris* in Estonia are on average 3–7 cm long (Laas 2004). East of the town of Kunda, in the direction of prevailing winds, the average length of pine needles was 4.9–6.3 cm (in the control area 4.1–5.5 cm) while during the years of the highest pollution load the average needle length was 3.6–4.8 cm (control 4.8 cm) (Ots 2002). These findings refer to a stimulating effect of small amounts of nutrient-rich cement dust on needle growth. Stimulation of needle length growth observed in *Pinus sylvestris* 2.5 and 5 km E of the pollution sources is reflected also in their biomass. Braniewski and Chrzanowska (1988) observed that the dust emitted from an aluminium plant reduces the dry mass of 1000 pine needles by 25–27%, but the same amount of dust from the filters of power stations has no significant impact on the dry mass of pine needles and the dust from cement production may increase the needle dry mass.

Irrespective of the stand age, the needle mass in the middle third of the canopy makes up about half of the total mass of needles. However, with increasing age the proportion of needles in the lowest third of the trees decreases continuously while in the highest third of the canopy it increases (Kollist 1970). The same tendency was found in the current study of mature pine stands. In forest monitoring several scientists have acknowledged damages of the crown as an indicator of vitality (Salemaa and Jukola-Sulonen 1990, Ozolinčius 1996, Juknys et al. 2003). Thinning of tree canopies due to air pollution in Northeast Estonia has been fixed for years (Pilt and Õunap 1999). Analysis of absolute data have shown that the density of needles on pine shoots in the immediate vicinity of the cement plant in Kunda is still lower than in the trees in the control area. The higher density of needles on shoots 3 km W of the cement plant is due to the inhibited length growth of the shoots there. Probably the deposition of cement dust containing large amounts of elements needed by trees on sample plots 2.5 and 5 km E of the cement plant had a fertilisation effect on the growth of pine needles.

Although the amount of cement dust emitted into the atmosphere in Northeast Estonia has been constantly decreasing thanks to harmonisation of cement production at Kunda with international environmental requirements, the effect of alkaline dust deposited in the environment is expected to last for years.

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ИЗМЕНЕНИЯ В РАЗДЕЛЕНИЕ ПИТАТЕЛЬНЫХ ВЕЩЕСТВ И БИОМАССЫ В КРОНАХ СОСЕН В РЕГИОНЕ ЦЕМЕНТНОГО ЗАВОДА СЕВЕРНО-ВОСТОЧНОЙ ЭСТОНИИ

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Резюме

Исследования проводились в 2005 году в лесах, расположенных в промышленном регионе (загрязнение окружающей среды цементной пылью) Северно-Восточной Эстонии. Отобранные сосновые леса (4) были одинаковые по природным характеристикам. Выбросы цементной пыли в окружающую среду за последние 10 лет уменьшились в несколько раз. В зоне с наибольшим загрязнением цементной пылью количество Ca, Mg и K в почве было несколко раз выше, чем на контрольном участке. Вертикальный градиент питательных веществ и их связи в кронах сосен варьируют в зависимости от щелочности почв и от загрязнения цементной пылью. Установили, что в зоне наибольшего загрязнения в сосновой хвое снизилось содержание N и повысилось K, Ca и Mg. Стимуляцию роста хвои у сосен, растущих на территории 2,5 км и 5 км от цементного завода на восток, можно отнести к позитивному влиянию небольших количеств цементной пыли.

Ключевые слова: щелочное загрязнение цементной пылью; ярус кроны; иголки; питательные вещества; *Pinus sylvestris,* почва