Effects of the reduction of cement plant pollution on the foliar and bark chemical composition of Scots pine

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The impact of dust emissions from the cement plant of Kunda, north-eastern Estonia, was studied by analysing Scots pine needles and bark. Element concentrations of needles were measured by atomic absorption spectrometry. A scanning electron microscope with an energy dispersive X-ray microanalyzer was used to study dust deposition on the surface of needles and bark. A thick cement dust layer covered plant surfaces, increasing the content of several elements of unwashed needles tenfold as compared with unpolluted sites. The distribution pattern of manganese was totally different to other elements, showing the highest concentrations at the remotest sample sites. Near the plant, pine needles suffered a serious deficiency of Mn. This was due to the unavailability of Mn caused by the alkalising of soil as an impact of dust pollutants. The renovation programme was aimed to reduce dust emissions by 97% over five years. Dust pollutants on pine needles and bark decreased remarkably. However, considerable dust deposition could be observed two years after the renovation. This was caused by the still continuing dust emissions (over 1000 tonnes y^{-1}) and by re-emissions of very large dust masses formerly deposited. The results gave indirect evidence that the recovery of the soil was slow.

Introduction

During the last decades the dust emissions of the cement plant of Kunda Nordic Cement Company in north-eastern Estonia have been permanently very high, rising in some years above 100 000 t yr^{-1}. The dust pollution has caused many problems relating to the environment,

health, hygiene, economy and aesthetics.

The dust of the Kunda cement plant is strongly alkaline. The pH of the snow melt may rise even over 12 in the vicinity of the Kunda plants, and the rainwater is alkalised, with a pH of 7.3–7.1 as far as 10–12 km from the cement plants (Tuulmets 1995). At the same distance, the concentrations of mainly Ca^{2+}, and also

Fig. 1. The location of Kunda in northeastern Estonia on the southern coast of the Gulf of Finland.

K^+ and Mg^{2+}, in the rainwater have increased, as well as the concentrations of sulphuric compounds.

Similarly, the air pollutants change soil and subsoil water pH and element concentration. The soil of the polluted area was in many places seriously alkalised, causing many ecological problems, one of which was the insolubility of manganese (Annuka and Mandre 1995). Therefore, Mn is not available as a plant nutrient, resulting in an exceptionally acute Mn deficiency in conifer needles (Mandre 1995b).

Many physiological and biochemical responses of plants to cement dust pollution have been observed (Mandre 1995b, Mandre 1995c, Mandre and Tuulmets 1995, Rauk 1995a, 1995b, Mandre and Ots 1995, Tohver 1995, Iqbal and Shafiq 1998, Bacic *et al.* 1999), as well as the effects on tree and herbaceous plant communities (Annuka 1995), epiphytic lichens (Nilson 1995) and bryophytes (Kannukene 1995). This area is characterized by easily observable indicator species. One of them is the green alga *Trentepohlia umbrina* (Chlorophyceae, Chaetophorales), which grows on pine bark only under alkaline conditions, coloring the trunks red. Another is the epiphytic lichen *Xanthoria parietina* (L.) Th. Fr., which normally grows on the aspen (*Populus tremula* (L.)), but under alkalised conditions it is common on all trees and bushes, including Scots pine (Haapala *et al.* 1996).

During 1993–1997 all the major sources of dust emissions were equipped with dust-collecting facilities. The quantity of dust was expected to decrease by 97% (Kunda Nordic Cement Corporation 1997). The aim of this research was to study how the environmental load will change as a result of this renovation. Scots pine bark and needles were used as bioindicators.

Material and methods

The study area is situated on the southern side of the Gulf of Finland, in north-eastern Estonia (Fig. 1). The relief is flat and the soils in the forest sample plots are mainly Gleyic Podzols on sands. The area belongs to the mixed-forest subregion of the Atlantic-continental region. According to the Kunda meteorological station, the mean annual temperature is about 5 °C and the annual amount of precipitation is 550 mm. The dominant winds blow from the south-west and south (Mandre 1995a). Total production of cement and clinker were at the level of 1 million tonnes in the 1990s and dust emissions were 80 000 tonnes at the beginning of the renovation programme (1992).

The studies were performed on 5 sample plots at different distances from the cement plant: Kunda 0.15 km (59°30´N, 26°30´E), Kunda 1.0 km, Lontova 2.5 km, Toolse 5.5 km and Eru 38 km. They were situated in Scots pine, *Pinus sylvestris* (L.), forests, and mainly on sandy heaths. Similar types of forest were selected for the study, but this was not possible in every case due to different geological conditions and to the effect on vegetation and soil by massive long-lasting pollution. The nearest sample plot is presently a park in Kunda town.

Samples from pine bark and needles were collected during April–May in 1996–1999. The age of pine trees was 50–80 years and three trees

were used for sampling at each site. Bark samples were taken at the height of 1.5 m on the side of the trunk exposed to the cement plant, 4 samples from each tree. Needles were collected from different sides of the mid-crown, 4–5 small shoots from each tree (from a height of approx. 10 m). In this article, one-year-old needles are those which were grown during the preceding summer, and 2-yr needles were grown two years before sampling. The samples were air-dried and stored in the laboratory in dustproof paper bags.

Two methods were used to study the impact of cement plant dust. Firstly, total element concentrations of unwashed pine needles were measured. The needles of composite samples were dried at 110 °C for 12 hours and extracted with HNO_3. We analysed the concentrations of Al, Ca, Fe, K, Mg, and Mn, which are important components of the dust from cement plants. The elements were determined with an atomic absorption spectrophotometer (Varian SpectrAA-400; for Al analysis a graphite furnace Varian GTA-96 was used). The results are given as mean values of two replicates. International Standard Reference Material was used for external quality control, the results are shown in Table 1.

Secondly, element concentrations on the surface of pine bark and needles were studied with the SEM/EDX method. Samples of bark and needles were mounted on sample stubs and coated with gold with a JEOL Fine Coat Ion Sputter JFC-1100 coating unit. The preparations were examined with a JEOL JSM 840 scanning electron microscope (SEM) and JEOL Semaphore image recording system operating at 6 kV. Analyses were performed directly in the SEM with the PGT energy dispersive X-ray microanalyser (EDX) and IMIX system. The voltage was 15 kV and the time of X-ray collection was 60 seconds. The analyses were performed applying the method developed by Haapala (1998).

1000-fold magnification was used during X-ray collection. For each sample plot, the concentrations of Al, Ca, Fe, K, Mg, S, and Si on bark and needle were measured with SEM/EDX as a mean of 40 randomly selected points on the bark surfaces. Differences among the samples were calculated with one-way ANOVA, a part of Statistix 7 software.

Results

A thick dust layer covers all the surfaces including the buildings in the surroundings of the cement plant. The same is true for pine bark and needles, which are covered with a crust to a distance of more than 2 km from the plant. Needle growth was seriously impaired, the length of needles being in many cases only 0.5–1 cm, and the needles of the same shoot and the same year more than 5 cm, respectively. In the vicinity of the plant the needles died when they were only one year old.

Element concentrations of pine needles

In the nearest vicinity of the cement plant (0.15 km), calcium concentrations were very high (max. 34 000 µg g⁻¹) exceeding the background values almost tenfold (Haapala *et al.* 1996b) and decreasing sharply with increasing distance from the plant (Fig. 2). Most of this calcium was deposited on the surface of the needles, which was covered by a thick crust. The Ca concentration of the 2-year needles was always clearly higher than that of one-year-old needles, indicating accumulating deposition (Table 2). The oldest needles studied in this work were grown in 1995, that is at the time when the renovation programme was already in

Table 1. Obtained (AAS, averages, $n = 8$) and certified metal concentrations of a standard reference material (NIST SRM 1573a, Tomato leaves); µg g⁻¹ dw.

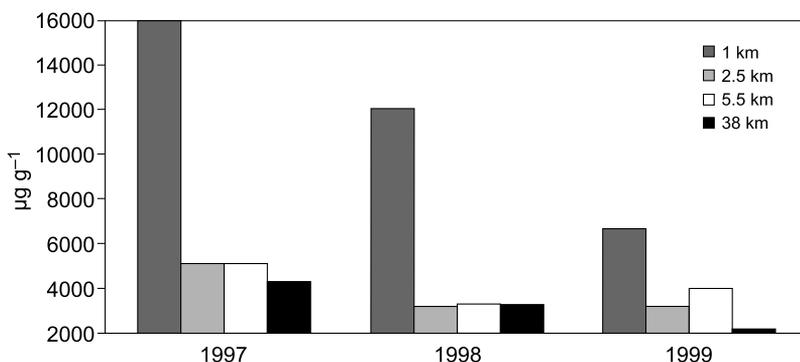|	Mn	Zn	Fe	Ca	K	Mg	Al
Obtained ± SD	229 ± 2.0	30.3 ± 0.4	380 ± 10	53000 ± 1000	2770 ± 60	1075 ± 15	240 ± 20
Certified ± uncertanties	246 ± 8	30.9 ± 0.7	368 ± 7	50500 ± 90	2700 ± 0.05	1200*	598 ± 12

* Determined, not certified

Fig. 2. Ca concentrations of unwashed one-year-old Scots pine needles collected in 1997–1999 at different distances from the cement plant.
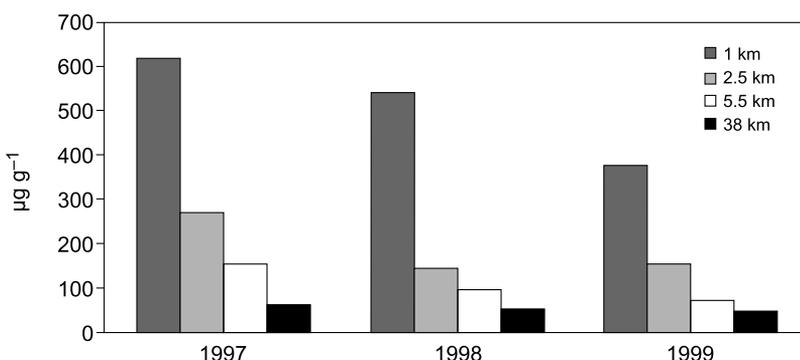

Fig. 3. Fe concentrations of unwashed one-year-old Scots pine needles collected in 1997–1999 at different distances from the cement plant.

progress. Since then, the Ca concentration has clearly decreased. However, Ca concentrations of the pine needles were still relatively high in needles grown during 1998 and sampled in 1999, two years after the renovation programme was completed.

Iron results were similar to those of Ca, with decreasing concentrations as the distance increased, with reducing concentrations during the study period (Fig. 3) and with higher concentrations in 2-yr as compared with 1-yr needles (Table 2). Considerable amounts of Fe were still

Table 2. Metal concentrations in current (1-yr) and previous year's (2-yr) needles collected in 1997. Risk levels (*p*) for pairwise differences between 1-yr and 2-yr concentrations (*significant differences) for all years and all distances (paired *t*-test).

	Age	0.15 km	1 km	2.5 km	5.5 km	38 km	*p*
Ca	2-yr	34000	25000	18000	6400	6200	0.003*
	1-yr	28000	16000	5100	5100	4200	
Fe	2-yr	940	800	670	260	79	0.013*
	1-yr	1000	620	270	150	61	
Mg	2-yr	1700	2000	1300	910	1200	0.313
	1-yr	1900	1800	1200	990	1200	
Mn	2-yr	31	52	75	48	350	0.104
	1-yr	35	56	47	50	220	
K	2-yr	4600	5400	6200	3600	2800	0.002*
	1-yr	5200	8800	8200	5500	3400	
Al	2-yr	820	520	590	330	400	0.318
	1-yr	1000	760	500	210	430	

Fig. 4. Al concentrations of unwashed 1-year-old Scots pine needles collected in 1997–1999 at different distances from the cement plant.
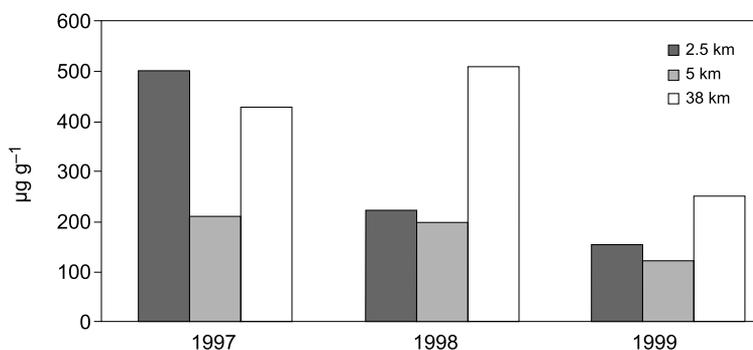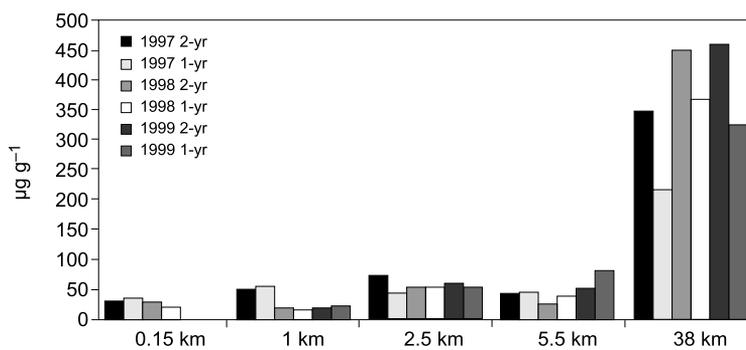

Fig. 5. Mn concentrations of unwashed 1-year and 2-year-old Scots pine needles collected in 1997–1999 at different distances from the cement plant.

deposited on the needle surfaces after the renovation programme was completed, the impact being observed up to a distance of 5.5 km.

Potassium and magnesium showed a different distribution pattern as compared with Ca, giving evidence of a more complicated impact of environmental factors. For potassium, the differences among the sample plots were not very clear, 2-yr needles had lower concentrations than 1-yr needles (Table 2) and the concentrations increased during the renovation. Pine needle aluminum concentrations were high (over 1000 µg g^{-1}) near the plant, indicating cement dust deposition. However, the Al concentrations of the remotest plot (38 km) were higher than those nearer the plant (2.5 and 5.5 km), especially at the end of the research period (Fig. 4), most distinctly at the remotest sample plots.

The distribution pattern for manganese is totally different from other elements (Fig. 5). The highest concentrations, between 200 and 500 µg g^{-1}, were measured at the remotest plot. Near the cement plant, pine needle Mn concen-

trations were very low (only 20 µg g^{-1}). It should be noted that practically no changes in needle Mn concentrations were found during the renovation process.

Element concentrations on the surface of pine bark and needles

A thick dust layer covers the pine bark and needles up to a distance of several kilometres from the Kunda cement plant. It is possible to get information about the amount of dust by measuring the element concentration on bark surfaces with SEM/EDX (Haapala 1998). However, in this case the deposited layer was in some cases so thick that not all of it was registered, since the electron beam of SEM did not penetrate the whole layer.

The calcium concentration on the surface of pine bark and needles was extremely high near the cement plant, being almost twentyfold to that measured about 38 km west from Kunda. Similar
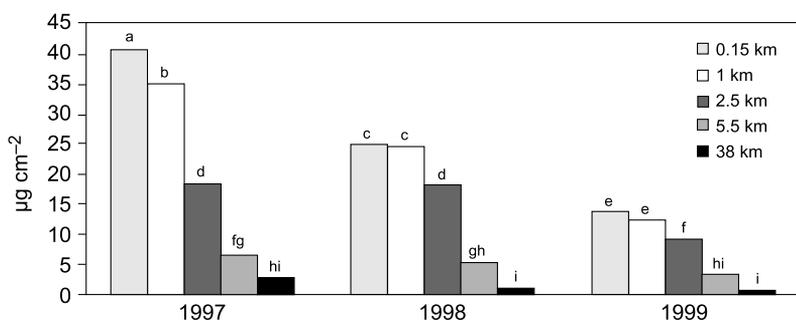
Fig. 6. Ca concentrations on the surface of Scots pine bark collected in 1997–1999 at different distances from the cement plant. Columns that do not share a letter in common were significantly different (at $\alpha = 0.01$, one-way ANOVA).

pollutant gradients were observed for all the included elements, even though the differences were smaller. In the case of silicon, the difference was about fivefold. Figure 6 shows that Ca concentration really decreases as a function of increasing distance and time, the differences being statistically significant. Even in Eru, 38 km west from the cement plant, the Ca concentration on pine bark decreased, showing that a small deposition was transported even this far.

However, the calcium level on pine bark in 1999 — two years after the completion of the renovation — is still relatively high near the pollutant source. The same was true of the needles: even visual examination indicated that the youngest needles grown in the summer of 1998 and sampled in May 1999 were covered by dust. SEM showed that a dust crust was accumulating on the surface of the needles. The SEM/EDX measurements supported these observations. The element concentration on the surface of the pine needles, which was much lower than on the bark, really did decrease. However, the main components of the emissions, Ca and Si, were still at a rather high level two years after the renovation was completed. Ca and Si concentrations were three to sixfold higher near the cement plant (1 km) than in the remotest plot (38 km). Dust element concentrations in Lontova (2.5 km) and Toolse (5.5 km) were also relatively high on the surface of needles collected in 1999.

Discussion and conclusions

Calcium is most suitable for monitoring the environmental pollution in the surroundings of

Kunda cement plant, because CaO is the most abundant component (over 40%) of cement dust emissions. Ca concentrations of unwashed pine needles were accordingly very high, exceeding somewhat the values measured earlier in the same area by Mandre (1995b). Background values in our present and previous work (Haapala *et al.* 1996b) were one-tenth of those levels. It was calculated that 20 000–30 000 μg g^{-1} of Ca was included in the cement crust deposited on the needle surface. On the other hand, SEM/EDX measurements showed that the Ca-content on pine needle surface may increase to 15 mg dm^{-2}, which makes approximately 15 000 μg Ca g^{-1} dry needle mass. As was stated earlier, all Ca was not registered by SEM/EDX in cases when the deposited layer was so thick that the electron beam of SEM did not penetrate the whole layer. Taking into account this limitation, we concluded that both the AAS-measurements and SEM/EDX-studies were fairly consistent with each other.

The dust deposited on natural surfaces and Scots pine bark and needles clearly decreased as a result of the renovation programme, which was aimed to reduce the dust emissions of the cement plant by 97%. The decrease was observed even on the control sample plot 38 km west from the plant, indicating that the emissions were transported even this far. At the beginning of this work the area showed elevated Ca levels (6000 μg g^{-1}), which decreased (to 3000 μg g^{-1}) as a result of reducing the emissions.

We assumed that one-year-old pine needles give the best information about the environmental change caused by the renovation programme, since only the emissions of the last year are deposited on their surface. Bark studies do not

give evidence of the real emission level, since dust pollution accumulated over many years does not easily disappear from the bark surface. By collecting 1-yr and 2-yr needles during the renovation programme and two years after its completion, we could show a decrease of dust pollution, if the dust level was very low on the surface of the one-year-old needles collected in 1999.

However, an unexpectedly high dust concentration was measured in all experiments including the surfaces of one-year-old needles even two years after the renovation was completed. At the distance of 1 km from the cement plant, the Ca-content of unwashed needles was 6600 µg g^{-1}, which was twofold compared with the background. The difference was still greater when needle surface element content was compared. These results can at least partly be explained by the following:

— The estimated amount of dust emissions after the renovation is still over 1000 tonnes y^{-1}, thus producing new particles in the air. The implementation of the renovation plan was not complete, causing higher dust pollution than those aimed for. According to Kaasik and Sõukand (2000), the snow melt water pH in Kunda area during January–March 1999 was still between pH 8–10 which, however, was remarkably lower than during 1984/1985 when pH 10–12 was recorded.
— Thick layers of dust have been deposited on all surfaces in the surroundings of the cement plant. Part of this dust is re-emitted by winds and deposited again, e.g. on recently grown needles. The amount of this recirculating dust remains at a high level for a long time.

The results concerning aluminum are affected mainly by two processes, upon which the alkaline pollution has an opposite effect. The emissions themselves contain quite a lot of Al. A pollutant gradient following increased distance from the plant should thus be expected. On the other hand, aluminum in the soil changes to an insoluble form as a result of soil alkalization due to these pollutants (Haapala *et al.* 1996b). For this reason, needle Al concentration should be highest in the remotest plots where the soil is most acidic (the pH from a KCl solution was 3.0–3.6, compared to 7.4–7.8 near the cement plant; Annuka and Mandre 1995). It is thus concluded that, during 1998 and 1999, the low Al concentration at distances of 2.5 and 5.5 km from the plant are caused by the simultaneous impact of these two factors. The deposited Al on the surface of the needles has been decreased to a low level after the restoration and at the same time the soil is still alkaline, thus decreasing Al solubility and uptake from the soil.

Variations in potassium and magnesium concentrations were not easy to explain. Deposited potassium and magnesium, which were important ingredients in cement plant emissions, increased the total concentration of these elements in the soil and on needle surfaces. However, element solubility and uptake by plants may be strongly reduced in dusted soils (Haapala *et al.* 1996, Annuka and Mandre 1995, Mandre 1995b), being affected by soil alkalinity and a very high Ca concentration. Two opposite processes occurred during the renovation: a decreasing deposition on needle surfaces and, to a greater degree, an increasing uptake from the soil. At different distances from the cement plant, the processes had different rates and therefore the total figure was quite complicated.

The manganese concentrations of pine needles were in the background sample plot similar to those of Finnish Scots pine needles (Raitio 1990), as well as to those of Norway spruce (*Picea abies* (L.) Karst.) needles in the Estonian background area of Mandre (1995b). However, pine needles suffered from a serious Mn deficiency on polluted plots, as did the spruce needles studied by Mandre (1995b). In an alkaline soil Mn^{2+} is oxidized to Mn^{3+} and Mn^{4+}, which are difficult for plants to assimilate (Mandre *et al.* 1999). The Mn deficiency causes damage to growing conditions having serious effects on conifer physiology including chlorosis (Mandre 1995c, Mandre and Tuulmets 1995, Mandre *et al.* 1999). In our study area, chlorosis was found especially in Norway spruce, mainly in young trees and current-year needles. Similarly, Cenni *et al.* (1998) observed that a severe decline of the Austrian black pine (*Pinus nigra* Arn.) consisting of widespread crown yellowing was caused by manganese deficiency. This was due to an antagonism between calcium and Mn on a

limestone substrate, the needle yellowing being called "limestone chlorosis". Also Hiltbrunner and Flückiger (1996) found needle chlorosis of silver fir (*Abies alba* Mill.) under soil Mn-deficiency.

The results give indirect evidence that the recovery of soil is slow. Soil is alkalised over wide areas and this alkaline impact is very strong. It is characterised by very low levels of available manganese in the soil and consequently in the pine needles. Continuing dust pollution — even though at a much lower level — can maintain the prevailing situation. However, the polluted area probably becomes gradually smaller.

Unwashed needles contained a lot of elements on their surface, a major part of the entire needle concentration being on the surface in the case of Ca, Si, Fe. Therefore, the measurement of total needle element concentration was a suitable method to study the decrease of dust pollution in the environment. However, the uptake from the soil for some other elements (Al, K, Mg and especially Mn) is very important for total needle concentration, making conclusions based on total needle analysis more difficult. In all cases, measurement of element concentration on the surface of pine bark and needles gives more direct information about the quantities of dust deposition. The current-year needles are most suitable for such monitoring.

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