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Quantitative variation in the elemental composition of Scots pine needles along a pollutant gradient

Kari Heliövaara & Rauno Väisänen

TIIVISTELMÄ: MÄNNYNNEULASTEN ALKUAINEKOOSTUMUS HARJAVALLAN TEOLLISUUSALUEELLA

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Quantitative variation in the elemental composition of living Scots pine needles was studied along an atmospheric pollutant gradient in the surroundings of the industrial town of Harjavalta, southwestern Finland. Two 9-km-long transects, each with nine sample plots, running to the S and SW from a distinctive factory complex were delimited in a homogeneous pine forest. Needle samples were taken from 10 trees at each site, and from two separate sites in Tuusula near Helsinki. There was considerable spatial variation in the elemental composition of the needles. Heavy metals (Cu, Fe, Zn) showed a clear pattern of exponentially decreasing concentration with increasing distance from the emission source. Sodium and potassium concentrations, as well as the ash weight and air-dry weight, also decreased. Magnesium, manganese and calcium concentrations increased with increasing distance.

Männynneulasten alkuainepitoisuuksia tutkittiin saastuneisuusgradientissa Harjavallan teollisuuslaitosten ympäristössä etelään ja lounaaseen osoittavalla 9 kilometrin linjalla, joilla kummallakin oli 9 näytealaa. Kultakin näytealalta kerättiin 10 männystä edellisvuotisia neulasia. Riippumatonta aineistoa kerättiin lisäksi kahdelta Tuusulassa sijainneelta näytealalta. Neulasten alkuainekoostumuksessa havaittiin huomattavaa paikallista vaihtelua. Tutkittujen raskasmetallien (Cu, Fe, Zn) pitoisuudet neulasissa vähenivät eksponentiaalisesti etäisyyden kasvaessa. Myös kalium- ja natriumpitoisuudet sekä neulasten tuhka- ja ilmakuivapaino vähenivät. Magnesium-, mangaani- ja kalsiumpitoisuudet lisääntyivät hieman etäisyyden kasvaessa.

Keywords: *Pinus sylvestris*, air pollution, herbivores, heavy metals, nutrients.
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1. Introduction

The concentrations of chemical elements in the green parts of plants vary depending on the environmental conditions, e.g. the edaphic quality and pollution level at the site. For example, the amounts of heavy metals and some other trace elements usually decrease with distance from the emission source (e.g. Little & Martin 1972, Buchauer 1973, Little 1973, Beavington 1975). Mineral nutrient concentrations also vary according to the physiological status of the plant, and may indicate the magnitude of external stress factors such as the effect of toxic chemicals and water deficiency (White 1974, 1984). There are considerable differences between different organological parts of plants, as has been demonstrated in several studies on coniferous trees (Lowry & Avard 1968, Morrison 1973, Lehtonen et al. 1976, Lehtonen 1977, Bringmark 1977). Nutrient levels also vary between seasons, years and even during different weather conditions (Tamm 1954, Aaltonen 1955, Viro 1955, Likens et al. 1967, Ellenberg 1969, Grier & Cole 1972, Christersson 1974, Lehtonen et al. 1976, Lehtonen 1977).

The effects of chemicals on plants vary according to the doses. Although metals such as copper and iron are essential for the normal development of plants in small quantities, they become toxic at high concentrations (Cottenie et al. 1976, Ormrod 1984). In addition to the direct effects, the various concentrations of chemicals have indirect effects on plants (Huttunen 1984). For instance, they may influence the quality and palatability of plant tissues from the point of view of herbivores. These secondary effects may compensate, or even increase, the direct impact of a chemical element on the plant. Elements which are concentrated in certain plant tissues through varying metabolic pathways also vary in respect to their chemical form, thus affecting their role in the physiology of the plant as well as their biological usability at higher trophic levels (Buchauer 1973, Ormrod 1984).

This paper continues our previous studies carried out in the industrial complex near the town Harjavalta in southwestern Finland (Heliövaara 1986, Heliövaara & Väisänen 1986 a, b, Heliövaara et al. 1987, Heliövaara & Väisänen 1988). In the present article the quantitative variation in the concentrations of elemental composition of living Scots pine (*Pinus sylvestris* L.) needles is studied along an atmospheric pollutant gradient in the surroundings of the industrial town of Harjavalta, in southwestern Finland. The concentrations of different elements were related to the distance from a distinctive pollution source. The correlation between various elements was investigated and the magnitude of mineral nutrient variation among sample trees was determined. The needle material examined here was also used in entomological experiments into the secondary effects of atmospheric pollution. The results of the laboratory studies on the effects of varying food quality on several insect species will be published separately.

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2. Material and methods

21. Study site and sampling

There is a distinctive emission source at Harjavalta (61°20' N, 22°10' E) consisting of two factories sited close to each other. One of these factories produces copper and nickel and the other factory fertilizers and sulphuric acid (Laaksovirta & Silvola 1975). The prevailing winds are from the southwest (Hynninen 1986, Hynninen & Lodenius 1986).

Two transects were delimited through the homogeneous Scots pine forest to the S and SW from the factory complex. Both transects were nine kilometres long, each containing nine sample plots set approximately at logarithmic distances. Each sample plot contained 10 Scots pine trees (mean height 12.5 m, SD 5.6). In addition, material was collected from two sample plots situated in Tuusula near Helsinki (60°22' N, 25°01' E).

Needles from 200 pines were sampled in June, 1987. Needle sampling is usually carried out during the dormant stage of conifers, but we wanted to obtain a more realistic picture of the mineral nutrient and metal composition of the needles from a herbivore's point of view. Only the previous year's needles were collected. About 3 g of needles were carefully removed from different branches around the lower part of the crown and placed in paper envelopes and transported to the laboratory.

22. Pretreatment and chemical analysis of the needle samples

The needles were stored at +20 °C until analysed in August. The samples were dried for 48 hours at +50 °C and then milled in a steel mill to pass through a 0.8 mm sieve. In order to reduce contamination the mill was wiped with 94 % ethanol after each sample. The homogenised needle samples were stored in plastic bags before analysis.

After determining the dry weight (105 °C) and ash weight (muffle furnace at 550 °C), the concentrations of total P, K, Mg, Mn, Fe, Al, Cu, Zn, Na and Ca were determined according to methods described by Halonen et al. (1983) on an ARL ICP 3580 Spectrometer. C, H and N were analysed on a LECO CHN analysor. Each sample was analysed in duplicate, and the mean value used.

The mean length and dry weight of 10 needles from five pines at each sampling site (50 needles per site) were also determined. Scanning electron microscope photographs were taken of the needles collected from both ends of the pollution gradient.

23. Statistical analyses

The differences between sample plots as regards the concentrations of analysed elements were first tested by an analysis of variance. The dependence of the concentration of each element on the distance from the emission source was made using linear regression analysis. Power curves were calculated to describe the concentrations of heavy metals as a function of the distance from the emission source.

The two transects were treated separately in the statistical analyses. Parametric correlation analysis was applied in comparing the concentrations of the different elements with each other. The main dimensions of variation in the elemental composition of the needles was investigated using principal components analysis (PCA).

3. Results

The needles were the smaller the nearer to the emission source they originated from. Distance from the pollution source explained 63−64 % of the variation in the needle length, and 66−72 % of that in needle dry weight (Fig. 1).

Scanning electron microscope photographs show differences in the morphology of the upper half of the needles taken from both ends of the pollutant gradient (Fig. 2). The waxy layer of the needles collected from near the emission source was largely destroyed, and the surface of the needle was covered with small dust particles. These were also visible inside the spiracle. At a distance of 8 km, the waxy layer was more or less intact.

The concentrations of different elements in relation to the distance from the pollutant

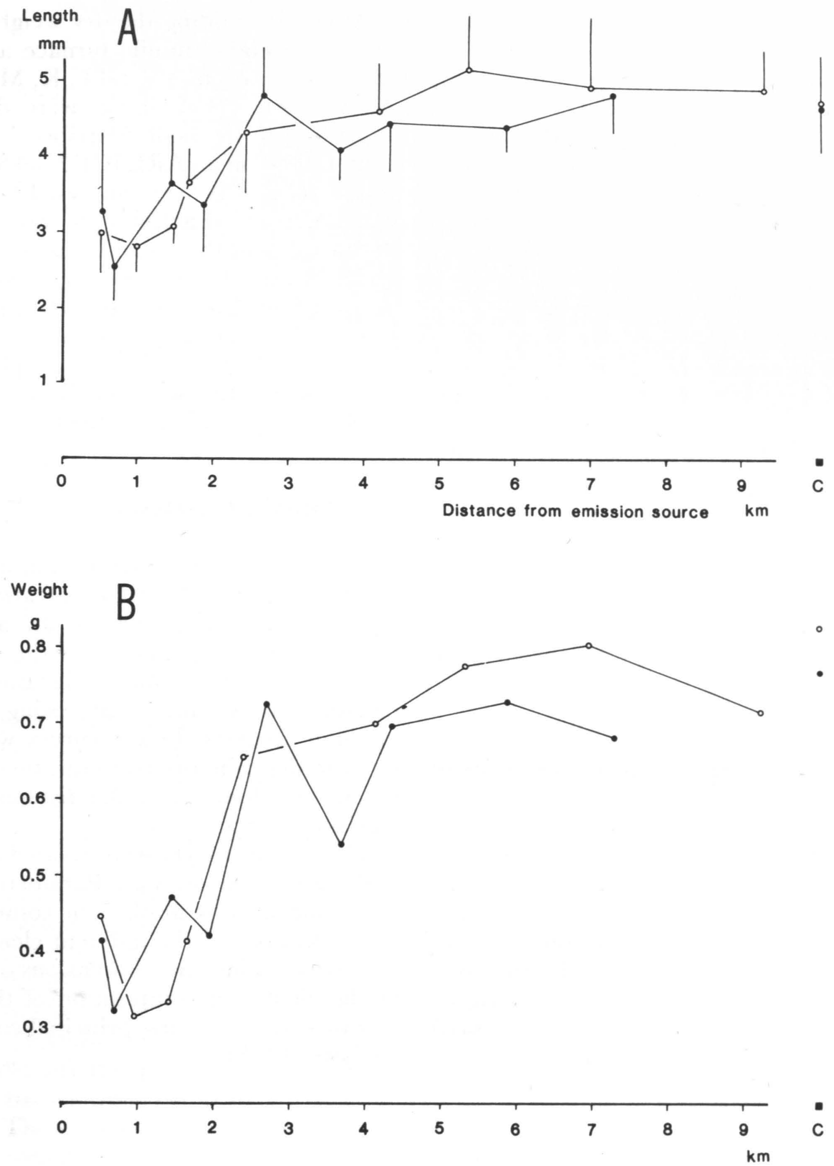


Fig. 1. Mean length ($\bar{x}$, SD, n = 10 needles per tree) (A) and air-dry weight (n = 50) of the previous year's needles (B) from five pines at each sample site. C represents the separate samples from Tuusula. Linear regression equations for length (Tuusula excluded) are y = 3.100 + 0.257x, P = 0.009 (S transect, dots), and y = 3. 084 + 0.260x, P = 0.002 (SW transect, circles), and for weight y = 0.389 + 0.054x, P = 0.007 and y = 0.383 + 0.053x, P = 0.006, respectively.

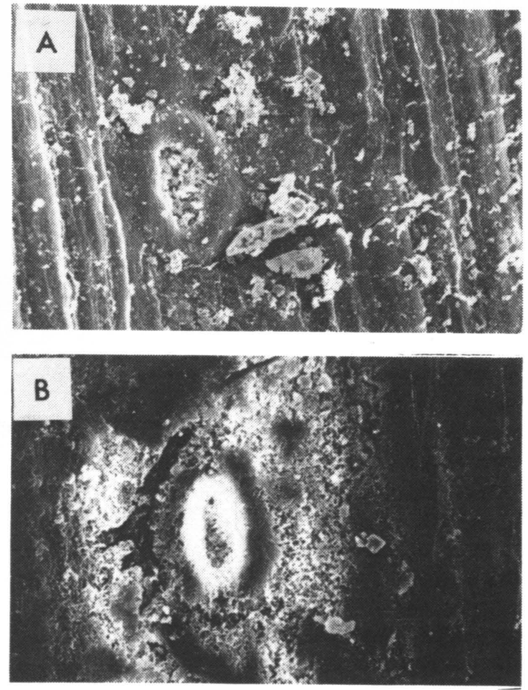


Fig. 2. Scanning electron microscope photographs of pine needles at (A) 400 m and (B) 8000 m distance from the pollutant source.

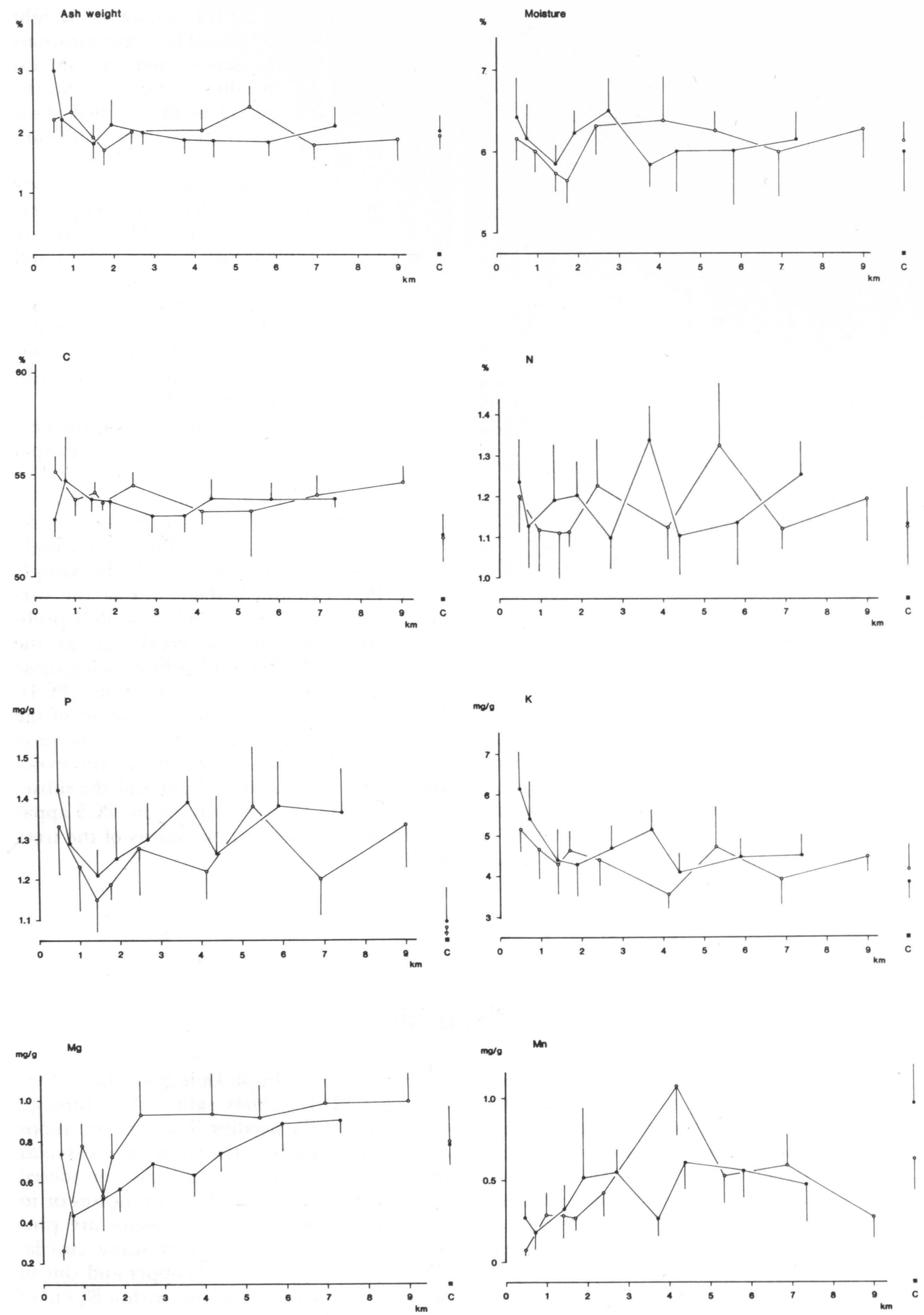
source are described in Fig. 3 and Table 1. The studied heavy metals (Cu, Fe, Zn) follow a clear pattern: exponentially decreasing concentrations with increasing distance from the emission source. The concentrations of sodium and potassium also tend to decrease. A similar pattern was observed for ash weight and air-dry weight, too. The concentrations of magnesium, manganese and calcium increased with increasing distance from the factories. The other elements studied show varying and/or statistically non-significant patterns.

All highly significant correlations ($p<0.001$) among the elemental concentrations are presented in Table 2. The results are partially self-explanatory for dry weight and ash weight, for example. The high positive correlations between the heavy metals are notable (Cu and Fe: r=0.937; Cu and Zn: r=0.783; Fe and Zn: r=0.787). The concentration of nitrogen is positively correlated with that of phosphorus (r=0.590).

The main variation trends among the needle samples were investigated using principal components analysis (PCA) (Table 3). The highest loadings for the principal component 1 (PC1), were given by the heavy metals. This is interpreted as a dimension closely related to the pollution level at the sample plots. PC1 explained about 1/4 of the variance in the data space. PC2 and PC4 probably reflect the edaphic conditions at the sample plots, the highest loadings being those for calcium (PC2) and phosphorus (PC4). PC3 was related to the water content of the needles, and is thus affected e.g. by the physiological state of the tree (stress) and exposure of the needles to sunlight and the subsequent level of evapotranspiration. PC5 apparently reflects the nutrient status of the trees and the needles.

4. Discussion

The results show that there is considerable spatial variation in the mineral nutrient and heavy metal composition of the pine needle material within a small area, and that a relatively large proportion of this variation could be explained by local atmospheric deposition. This was especially clear in the case of heavy metals. Part of the needle heavy metal content is accounted for by dry deposition, observed as dust on the needle surface. Most heavy metal uptake is from the soil via the roots (Tyler 1984) rather than through the surface of the needles. The concentrations of metal pollutants in the biomass of vascular plants, such as trees, are usually considered to be much lower than those in mosses or in litter (Roth 1985), as their tissues are protected by the epidermis and waxy cuticle. The 3-month deposition of copper and zinc in *Sphagnum* moss bags laid out within 800 m of



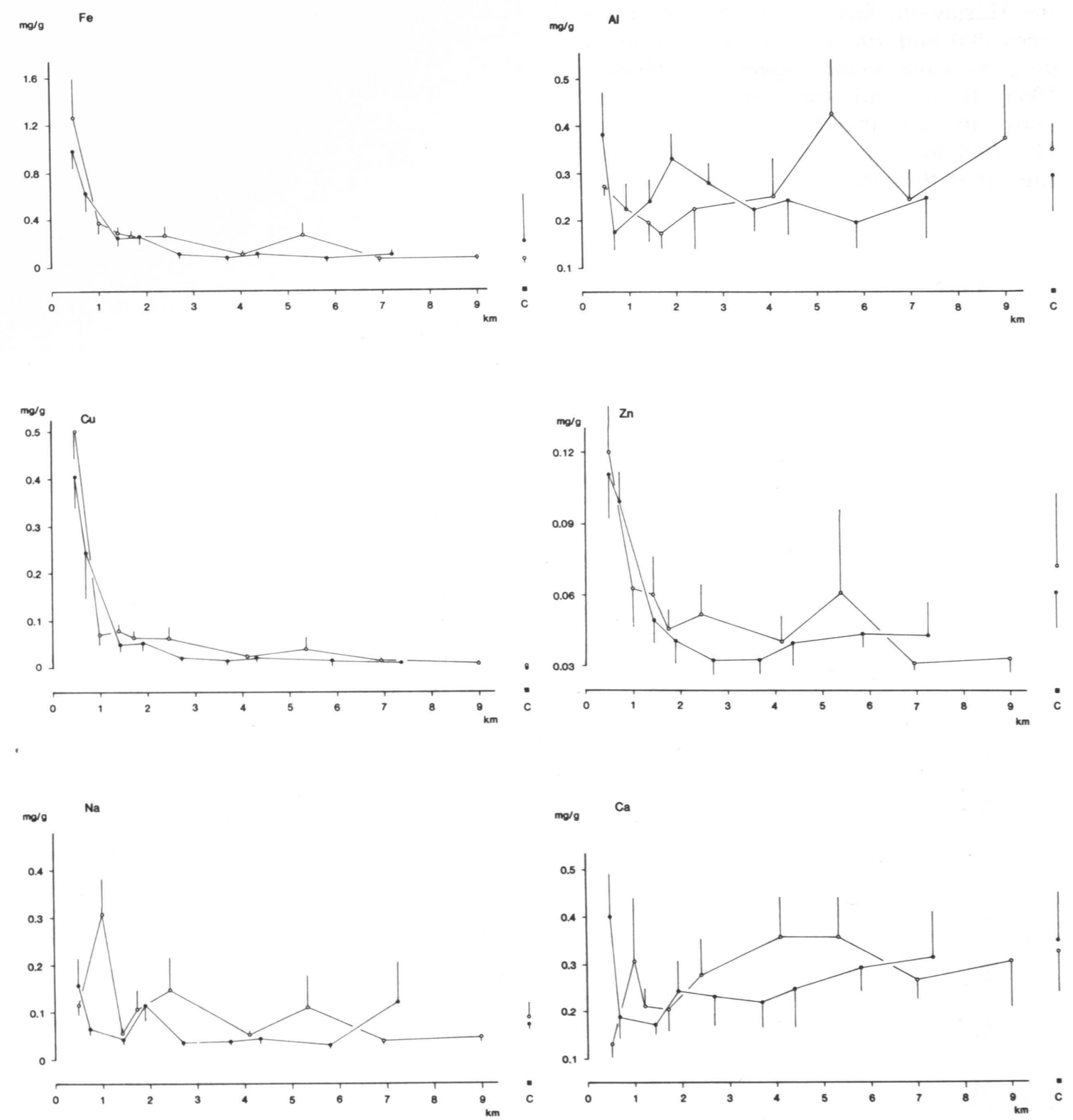


Fig. 3. Means of different elemental concentrations in relation to the distance from the pollutant source at Harjavalta. Vertical bars indicate standard deviations. Respective results for Tuusula are given on the right (C). For regression equations, see Table 1.

the Harjavalta factory complex varied between 200 and 700 and between 25 and 100 µg/g, for each metal respectively (Hynninen 1986). In our study the respective concentrations in 2-year-old pine needles were 317–898 for copper and 98–164 µg/g for zinc. It is thus clear that the pines near the factories are relatively heavily contaminated.

The causal factors lying behind the dependence of Mg, Mn, Na, K, Ca concentrations on the distance from the pollutant source remain to be investigated in more detail. However, it seems probable that the indirect effects of air pollution can bring about both major changes in the metabolism of the plant and in the plant-herbivore relationships (see Huttunen 1984). The edaphic conditions of the site may in certain cases also compensate for the effect of pollution on pines.

The pines growing close to the the pollutant sources were no doubt badly stressed by air pollution. However, we were not able to demonstrate any significant differences between the nitrogen contents of pines growing on heavily, moderately and slightly polluted sites (cf. White 1984). This result does not mean that there are no differences in the actual amount of nitrogen available for herbivores since differences in the amounts of defensive organic compounds in pine needles were not examined.

The observed gradients in the morphology and elemental composition of the pine needles increase variation in the food quality and palatability for pine herbivores and consequently may affect the susceptibility of pines to insect defoliators. The impact of air pollutants on forests has been shown to be related to the outbreak pattern of several insect species (e.g. Lechowicz 1987). Plants weakened by air pollutants are especially predisposed to attacks by aphids (Braun & Flückiger 1984 a, b, Dohmen 1985) and, in the case of forest trees, also moths (Templin 1962, Sierpinski 1966), sawflies (Wentzel & Ohnesorge 1961, Wentzel 1965) and bark beetles (Stark et al. 1968). The pollutant concentration affects the population dynamics of different insect species in markedly different ways, and all species do not, of course, gain any benefit from a heavy pollution load.

Alstad et al. (1982) have concluded that exposure of a plant to air pollution can alter the quantitative and qualitative composition of secondary products. Plant palatability, nutritional quality, fungal and bacterial epifloras or the attractiveness of the plant as an oviposition site may also be altered as a result of pollution stress. Sulphur dioxide and ozone cause major injury to plants. However, near to distinctive sources of pollution, such as smelters, much damage is caused by fluorides, dust, and – as in the present case – heavy metals, which are generally considered to be of minor importance (Kozlowski 1980).

Table 1. Statistical trends in the elemental concentrations in relation to the atmospheric pollution level. Differences between sample plots were studied using ANOVA, df1=9, df2=90. The dependence of the concentration on the distance from the pollutant source was studied using linear regression models, df=88. The elements have been arranged according to their correlation relationships (see Table 2).

Element	Transect S/SW	ANOVA F	p<	Regression model Equation	r	p<
Ash weight	S	18.04	0.001	y=2.276–0.001x	–0.323	0.01
	SW	7.35	0.001	y=2.094–0.000x	–0.145	NS
Dry weight	S	1.87	NS	y=93.815+0.000x	0.108	NS
	SW	3.86	0.001	y=94.050–0.000x	–0.261	0.05
Moisture %	S	1.87	NS	y=6.189–0.000x	–0.108	NS
	SW	3.73	0.001	y=5.961+0.000x	0.244	0.05
C	S	5.38	0.001	y=56.539+0.000x	0.051	NS
	SW	11.58	0.001	y=57.152–0.000x	–0.045	NS
H	S	10.93	0.001	y=6.559+0.000x	0.114	NS
	SW	8.99	0.001	y=6.718–0.000x	–0.367	0.001
N	S	6.22	0.001	y=1.176+0.000x	0.068	NS
	SW	3.94	0.001	y=1.170+0.000x	0.079	NS
Air dry w.	S	18.74	0.001	y=14.116–0.370	–0.474	0.001
	SW	1.02	NS	y=13.482–0.000x	–0.014	NS
P	S	6.85	0.001	y=1.291+0.000x	0.134	NS
	SW	6.97	0.001	y=1.222+0.011x	0.236	NS
K	S	10.68	0.001	y=5.296–0.150x	–0.368	0.001
	SW	5.03	0.001	y=4.655–0.064x	–0.234	NS
Mg	S	16.06	0.001	y=0.507+0.054x	0.634	0.001
	SW	21.75	0.001	y=0.573+0.055x	0.578	0.001
Mn	S	18.55	0.001	y=0.297+0.036x	0.388	0.001
	SW	42.50	0.001	y=0.309+0.031x	0.300	0.01
Fe	S	38.10	0.001	$y=0.436x^{-1.17}$	0.708	0.001
	SW	93.94	0.001	$y=0.499x^{-1.29}$	0.742	0.001
Al	S	9.06	0.001	y=0.291–0.010x	–0.259	0.05
	SW	10.41	0.001	y=0.203+0.016x	0.435	0.001
Cu	S	154.45	0.001	$y=0.131x^{-1.73}$	0.914	0.001
	SW	63.28	0.001	$y=0.116x^{-2.09}$	0.945	0.001
Zn	S	54.94	0.001	$y=0.076x^{-0.54}$	–0.045	0.05
				(y=0.079–0.076x	–0.569	0.001)
	SW	17.82	0.001	$y=0.077x^{-0.44}$	–0.240	0.001
Na	S	17.92	0.001	$y=0.096x^{-0.35}$	–0.316	0.001
	SW	31.50	0.001	$y=0.146x^{-0.35}$	–0.360	0.001
Ca	S	9.64	0.001	y=0.239+0.005x	0.125	NS
	SW	8.33	0.001	y=0.221+0.013x	0.356	0.001

Table 2. Simple correlations among the elemental concentrations in all the sample trees. All statistically highly significant correlation coefficients (p<0.001), either positive (+) or negative (–) are given. Cases in which r>0.500 are indicated by double markings (++ or - -). Abbreviations: Dw = dry weight, Aw = ash weight, Adw = air dry weight, M% = moisture %.

	Dw	Aw	M%	C	H	N	Adw	P	K	Mg	Mn	Fe	Al	Cu	Zn	Na	Ca
Aw																	
M%	- -																
C	–		+														
H		–		++													
N			+														
Adw																	
P		+				++											
K		+			–	+		++									
Mg									–								
Mn					–			–	- -	+							
Fe		+								+	–	–					
Al		+			–	+											
Cu		+						+	+	–	–	++					
Zn		++					+		+	–	–	++		++			
Na		+										+		+	+		
Ca		++		–	–				–	++	+		+				+

Table 3. Results from the principal components analysis (PCA) on the main trends in elemental variation of the pine needles. The loading matrix has been rearranged so that the columns appear in decreasing order of variance explained by the principal components. The rows have been rearranged so that loadings greater than 0.500 appear first for each successive principal component. Loadings less than 0.250 have been omitted and loadings less than 0.450 have been indicated by + or — only.

	PC1	PC2	PC3	PC4	PC5
Eigenvalue	4.191	3.237	2.151	1.858	1.177
% variance explained	24.7	19.0	12.7	10.9	6.9
Cumulative % variance explained	24.7	43.7	56.4	67.3	74.2
Sorted factor loadings					
Cu	0.902				
Fe	0.886				
Zn	0.801			–	
K	0.690			0.453	
Ash weight	0.675	0.569			
Mn	–0.549	0.457		–	
Ca		0.778		–	
H		–0.729			0.482
Mg	–	0.604			+
Al		0.602			
C		–0.535	0.531		0.454
Dry weight		–	–0.930		
Moisture %		+	0.930		
P	+	+		0.686	+
N	+	+		0.502	0.485
Na	+			–	+
Air dry weight		–		–0.485	

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