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Observations on trees of Scots pine (Pinus silvestris L.) and lichens around a HF and SO_2 emission source

Iakttagelser på tall (Pinus silvestris L.) och lavar runt en emissionskälla för HF och SO_2

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Abstract

ODC 425.1 : 174.7 : 172.9 Pinus silvestris

The influence of air pollutants from an iron sintering plant in the northern part of Sweden on Scots pine trees and lichens has been studied. The emission has been around 175 ton HF and 2000 ton SO_2 yearly. The total fluorine and sulphur contents of Scots pine needles from different sample plots around the factory have been analysed. Three years old needles, which had the highest concentrations of fluorine, were used in the analyses. An increased fluorine content was evident within an area of 5 km radius from the emission source. The corresponding sulphur analyses did not show an equal clear relationship to the distance from the factory. The accelerated needle drop of Scots pine trees has been quantitatively recorded. A shorter lifespan of needles coincided with the proximity to the emission source. Together with the observations of the occurrence of the lichens Hypogymnia physodes (L.) W. Wats., Alectoria spp. and Nephroma arcticum (L.) Torss. their photosynthetic and nitrogen fixation activity was measured under laboratory conditions. The photosynthesis, measured by 14C-technique, was inhibited in samples of Hypogymnia physodes taken within the influence area mentioned above. Also the nitrogen fixation activity in excised cephalodia of Nephroma arcticum sampled near the emission source was significantly inhibited. The results presented are discussed in relation to the techniques used and the ecological consequences of the emission and imission in the area.

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1 Introduction

The effects of polluted air from industrial emission sources on surrounding vegetation has been well documented, especially regarding the effects on epiphytic lichens and coniferous trees. This research has mainly been concentrated on the destructive effects of sulphur dioxide emissions. Much information, however, is also available concerning fluorine compounds identified as having deleterious effects. The effects of sulphur dioxide and fluorine compounds on lichens has recently been reviewed by Ferry et al. (1973) and Gilbert (1971, 1973 a, b). The corresponding effects on coniferous trees have also been thoroughly reviewed (Scurfield 1960 a,b, Garber 1967, Tamm and Aronson 1972, Mudd and Kozlowski 1975).

The aim of the present study has been to evaluate the extension of eventual influences on the surrounding vegetation of the emission from the iron-sintering plant in Vitåfors, Norrbotten, Sweden-in close proximity to the communities Koskullskulle, Malmberget and Gällivare. The yearly emission of sulphur dioxide and hydrogen fluoride has been around 2000 ton and 175 ton, respectively for the last years. The content of sulphur and fluorine in the needles of Scots pine from the sample plots has been analysed and correlated to the emission source. The needle drop of coniferous trees, well-known from air polluted areas, has been quantitatively recorded and correlated with the occurrence and physiological reactions in some lichen species from sample plots at different distances from the emission source. Earlier reports on the impact of iron-sintering plants on their biological environment seem to be limited to studies on predominately lower levels of the vegetational strata (Gordon and Gorham 1963, Rao and LeBlanc 1967).

2 Material and methods

2.1 The sampling area

The LKAB sintering plant in Vitåfors is located in Norrbotten, the most northern county of Sweden at a latitude $67^{\circ}N$ and at a longitude $20^{\circ}E$ on an altitude of about 450 m. The near surroundings of the plant has a marked topography with altitude variations between 270 m (at Lina river) and 600 m (on the top of the hill Kungsryggen). For a survey of the area, cf. Fig. 1. The tree-limit is just above 600 m.

2.2 The emission source

The emission source is situated on the northern slope of the hill Kungsryggen in the Lina river valley, the main direction of which is northwest to southeast. The emission of sulphur dioxide and fluoride compounds, respectively, for the last years at this industry is given in Fig. 2. Actually there are several chimneys emitting both SO_2 and HF, the biggest emission products. The two sintering furnaces have chimney

heights of 45 and 70 m, respectively. Particulate fluorides and nitrogen oxides (NO_x) are also emitted from the industry. The exact amount of NO_x is not known but is probably less than the quantity of HF. According to estimations made by SMHI, Norrköping, calculating with a total emission of fluoride compounds of 1100 kg day-1, the monthly mean value for February in Koskullskulle, 1.7 km SE the industry (near SO1, cf. Fig. 1) would be 1.09 μ g F.m⁻³ and for an unfavorable summer month 0.77 μ g F.m⁻³ (data after Bringfelt and Persson 1974). However, also the broken ground in this area may have considerable influence on the actual distribution of smoke-gases from the factory. This may be considered together with the prevailing wind directions presented in Tab. 1. It should also be noted that these wind directions are based on pooled data from the climate stations Malmberget airport and Gällivare 10 km SSE and S, respectively the emission source.

Table 1. Monthly wind distribution in the Gällivare—Malmberget area in the period 1965—1974 calculated after data from SMHI, Norrköping.

Month	Per cer	nt distribu	tion						
	N	NE	Е	SE	S	SW	W	NW	CALM
1	5.1	7.4	9.2	8.2	4.5	7.8	11.9	7.9	37.9
2	1.4	9.4	8.2	6.5	2.9	11.7	8.9	10.2	40.9
3	6.5	8.1	8.4	9.6	5.0	12.8	15.1	10.0	24.5
4	11.1	10.0	12.6	12.2	5.4	10.7	14.0	8.4	15.6
5	14.1	13.2	11.3	8.8	7.2	9.2	11.8	12.8	11.5
6	6.4	6.0	8.2	11.0	8.8	16.4	21.7	12.8	8.7
7	5.4	5.6	9.7	15.5	10.6	12.8	15.9	9.9	14.6
8	8.9	5.9	6.3	14.8	8.5	10.7	15.4	12.1	17.3
9	6.5	5.2	11.0	13.5	5.9	13.8	14.3	12.0	17.9
10	5.7	7.0	7.4	11.6	3.6	11.3	18.7	13.5	21.1
11	6.8	8.9	7.6	9.9	4.6	9.2	13.8	12.4	26.9
12	4.5	9.0	11.5	10.1	5.6	9.9	14.9	9.5	25.0



Figure 1. The distribution of the sample plots around the LKAB sintering plant in Vitåfors, Malmberget, Norrbotten, Sweden $(67^{\circ}0'-67^{\circ}20'N \text{ lat.}, 20^{\circ}25'-20^{\circ}40'E \text{ long.})$.

0-point at the emission source of the industry. Marks on the "Topografisk Karta över Sverige, Rikets Allmänna Kartverk, fältkarta 28 K, Gällivare SV, 1: 50.000".



Figure 2. The cumulative emission of SO₂ and HF from the LKAB sintering plant in Vitåfors, Norrbotten during the period 1962—1973.

After data from LKAB. The two curves for the HF-emission represent the results from the highest and lowest estimated levels of emission.

2.3 The sample plots

Efforts were made to locate the sampling plots in as comparable areas as possible and symmetrically around the industry with the smoke stacks as central-point. For practical reasons it was not possible to carry out this intention strictly but plots in most of the main directions were located up to a distance of about 15 km from the emission source. A survey of the sample plot locations and their vegetation is given in Fig. 1 and Tab. 2, respectively. The plot designations and topographical profiles between the source and the sampling plots, respectively, are presented in Fig. 3 A—7 A.

2.4 The needle recording

During the period 27.8—5.9 1974 material was collected from trees of Scots pine, *Pinus silvestris* L., which is the prevailing coniferous species in the area. In each sample plot four trees were chosen, two with a vigorous growth and two of an

inferior kind. Each couple was as similar as possible with respect to general appearance. From each tree two branches were taken from the lower part of the crown (for an exception, cf. Fig. 7). Branches were avoided, which by flowering had a disturbed needle pattern. Trees near roads or with resinous wood or otherwise directly visibly affected by insects etc. were also avoided.

In the laboratory the numbers of needle pair of each yearly shoot were recorded from the main shoot and one side branch of sufficient age permitting recording the numbers from each annual section up to a total age of ten years. Simultaneously the lengths (in mm) of the annual sections, respectively, were measured. For each sampling plot and shoot-age the total number of needle pairs and needle pairs per cm were pooled (Tab. 3). The data in Fig. 3—7 give both types of values relative to the corresponding plot values from the current year (1974), referred to as 0 year. The age with a 50 per cent reduction of the relative

Sample plot designations	Vegetation type
N1	near clear felled-area, dry dwarf shrub type
S 1	boulder rich hillside slooping abruptly down to the industry, mesic dwarf shrub type
S2	old pine forest, mesic dwarf shrub type
S3	thinned stared mesic dwarf shrub type
S4	boulder rich pine forest, near railroad, mesic-dry dwarf shrub type
SO1	relatively-young stand within Koskullskulle community, mesic dwarf shrub type
SO2	relatively young pruned stand, dry dwarf shrub type on a slope, exposed to industry
SO3	older stand of pine, very dry dwarf shrub type
01	old pine forest, boulder rich ground, mesic dwarf shrub type
OSO1	thinned loftly pine stand at the foot of slope, dry dwarf shrub type
02	older stand, dry dwarf shrub type
OSO2	dry dwarf shrub type near clear-felled area
O3	old stand, dry dwarf shrub type
NO2	mesic dwarf shrub type (Empetrum-type), spruce and birch dominating
NO1	dry dwarf shrub type, bole scars on pine from fire, hillside slooping towards south
SV1	mesic dwarf shrub type (Empetrum-type), birch dominating
SV2	dry dwarf shrub type, relatively young stand, thinned
SV3	moist dwarf shrub type, near clear-felled area
SV4	near mire, wet dwarf shrub type, spruce dominating
NV5 ¹	pine forest heath, mesic dwarf shrub type
NV4	old pine forest with resinous wood (Peridermium), very dry dwarf shrub type
NV3	very dry dwarf shrub type
NV2	dry dwarf shrub type near newly clear-felled area exposed to industry
NV1	near the industry, relatively young stand, mesic dwarf shrub type

Table 2. Some vegetation characters of the sample plots around the LKAB sintering plant, Vitåfors, Norrbotten. Classification system according to Arnborg (1964).

¹ Located 23.5 km, 382° (WNW) the emission source near Sjaunja National Park; other sample plots according to Fig. 1.

needle frequencies and the relative number of needle pairs per cm was obtained by interpolation.

2.5 The lichens

At the same time as the collection of pine material and recording of lichens on the individual sample plots materials of *Hypo*- gymnia physodes (L.) W. Wats., Nephroma arcticum (L.) Torss. and Alectoria spp. were collected for further measurements in the laboratory. Lichens were also taken on a later occasion (11–13.11 1974, see Material II in Tab. 8). The materials were air-dried and then kept dry at $+4^{\circ}$ C in the dark until further analyses, which were carried out within two weeks after the collection.

Before the start of the photosynthetic or nitrogen fixation measurements the lichens were rewetted in tap water for one minute, rinsed and transferred to moist filter paper into a growth chamber. There they were kept for 14—18 hours at 75 per cent relative humidity and $+18^{\circ}$ C in continuous fluorescent light (General Electric Power Growth, type "Daylight" and "White de Lux" in the proportion 1:1). The irradiance was 55 W.m⁻² measured with a Moll-Gorzynski Solarimeter (Kipp & Zoonen CM2).

After this pretreatment lichens of the same species but from different sample plots were at the same time transferred into a plexiglas-chamber for the photosynthetic maasurements. The conditions were the same as described above but with a circulating air supply containing about 350 ppm CO₂ with ¹⁴CO₂ (0.5 Ci.mol⁻¹) during one hour. Immediately afterwards the lichens were killed by being submersed in liquid nitrogen and then dried at +104°C for 24 hours. Parallel samples were taken for water content determinations. After weighing the samples were combusted (Packard Tricarb Sample Oxidizer, Mod. 306) and the evolved CO₂ was trapped in Carbosorb. The samples-10-20 per species and sample plot-were counted by liquid scintillation (Packard No. 3375) with Permafluor V as a scintillation liquid. After appropriate quenching corrections the photosynthetic values have been expressed as the quantity of incorporated ¹⁴C per 10 mg dry weight and hour.

The nitrogen fixation activity was studied on excised cephalodia from *Nephroma arcticum*. One piece from each thallus, 12 mm in diameter containing one cephalodium, was puched out and put into a 7 ml glass bottle. The ethyne reduction technique earlier described (Hällgren and Huss 1975) was used. The nitrogen fixation activity has been expressed as μ mol C₂H₄ produced per cm² and hour.

2.6 The fluorine and sulphur analyses

The materials from the recording of the needle frequencies were preserved in paper bags at $+4^{\circ}$ C until further analyses. The fluorine and sulphur content were determined at the Department of Chemistry, Agricultural College, Uppsala according to Kirsten (1976). The determinations were done on duplicated, random samples with 20 needle pairs in each sample. The results have been expressed as ppm fluorine and as weight per cent sulphur of the needle dry weight (+110°C, 48 hours). The determinations of the needle content of water soluble fluoride was performed by G. Nilsson, the Laboratory of LKAB, Vitåfors.

3 Results

The well-known needle drop increase of coniferous trees in air polluted areas has in this case been quantitatively recorded as the number of needle pairs in relation to the age and the length of the corresponding shoot. The results show an extended lifespan of the needles up to nine—ten years in this area. The total numbers of needle pairs per year and sample plot are seen in Tab. 3 and expressed as number per unit length of

Table 3. The total number of needle pairs of Scots pine, *Pinus silvestris* L. per age-class of needles and sample plot.

Each figure represents the total number from four investigated trees and two branches per tree. 24 sample plots. Collected 27.8—5.9 1974 around the LKAB sintering plant, Vitåfors, Norrbøtten. For plot designations, cf. Figure 1 and Table 2.

Sample	Total number of needle pairs per age-class of needles											
plot	0*	1	2	3	4	5	6	7	8	9	10	Sum
NV5T	534	668	462	521	475	384	51	15	0	0	0	3.110
NV5B	323	405	291	260	283	224	97	99	14	0	0	1.996
NV4	365	403	330	318	226	117	42	2	0	0	0	1.803
NV3	434	502	330	400	248	162	86	0	0	0	0	2.162
NV2	324	324	212	268	258	190	119	23	0	0	0	1.718
NV1	400	424	297	288	290	153	79	10	0	0	0	1.941
NV1A	408	419	308	360	333	251	223	83	36	0	0	2.421
NV1B	431	422	217	234	24	2	0	0	0	0	0	1.330
SO1	538	549	414	329	146	88	29	13	3	0	0	2.109
SO2	451	557	350	339	192	158	71	11	0	0	0	2.129
SO3	437	482	307	436	250	251	147	33	45	1	0	2.389
NO2	480	484	312	304	211	138	81	69	0	0	0	2.079
NO1	274	293	187	254	225	208	200	54	0	0	0	1.695
SV1	429	491	338	400	305	152	61	25	0	0	0	2.201
SV2	369	419	276	281	213	161	75	22	0	0	0	1.816
SV3	495	516	380	484	414	212	34	0	0	0	0	2.535
SV4	405	383	295	342	269	209	65	26	0	0	0	1.994
N1	300	396	245	349	181	137	41	30	19	7	0	1.705
S 1	338	332	154	143	138	56	24	24	5	0	0	1.214
S2	386	557	258	338	282	168	25	0	0	0	0	2.014
S 3	238	307	189	199	159	134	58	6	0	0	0	1.290
S4	375	429	307	333	177	139	144	52	8	1	0	1.965
01	350	280	197	251	213	151	121	72	23	0	0	1.658
OSO1	459	525	321	340	352	220	228	35	0	0	0	2.480
02	339	407	247	272	287	205	105	52	26	14	0	1.954
OSO2	385	408	241	221	133	124	53	6	0	0	0	1.571
O3	372	391	248	283	239	223	72	19	0	0	0	1.847
Sum	10.639	11.773	7.713	8.547	6.523	4.617	2.331	781	179	23	0	53.126
x	394	436	286	317	242	171	86	29	7	1	0	
%	100	111	73	80	61	43	22	7	2	0.2	0	

* The age-class 0 represents needle pairs developed during the year 1974.

Table 4. The mean numbers of needle pairs per unit length of shoot and corresponding shoot lengths of Scots pine, *Pinus silvestris* L. from some sample plots around the LKAB sintering plant, Vitåfors, Norrbotten. 27.8-5.9 1974.

Sample plot	Shoot and needle age year	Number of needle pairs per cm shoot length (n.p.cm ⁻¹)	Corresponding shoot lengths cm (s.1.cm)	Average number of needle pairs per shoot (n.p.cm ⁻¹ ×s.l.cm)
NV4	01	$13^2 \pm 1^3$	3.7±0.6	49
	5	6 ± 2	2.7 ± 0.5	17
NV1	A-trees			
	0	20 ± 2	2.8 ± 0.5	56
	5	14 ± 2	2.5 ± 0.4	36
	B-trees			
	0	18 ± 2	3.1 ± 0.5	57
	5	0 ± 0	2.9 ± 0.2	0
SO3	0	15 ± 1	3.8 ± 0.5	58
	5	12 ± 2	2.8 ± 0.4	33
NO2	0	13 ± 1	4.7 ± 0.6	63
	5	8 ± 3	2.8 ± 0.4	22
S1	0	17 ± 1	2.6 ± 0.2	43
	5	2 ± 1	2.3 ± 0.4	5
SV4	0	15 + 1	3.7 ± 0.7	54
-	5	11 ± 2	2.9 ± 0.6	33

For further details, cf. Figure 1 and 7 B.

¹ represents the investigation year 1974.

² mean value from four trees, two branches per tree, n = 8.

³ mean error.

shoot for some sample plots in Tab. 4. In Fig. 3 B-7 B and 3 C-7 C the results are given for the relative frequencies of needle pairs and the relative numbers of needle pairs per unit length of the shoot, respectively. The needle frequencies naturally decrease with age but deviations are evident among the three-five years old needles. The relatively increased number of needles from 1973 year's shoots is also evident. As seen in Fig. 3 B-7 B the estimated needle age at a 50 per cent reduction of the needle frequencies has a variation width from 1.8-6.4 years between different sample plots. Correspondingly the results in Fig. 3 C-7 C show a broad variation for the 50 per cent reduction between 2.3-6.3 years of the number of needle pairs per unit length of the corresponding shoot lengths. Similar types of irregularities also can be observed here among the three-five year old needles, but the 1973 year's peaks are rarely found (for exceptions, see S2 and SV4). This may suggest a maximum, correlative value for the number of needle pairs per unit length of the corresponding shoots.

In spite of the fact that the used interpolation method has not been taken into account the variation between individual branches and individual trees inside each sampling plot (cf. Tab. 4) it is suggested that the needle pair frequencies and the number of needle pairs per unit length of the shoots of most of the materials show a time interval of 4—6 years for the 50 per cent reduction level. However, in some cases there is both a marked tendency for a shorter life-span of the needles at the 50 per cent reduction level (sample plots S1, SO1, SO2, OSO2, NO2, NV1/B) and at the same time absence (with exception for NO2)

Table 5. Ranking list for the estimated needle ages of Scots pine, *Pinus silvestris* L. at a 50 per cent reduction of the number of needle pairs in relation to the numbers for the current year (1974). From sample plots around the LKAB sintering plant, Vitåfors, Norrbotten 27.8—5.9 1974.

Ranking list	Sample plot	Needle age at a 50 % reduction of the number of needle pairs year*
1	NO1	6.4
2	NV1A	6.2
3	02	5.8
3	OSO1	5.8
4	O 1	5.5
5	NV5T	5.4
5	NV2	5.4
6	NV5B	5.3
7	SO3	5.2
7	S3 -	5.2
7	O3	5.2
8	SV4	5.0
9	S2	4.8
9	SV3	4.8
10	NV1	4.6
10	N1	4.6
11	SV1	4.5
11	SV2	4.5
12	NV4	4.4
13	NV3	4.3
14	S4	4.0
15	NO2	3.6
15	SO2	3.6
16	OSO2	3.3
17	SO1	3.2
18	NV1B	2.3
19	S 1	1.8

* Interpolated values from Fig. 3 B-7 B.

of the above mentioned irregularities for the three—five year old needles. Thus in these cases an enhanced needle drop may indicate a disturbance of their environment. In order to demonstrate any possible correlation to the emission source all the estimated ages for the 50 per cent reduction of the number of needle pairs per unit length have been plotted in Fig. 8 in relation to distance and direction from the emission source. As shown there is no dramatical change of the life-span of the needles in relation to the emission source except for the localities immediately south (S1) and southeast (SO1 and SO2) the sintering plant, where the intensified needle drop could be caused by emissions from the plant.

A ranking list for the ages at the 50 per cent reduction of the needle frequencies (Tab. 5) shows a similar correlation concerning the sample plots relatively close to the emission source. However, this list also shows a pronounced accelerated needle drop at the sampling plot OSO2, 11.5 km ESE of the emission source. The reason for this may be an unfavorable local climate (cf. the profile in Fig. 5 A). Also the pines at the elevated locality NO2 (Fig. 6 A) at the forest limit may be suspected to be primarily influenced by unfavorable climate conditions rather than by the emission from the sintering plant. The samples from the plot NV1, close to the emission source (cf. Fig. 7 A) are supposed to be influenced from the industry both by its proximity and the observed loading of the needles with dust of iron oxides. By use of the same sampling technique (cf. the methods), however, the results in Fig. 7 B/NV1 and 7 C/NV1 do not suggest any effects. A selective collection of material from two vigorously growing trees and two of an inferior kind from the same plot in which the branches from the two types were analysed separately gave the results presented in Fig. 7 B/NV1/A and 7 C/NV1/B, respectively. Here an apparent difference between the reactions of different individuals from the same sampling plot is evident (see also Tab. 4). This demonstrates at the same time the risks of biasing the results by selective sampling. By use of the technique here generally adopted with sampling from two of the best and two of the inferior trees from each sampling plot neither an over- nor an underestimation of the environmental effects on the needle drop of the Scots pine trees probably has been attained.

If thus the observed cases with an accelerated needle drop is related to the distance and the direction from the emission source evidences for a 5 km zone of inTable 6. The occurrence of some terricolous and epiphytic lichens on sample plots around the emission source at the LKAB sintering plant, Vitåfors, Norrbotten.

27.8—5.9 1974. + = present; - = absent; for *Alectoria* spp. also estimations of the abundance. For plot designations, cf. Figure 1.

Sample	Terricolous li	chens	Epiphytic licher	ns on Scots pine			
plot	Peltigera	Nephroma	Hypogymnia	Alectoria spp.			
	apritosa	arcticum	physodes	Occurrence	Abundance		
O3	+	+	-4-	+	5		
NV4	+	+	+	+	5		
OSO2	+	+	+	+	5		
NO1	+	+	+	+	5		
S 4	+	+	+	+	4		
NV3	+	+	+	+	4		
OSO1	+	+	+	+	4		
SV4	+	+	+	+	3—4		
NO2	+	+	+	+	3		
N1	+	+	+	+	3		
01	+	+	÷	+	3		
NV2	+	+	+	+	2		
S3	+	+	+	+	2		
SO3	+-	+	+	+	2		
02	+	+	+	+			
SV3	-+-	+	+	+	1		
SV2	+	+	+	-	0		
S2	+	+	+ 1	_	0		
SV1	+	+	+1	-	0		
SO2	+	+	+ 1	_	0		
NV1	+		+ 1	-	0		
S1	+	+ 1	-	_	0		
SO1	+	_	+ 1	-	0		

¹ with visible damages.

fluence with preference for a topographically dependent extension in northwest southeast seems probable.

Since lichens are known as especially sensitive indicators of air quality, studies on the occurrence and activity of some lichens were carried out. Here *Alectoria* spp. (mainly *A. Fremontii* Tuck.) has been used as one of these bioindicators, the occurrence of which and estimated abundance in the area are presented in Tab. 6 and Fig. 9. The results support the evidence from the needle drop studies above and are also consistent with an earlier investigation of *Alectoria* spp. and *Usnea* spp. in the area (Nilsson, unpubl. report 1974). Also thalli of *Hypogymnia physodes* were absent close to the industry (S1) and showed morphological damages in other sample plots near the emission source.

The lichens close to the sintering plant were more or less covered with dust, mainly consisting of iron-oxides, and this was also the case of *Hypogymnia physodes* from sample plot NV1. Here the terricolous lichen *Nephroma arcticum* was missing and in the sample plot S1 it frequently showed yellow margins of the thalli, which was not observed in any of the other investigated plots. The terricolous lichen species of *Peltigera* as well as *Cladonia* and *Stereocaulon* from more dry habitats were never observed to have visible damages.

Concerning directly visible damages it may also be mentioned that the pine trees from the sample plots S1, NV1 and SO1, Table 7. Comparisons between the life-span of needles of Scots pine, *Pinus silvestris* L. and the presence of lichens of *Alectoria* spp. in the same sample plots around the LKAB sintering plant, Vitåfors, Norrbotten.

27.8—5.9 1974. For the estimations of the age at the 50 per cent reduction, cf. Figure 3 B and 3 C, respectively.

A: Based on the relative needle frequencies.

B: Based on the relative numbers of needle pairs per unit shoot-length.

The occurrence of Alectoria spp.	Number of sample plots with trees showing a 50 per cent reduction of the needles at an age of							
	A		Sum	В		Sum		
	< 5 years	≥5 years		< 5 years	≥5 years			
Number of plots without Alectoria spp.	7	0	7	4	3	7		
Number of plots with Alectoria spp.	7	9	16	3	13	16		
Sum	14	9	23	7	16	23		
	χ. 0.	$P_{2} = 4.323$ 0.5 > P > 0).01*	$\chi^2_{\rm C} = 0.20$	1.819 > P > 0.10)°		

showed partly miscoloured needles (yellowred—greyish-red). This was observed either among the current year's needles or among older ones. Only on very few branches the needles of several ages were simultaneously discoloured. It must be emphasized that the majority of the trees was lacking such visible damages.

To evaluate the similarities in reaction to the environment of Scots pine and the lichen *Alectoria* spp. the number or sample plots with and without the occurrence of *Alectoria* and the corresponding needle drop reaction of the pines are given in Tab. 7. These results suggest a positive relationship between the occurrence of needle ages of five years or older and the presence of *Alectoria* in the same plots thus supporting a similar reaction of the different systems in relation to the emission source.

Not only directly visible effects of an actual air pollution are of interest, but also possible disturbances of the basic physiology of the plants in such an environment. Therefore the photosynthesis and nitrogen fixation of some of the affected lichen species mentioned above were studied. Material of *Hypogymnia physodes* from plots inside an area of 5 km radius from the

emission source showed pronounced diminished photosynthetic capacity than material from more distant localities (Tab. 8, Fig. 10). This was evident even in cases, where no directly visible damages were observed (cf. Tab. 6). A similar correlation between the photosynthetic activity of the Alectoria samples and the proximity to the emission source could not be established, however. A partly repeated investigation of the same area two months later confirmed in the main these differences between Hypogymnia and Alectoria. Concerning the nitrogen fixation capacity of excised cephalodia of Nephroma arcticum this was significantly lower-tested with Wilcoxon's test-in samples from the plots S1 and SV1 in comparison with material from more distant plots (SV2 and SV3, Tab. 9). It was also observed that several samples from the localities S1 and SV1 did not show any nitrogen fixation activity at all.

A further approach to mapping the actual imission situation was carried out by means of analyses for fluorine and sulphur in pine needle material.

By kind permission from G. Nilsson, the LKAB Laboratory, Vitåfors, the results from a study on the water-soluble content

Table 8. Photosynthetic ¹⁴CO₂-fixation by the epiphytic lichens *Hypogymnia physodes* (L.) W. Wats. and *Alectoria* spp. from sample plots around the LKAB sintering plant, Vitåfors, Norrbotten.

Sample plot	Photosynthetic Dpm • 10 ³ • 10 1					
	Hypogymnia pl	nysodes	Alectoria spp.			
	Material I	Material II	Material I	Material II		
NV4	20.3 ± 1.89	n.i.	7.1 ± 1.27	n.i.		
NO2	24.4 ± 1.92	n.i.	n.i.	12.0 ± 0.87		
NV3	20.2 ± 1.62	n.i.	4.3 ± 0.50	n.i.		
NV2	4.5 ± 1.40	6.6 ± 0.01	2.5 ± 0.51	5.1 ± 1.3		
NO1	n. i .	n.i.	n.i.	n.i.		
N1	14.7 ± 1.38	12.1 ± 0.77	2.7 ± 0.43	n.i.		
NV1	0.3 ± 0.07	0.1 ± 0.01	n.d.	n.d.		
S1	n.d.	n.d.	n.d.	n.d.		
SO1	0.1 ± 0.01	n.i.	n.d.	n.d.		
01	14.4 ± 1.23	n.i.	3.5 ± 0.92	6.4 ± 0.90		
SV1	n.i.	n.i.	n.d.	n.d.		
S2	n.i.	n.i.	n.d.	n.d.		
OSO1	n.i.	n.i.	2.4 ± 0.44	10.0 ± 1.92		
02	n.i.	n.i.	n.i.	n.i.		
SO2	7.8 ± 0.97	n.i.	n.d.	n.d.		
SV2	n.i.	n.i.	n.d.	n.d.		
\$3	11.0 ± 0.95	n.i.	n.i.	7.1 ± 0.5		
SO3	19.2 ± 1.12	21.4 ± 3.15	3.5 ± 0.58	13.0 ± 1.4		
SV3	n.i.	n.i.	n.i.	n.i.		
S4	20.9 ± 2.49	n.i.	n.i.	n.i.		
OSO2	n.i.	n.i.	7.9 ± 0.87	5.0 ± 0.6		
03	18.1 ± 1.17	n.i.	n.i.	4.2 ± 1.3		
SV4	n.i.	n.i.	n.i.	5.2 ± 0.7		

Material I collected 27.8—5.9 1974, material II collected 11.11—13.11 1974. Measurements done in the laboratory, cf. the methods. For sample plot designations, see Figure 1 and Table 2.

n.i.: not investigated.

n.d.: not detectable.

of fluoride of Scots pine needles from sampling plots around the sintering plant are given in Fig. 11. This material was collected in 1972 and the analyses were done on one—several year old needles. It shows a higher content in material from the nearest 5 km area around the emission source with a marked distribution inside this area in a northwest—southeast direction. The maximum value was 18 ppm fluoride per dry weight from a sample plot close and north to the emission source, the lowest ones were 0.3 ppm from plots at a distance of 10—15 km.

In order to permit a direct comparison

between the fluorine content and the physiological observations presented above a repeated study of the total fluorine content was carried out on material from the needle drop studies and thus from the same sample plots as which where the lichen samples were collected. As a beginning the content of fluorine and sulphur in needles of different ages was studied. The results for fluorine are given in Fig. 12 made on material from plot SO1 in which disturbances have been found (cf. Tab. 5 and 6). The highest accumulation was detected in the needles from the year 3 (1971) declining with increasing age. A comparison between Table 9. The nitrogen fixation activity of excised cephalodia from *Nephroma arcticum* (L.) Torss. collected at different sample plots around the LKAB sintering plant, Vitåfors, Norrbotten.

27.8—5.9 1974. Tested in the laboratory with the ethyne reduction technique.

Sample plot	Nitrogen fixation activity nmol $C_2H_4 \cdot cm^{-1} \cdot h^{-1}$							
	Mini- mum	Mean	Maxi- mum	No. of determi- nations				
S1	0.0	1.6	28.2	28				
SV1	0.0	5.0	29.2	13				
SV2	4.8	10.7	24.7	13				
SV3	1.1	12.4	29.0	12				

the fluorine content of the current year's needles and the three years old ones showed about a fourfold increase not reflected in the corresponding hydrogen fluoride emission from the industry (cf. Fig. 1). Concerning the sulphur contents these varied between 0.08-0.09 per cent of the dry weight for the needle ages one—five years. The only tendency to a higher content was observed in the needles from the current year (1974, 0.13 %).

Based on these results the main analyses

were carried out on three years old needles from the different plots. The results for the fluorine content are given in Fig. 13 as a survey in relation to the emission source. Here all analytical data represent the total fluorine content. The highest one was observed in needles collected close to the sintering plant (plot S1) representing a content of 163 ppm, which may be compared with the minimum value 10 ppm (from the plot NV4), the latter obviously reflecting the magnitude of the fluorine content of Scots pine needles from sampling plots more than 5 km from the emission source. Concerning the distribution of the highest values a main direction in northwest-southeast is suggested also in this case.

The corresponding sulphur analyses were carried out on the same materials, the results of which are given in the same way in Fig. 14. The width of variation was 0.07-0.12 per cent sulphur of the dry weight between the used plots. These results do not permit any conclusions concerning an increased total sulphur content of the Scots pine needles in relation to the emission source. However, it was observed that the material from the plots S1 and SO1 showed the highest mean values inside the variation mentioned above.



Figure 3. The relative needle frequencies and numbers of needle pairs per unit length of shoot of Scots pine, *Pinus silvestris* L. in relation to needle age, distance and topography *north—south* the emission source at the LKAB sintering plant in Vitåfors, Norrbotten.

27.8—5.9 1974. For details concerning the sample plots, cf. Figure 1 and Table 2.

A: Topographical profiles—with 25 metre intervals—between the emission source and the sample plots, respectively. 0-point at the emission source of the industry. Zero level (0 m) at the level of the plant location. The dotted line represents an average height of the smoke-stacks.







C: Relative numbers of needle pairs per unit length of shoot. Interpolated ages (in years) at a 50 per cent reduction related to the current year's (1974) numbers of needle pairs per unit length of shoot.



Figure 4. The relative needle frequencies and numbers of needle pairs per unit length of shoot of Scots pine, *Pinus silvesiris* L. in relation to needle age, distance and topography *southeast* of the emission source at the LKAB sintering plant in Vitåfors, Norrbotten. For further details cf. Figure 3.

A: Topographical profiles.





Figure 5. The relative needle frequencies and numbers of needle pairs per unit length of shoot of Scots pine, *Pinus silvestris* L. in relation to needle age, distance and topography *east—eastsoutheast* the emission source at the LKAB sintering plant in Vitâfors, Norrbotten. For further details, cf. Figure 3.

A: Topographical profiles.

C



B: Relative needle frequencies.



C: Relative numbers of needle pairs per unit length of shoot.



Figure 6. The relative needle frequencies and numbers of needle pairs per unit length of shoot of Scots pine, *Pinus silvestris* L. in relation to needle age, distance and topography *northeast southwest* of the emission source at the LKAB sintering plant in Vitåfors, Norrbotten.

For further details, cf. Figure 3. A: Topographical profiles.







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C: Relative numbers of needle pairs per unit length of shoot.



Figure 7. The relative needle frequencies and numbers of needle pairs per unit length of shoot of Scots pine, *Pinus silvestris* L. in relation to needle age, distance and topography *northwest* the emission source at the LKAB sintering plant in Vitåfors, Norrbotten.

For further details, cf. Figure 3.

A: Topographical profiles. Note that the profile for the sample plot NV5 is not included in this figure. Cf. Table 2.



B: Relative needle frequencies. Note that NV1/A represents two apparently vigorous trees from the sample plot NV1, NV1/B two distributed ones in the sample plot. NV1 shows the mean values from four randomly collected trees in the same location. In the plot NV5 material was sampled in the usual way (NV5/B—bottom branches) and also from the corresponding top-branches (NV5/T).





Figure 8. The distribution of needle ages at a 50 per cent reduction of the relative numbers of needle pairs per unit length of shoot of Scots pine, *Pinus silvestris* L. in the sample plots around the LKAB sintering plant in Vitåfors, Norrbotten.

Emission source in the origo. Column height at the sample plot S1 south of the emission source represents 2.3 years, other column heights in corresponding proportions. 27.8—5.9 1974.



Figure 9. The estimated abundances of the lichens *Alectoria* spp. on Scots pine, *Pinus silvestris* L. in the sample plots around the LKAB sintering plant in Vitâfors, Norrbotten.

Emission source in the origo. Column heights proportional to the estimated abundances (cf. Table 6). 27.8–5.9 1974.



Figure 10. A survey of the photosynthetic capacity of the lichen *Hypogymnia physodes* (L.) W. Wats collected on Scots pine trees, *Pinus silvestris* L. in the sample plots around the LKAB sintering plant in Vitåfors, Norrbotten.

Emission source in the origo. The column heights proportional to the photosynthetic capacities, respectively (cf. Table 8). 27.8—5.9 1974.



Figure 11. A survey of the water soluble content of fluoride in needles of Scots pine, *Pinus silvestris* L. from locations around the LKAB sintering plant in Vitåfors, Norrbotten.

Emission source in the origo. The column heights proportional to the fluoride contents, respectively expressed as ppm of the needle dry weight. The maximum value corresponds to 18 ppm, the minimum ones to 0.3 ppm. Analyses carried out with ion-selective electrode in water extracts from one to several year old needles collected 22.6–11.7 1972. After data from G. Nilsson, 1973, the Laboratory of LKAB, Vitåfors, Norrbotten.



Figure 12. The fluorine content in needles of different ages of Scots pine, *Pinus silvestris* L.

Material from the plot designed SO1, 2.5 km southeast of the emission source in Vitåfors, Norrbotten collected 27.8—5.9 1974. The contents are related to the dry weight $(+110^{\circ}C, 48 \text{ h})$. Mean values from duplicate analyses of 20 randomly selected needles of the ages, respectively. Concerning the analyse method, cf. p. 10.



Figure 13. A survey of the fluorine content in three years old needles of Scots pine, *Pinus silvest*ris L. from the sample plots around the LKAB sintering plant in Vitåfors, Norrbotten.

Emission source in the origo. The column heights proportional to the fluorine contents expressed as ppm of the needle dry weight. The maximum value (sample plot S1) corresponds to 163 ppm, the minimum one (in NV4) to 10 ppm. Concerning the analyse method, cf. p. 10 and Figure 12.



Figure 14. A survey of the sulphur content in three years old needles of Scots pine, *Pinus silvestris* L. from the sample plots around the LKAB sintering plant in Vitåfors, Norrbotten.

For details, cf. Figure 13. The maximum column heights correspond to 0.12 % sulphur of the needle dry weight (sample S1 and SO1), the minimum ones to 0.07 %.

4 Discussion

Any studies on physiological phenomena under ecological conditions naturally have to be faced with a lot of variable conditions. Not only variations as here of the qualities and quantities of pollutants but also intraand interspecific variation of the responses make the evaluation of such an ecophysiological study difficult. In cases where the pollution as in this Vitåfors case consists of several toxic gases together the analysis may be even more complicated. The use of some alternative and independent techniques at the same time may be of help in such a situation.

The recording of damages or other deleterious effects on coniferous trees in air polluted areas generally includes a statement of an accelerated needle drop. Such habitus changes are described by e.g. Pelz and Materna (1964) but attempts to quantify them seem to be lacking. Other quantitative studies have been done on the annual, radial growth (Lux 1965, Vinš 1965, 1971, Pollanschütz 1972, 1975, Huttunen 1975 among others), the annual height growth (Scheffer and Hedgecook 1965, Wentzel 1971) or combinations of both (Vinš 1965), the success of which all are dependent on the establishment of a representative control level.

The present attempt to evaluate the influences on the Scots pine trees has been done with a quantitative method for the needle drop, which depending on its analytical simplicity and at the same time maximum life-span of the needles may offer some advantages. The life-span of Scots pine needles varies from three to four years in the southern parts of the country up to eight to ten years as found in this case in the northernmost part of Sweden. By recording the number of needle pairs in relation to the ages and the length of the corresponding annual shoots followed by an interpolation as estimation of the ages at a 50 per cent reduction of the needles was received. By reference to corresponding data from remote localities this method in a simple way may permit the showing of any influence from emission sources. In the present case this method also permitted the demonstration of differences in susceptibility to the actual emission between Scots pine individuals, a phenomena which earlier has been shown and discussed both for this and other tree species (Vogl and Börtitz 1968, Dässler et al. 1972).

The diagnosis of fluorine and sulphur dioxide damages from the needle appearance is a disputable task (Treshow and Pack 1970, Stewart et al. 1973). The few here observed discoloured needles could not be identified as typical for either fluorine or sulphur dioxide damages.

The content of fluorine or sulphur in needles of different trees, from different sample plots and of different ages may show considerable variations (Guderian 1970, Stephan 1972, 1975, Halbwachs 1975). In order to decrease this phenomenon all analyses presented here have been restricted to a selected age of the needles, which in the introductory studies showed the highest content of fluorine. Earlier investigations have shown negligible differences between different needle-ages from pollutant-free areas but higher contents in older ones from areas with immission (Guderian 1970, Stephan 1975), which, however, has not been possible to reinvestigate here. The reason for the three years old needles to show the highest fluorine content is not known. One explanation could be that the needles more than three years old are more inclined to drop with an individually increased fluorine content. A redistribution within the plant of this element, earlier considered as rather immobile, has been shown to exist by Keller (1974). This may be another explanation as good as more leaching by rainfall from the older needles.

The fluorine analyses have shown an increased content of both water-soluble fluorides and total fluorine with a decreased distance to the emission source and with maximum values in the close neighbourhood of it. At a distance of about 5 km or more the total fluorine content was about 10 ppm. This is comparable to the fluorine levels reported for Scots pine from rural areas without emission of fluorides (Dässler 1969, Havas 1971, Stephan 1972). In the present case all the needles were collected from the lower parts of the crowns. Perhaps a more differentiated and clear picture could have been obtained with needle materials from the top of the trees, where a higher total immission may be more probable. Nevertheless, the figures point out that in this ecosystem with an expected slow turn-over rate of elements, considerable amounts of fluorine may accumulate due to the present emission source. In order to find out the consequences of this for the future a thorough investigation of the total fluorine distribution in this ecosystem ought to be done.

Concerning the sulphur contents evidence for a higher sulphur content possible to correlate with the emission source are not proved. The values found here, 0.07--0.12 per cent sulphur of the dry weight seem to correspond to the content of needles of Scots pine from emission free areas (Themlitz 1960, Pavlik 1965 quoted from Stephan 1975). Stephan (1972) has also pointed out that generally the sulphur content need not be markedly changed in relation to any actual emission source of sulphur dioxide. Corresponding results have also been presented by Guderian (1970).

Zonation of epiphytic lichens around more or less isolated industrial plants are well documented (Hawksworth 1973, Gilbert 1973 b). *I.a.* this has been shown even for a sintering plant in Wawa, Ontario, Canada by Rao and LeBlanc (1967). In that case, however, only sulphur dioxide was emitted and in much higher amounts than in Vitåfors, which along with other ecological differences hamper further comparisons. Under the climatic conditions in the Vitåfors area epiphytic lichen growth is not favoured. Normally the trunks of the Scots pine trees have a very poor lichen growth although occasionally on older ones there was a luxuriant growth of Alectoria and Usnea spp. Concerning Alectoria spp. these were completely lacking in the sample plots near the sintering plant, but the distribution was not exclusively possible to correlate to the emission source. In several sample plots northwest-east of the plant Alectoria spp. were abundant, regardless of the proximity to the smokestacks. In remote localities specimen of A. Fremontii often reached 40 cm length, while they generally were much shorter closer to the emission source. Here it may be noted that the recordings of the Alectoria spp. have not been strictly separated with regard to different species at the same time as A. Fremontii has been noticed as a common one in the area. Therefore it may be of interest to note that the same species has been shown to be one of the more tolerant lichens to airpollution from a study around a pulp-mill at Örnsköldsvik, Sweden (Westman 1975). This perhaps also may illustrate the lack of complete conformity observed between the occurrence of Alectoria spp. and the premature needle drop of Scots pine (Tab. 7).

It may also be emphasized that, in case where several gases toxic to lichens are simultaneously present, their eventual synergistic effects are difficult to evaluate (Hawksworth 1973) as well as interindividual variations of susceptibility. For example, in sample plots near the sintering plant epiphytic lichens such as Hypogymnia physodes, Parmelia olivacea (L.) Ach. and P. sulcata Tayl. could vary from being completely white, small and crustaceous or appearing with red-brown, curled margins or looking absolutely unaffected. This probably reflects ecotypic differences, which are supposed to be genetically determined (Gilbert 1973 a). Also the possibilities for modifying effects of shelters may be observed under ecological conditions. In transplant and laboratory studies, however, several lichens have been proved to be sensitive also to hydrogen fluoride (Pyatt 1970, Nash 1971, LeBlanc et al. 1971, 1972, Gilbert 1971, Börtitz and Ranft 1972). Its deleterious effects on *Hypogymnia physodes* have also been described by Comeau and LeBlanc (1972). In spite of this the diversified reactions of the observed lichens could not give any indication of the ultimate source of the harmful effects.

It has earlier been shown from transplant studies in air-polluted areas (Kallio and Varheenmaa 1974) and at laboratory investigations (Hällgren and Huss 1975) that nitrogen fixation and photosynthesis of lichens are easily disturbed by sulphureous air pollutants. Richardson and Puckett (1973) have also pointed out the disturbed photosynthesis as one of the first detectable effects on lichens in such air-contaminated environments (cf. also Sundström and Hällgren 1973). The ultimate dependence of the lichen growth on an unaffected physiological status thus may indicate the value of such an experimental approach for evaluation of environmental disturbances. Even if the present results are restricted in number and not unequivocal for all sampling plots concerning correlations between growth reactions, photosynthesis and nitrogen fixation (cf. Tab. 6 and 8, see also Fig. 9 and 10) they may support the further use of measurements on the photosynthetic and nitrogen fixation activities of lichens as sensitive, quantitative and rapidly available expressions for influences of air-pollution.

Even if the results presented here from combined ecological, physiological and chemical studies on different plant species around an emission source of mainly sulphur dioxide and hydrogen fluoride suggest a zone of influence within a radius of 5 km-in mainly northwest--southeast direction on account of topographical reasonsthis does not exclude the possibilities for the occurrence of a wider affected area possible to detect by the use of other and more sensitive analytical techniques and perhaps other species. The importance of studies on such low dosage conditions has also been stressed by Smith (1974) for a better understanding of the real impact of air pollution on forest ecosystems.

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Sammanfattning

Barrfällningstendenser hos tall jämte förekomst av och fysiologiska reaktioner fotosyntes och kväveassimilation — hos några lavarter har studerats på material från lokaler omkring LKAB:s sintringsanläggning vid Vitåfors, Malmberget, Norrbotten. Därjämte har kemiska analyser av tallbarr med avseende på fluor- och svavelhalt utförts. Emissionen av fluorväte och svaveldioxid i området uppgår till genomsnittligt 175 ton respektive 2000 ton per år.

Inom en yta med radien 5 km från emissionskällan kunde en förhöjd fluorhalt med maximalvärdet 16 ggr bakgrundsvärdet noteras. Analyserna utfördes på treåriga barr, vilka vid inledande undersökningar visat den högsta ackumuleringen av fluor. Ett motsvarande åldersberoende för svavelhalten kunde ej observeras, ej heller kunde någon klart förhöjd svavelhalt i förhållande till emissionskällan noteras.

Genom registrering av antalet barrpar per skottaxelgeneration och dito längd samt interpolation erhölls information om den genomsnittliga barråldern vid 50 %-ig reduktion i förhållande till barrförekomsten under undersökningsåret 1974. En accentuerad barrfällning kunde därvid sättas i relation till närheten till emissionskällan även om exempel erhållits på avvikande, extrem individuell resistens. Påskyndad barrfällning kunde noteras inom samma område, som ovan visats ha ökad fluorbelastning.

Lavförekomsten och eventuella, direkt synliga skador på lavbålarna har registrerats

för Peltigera aphtosa (L.) Willd., Nephorma arcticum (L.) Torss., Hypogymnia physodes (L.) W. Wats. och Alectoria spp., varvid successivt ökad känslighet i nämnd ordning har observerats. Jämförande studier över fotosyntesaktiviteten med ¹⁴C-teknik på Hypogymnia-material och kvävefixeringsförmågan hos Nephroma-material med etynreduktionsmetoden har vid mätningar under kontrollerade laboratorieförhållanden visat störningar hos respektive material, vilka har kunnat sättas i relation till närheten av emissionskällan vid provtagningstillfället. Vidare har viss samvariation mellan lavförekomst (Alectoria spp.) och tallens barrfällningstendens noterats.

Kemisk, växtfysiologisk och ekologisk analysteknik på tall- och lavmaterial har sålunda var för sig visat på en påverkanszon omkring sintringsanläggningen, vars detaljer emellertid synes vara mer beroende av lokala topografiska än allmänna vindklimatologiska förhållanden. Observerade effekter på det biologiska materialet har dock ej explicit kunnat relateras till någon av de aktuella imissionsprodukterna.

Svårigheterna vid utvärdering av påverkanszon från emissionskällan har diskuterats i förhållande till använt biologiskt material och analytisk teknik. Värdet av lavfysiologiska undersökningar i sammanhanget har betonats. Slutligen har framhållits det angelägna i vidgad kunskap om förhållanden i svagt immissionsbelastade områden, där ännu inga direkt synliga skador kan observeras.

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