# ACTA FORESTALIA FENNICA

## Vol. 121, 1971

Injury to Pines in the Vicinity of a Chemical Processing Plant in Northern Finland

Männyn vaurioista erään Pohjois-Suomen kemiallisen tehtaan läheisyydessä

Paavo Havas

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SUOMEN METSÄTIETEELLINEN SEURA

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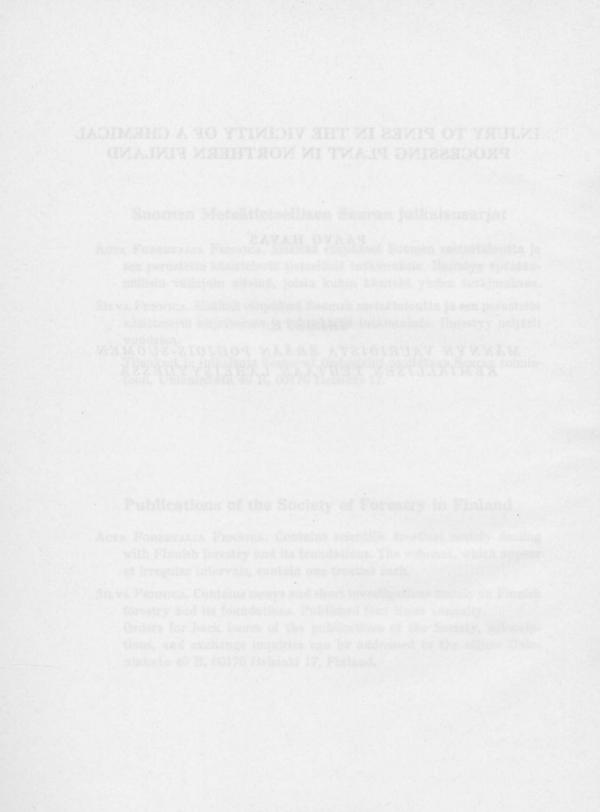
### INJURY TO PINES IN THE VICINITY OF A CHEMICAL PROCESSING PLANT IN NORTHERN FINLAND

### PAAVO HAVAS

### SELOSTE:

MÄNNYN VAURIOISTA ERÄÄN POHJOIS-SUOMEN KEMIALLISEN TEHTAAN LÄHEISYYDESSÄ

HELSINKI 1971



Suomalaisen Kirjallisuuden Kirjapaino Oy Helsinki 1971

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### **1. INTRODUCTION**

The spring of 1966 was the first time when any largescale damage was noted in the trees, particularly pines (*Pinus silvestris*) and birches (*Betula pubescens*), surrounding a processing plant, Typpi Oy, which produces nitrogen fertilizers in Oulu ( $65^{\circ}$  N). Since that time, the damage has increased to such an extent that the area of serious damage in 1970 comprised appr. 250 hectares. Typpi Oy financed the comprehensive investigations which were carried out in winter 1969—70, in order to elucidate the causes of the damage to the pines. Typpi Oy has had the condition of the trees observed since 1966, and chemical analyses have been carried out to determine the amounts of fluorine, sulphur and nitrogen present in the needles of the pines and the leaves of the birches.

The emission inventory (SÖYRINKI & ES-TAMA & MERILÄINEN 1970) of the department of Typpi Oy producing mixture-type fertilizers, which started operating in 1957, shows a fluorine emission (hydrogenfluoride, silicon tetrafluoride, and the other fluorides present in the dust of mixture-type fertilizers) of 20—30 tons/year. The sulphur emission of Typpi Oy is probably appr. 450 tons/year, and the emission of nitrogenoxides appr. 1000 tons/year, calculated as elementary nitrogen.

### 2. MATERIALS AND METHODS

The following pine samples were used: 1) Trees growing on the damaged area (mainly NW of Typpi Oy, where the damage is most severe) and a control material of trees from an unpolluted area. 2) During the winter 1969-1970, pines of 2-4 meters were felled on the unpolluted area (6-7 km SW of Typpi Oy, on the southern side of the town). Part of these were thereafter brought to the site of Typpi Oy, part were left on the unpolluted area. Thus the pines brought near the process operations came under the pollutant infuence as quickly and effectively as possible. The first few of these »test trees» were placed on the designated field on Nov. 4th 1969 (the socalled November trees). More trees were brought on February 4th, March 9th and April 2nd 1970 (the February, March and April trees). A Number of these trees were protected in such a way that they were not, at least immediately, exposed to the effect of the fallout of particulate pollutants such as fertilizer dust.

The needles of the pines investigated were divided into two groups: those over 1 year old, and those of shoots less than 1 year old. Before chemical analyses the needles were rinsed (appr. 2 min/sample) on a sieve with a spray of distilled water. After that they were dried at 90° C, ground, and preserved in an exicator until analyzed. The determinations of fluorine, sulphur, nitrogen, potassium and phosphor were carried out in the laboratories of Typpi Oy by the following methods.

Fluorine: ashing, NaOH-fusion, suspension in water, distillation from  $HCl0_4$  by water vapour, assay of NaOH-solution by the spectrophotometric alizarine method. Sulphur: Na<sub>2</sub>O<sub>2</sub>-Na<sub>2</sub>CO<sub>3</sub>-fusion, suspension in water, filtering, precipitation as BaSO<sub>4</sub>. Nitrogen: the wet-Kjelldahl method. Potassium: ashing, dissolution in HCl, atomic absorption spectrophotometry. Phosphor: Mg(NO<sub>3</sub>)<sub>2</sub> treatment, ashing at 500° C, dissolution in H<sub>2</sub>SO<sub>4</sub>, spectrophotometric molybdophosphate method.

The pH determinations of the washing water of the needles, melted snow and frost were carried out using Beckman's Expandomatic pH meter in the Botany Department of the University of Oulu.

The chlorophyll present in the needles was also determined in the Department of Botany: the needles were ground in a Sorwall Omni-Mixer and extracted in acetone,  $Ca(CO_3)_2$  was added, the mixture was centrifuged (5 min., 5000 rpm) and filtered. The chlorophyll content was determined by a spectrophotometer (Hitachi).

The photosynthesis and respiration experiments on the needles were made in the Department of Botany, using Hartman & Braun's URAS II-analyzer and chambers built for this purpose.

In order to determine the degree of xeromorphism of the pine needles, samples of the most recently grown needles were collected on Oct. 27th 1969 both near Typpi Oy and further away in the unpolluted area (Oulu, Sanginjoki), where samples were taken from fertilized forests on the one hand and from areas not fertilized on the other. All the samples come from dryish, mainly Empetrum-Vaccinium type pine forests, though both near Typpi Oy and in the fertilized forests at Sanginjoki the species of the field and bottom lavers had been somewhat changed by the effect of fertilizers. The samples were obtained about 1.5 meters above the ground from the southern side of nore or less healthylooking trees 2-3 meters high. The average length of the needles was determined from 200 needles of the youngest shoots randomly obtained in the Typpi Oy area, while in the other two areas 100 needles were measured in each case. The other measurements were carried out on preparations made from the middle parts of needles obtained from about 2 cm below the tip of a young shoot. 60 needles were taken from the Typpi Oy area, 30 needles from each of the two others.

The water uptake experiments of pine shoots were made under dark conditions and 0 to  $+5^{\circ}$  C temperature, so as to eliminate the effect of photosynthesis and respiration on the alternation of weight. The relative humidity of the air was 80—100 % throughout the water uptake experiment, during which the shoots were sprayed for appr. 20 seconds at intervals of about 1/2 hour. The shoots, dried on the surface, were weighed twice a day, and the water content was determined at the beginning and at the end of the experiment.

### **3. RESULTS AND DISCUSSION**

### **31.** Symptoms in the Trees

The symptoms in the trees in the neighbourhood of Typpi Oy are clearly visible (fig. 1). Yet it is difficult to say on the basis of these which contaminants have caused the damage (cf. e.g. THOMAS, 1964, p. 230 and TRESHOW, 1970, р. 295—297). The damaged needles are greyish brown or brown. The brown colour is not clearly concentrated to the tips of the needles. There is often a thin (1/2-1 mm), dark brown or reddish brown »band» to separate the dead brown tissue from the live green parts. There is no clear difference in the sensitivity of old and young needles. In the most polluted area the trees in the spring retain little more than the needles grown during the previous summer older needles are turning brown and falling off. Thus the trees keep their needles only for 1-2 years. During the early summer months new needles grow to compensate for the winter defoliation.

It can be clearly seen from the trees especially on and around open places — that the sides facing Typpi Oy are more damaged than the opposite sides. Even in one branch the needles protected by others (from exposure to the effects of the process operations) may remain green, although the ones exposed to the injurious effect are destroyed. Greatest damage has occurred in individual trees growing on open places, and in the margins of forests. It can often be seen that the tops of high trees are more badly damaged than are small trees, which may survive even near Typpi Oy if protected by buildings, etc. Tops of high birches are damaged as far as the centre of the town (2—3 km SW of Typpi Oy), as are also high pines at a similar distance in the NW direction.

Snow cover is a good shield against damage. Small pines and the lowest branches of high pines, which are covered by snow, remain green even in the worst damage areas (fig. 2). This fact, among others, suggests that the injury mainly occurs in winter conditions. In the immediate surroundings of Typpi Oy the leaves of some deciduous trees are badly damaged even in summertime.

The symptoms appear on the pine needles in the spring, though not very suddenly. In

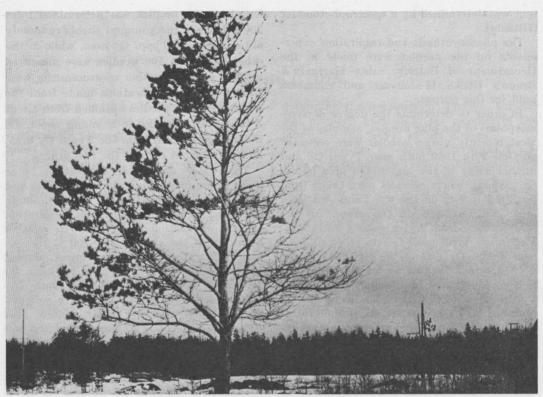


Fig. 1. Damaged pine near Typpi Oy. The parts directly facing the plant are ones most severely damaged.

the spring of 1970, for example, the new injuries were slightly visible as early as the middle of April, but became apparent at the beginning of June.

The symptoms in the test trees brought to the site of Typpi Oy were similar in quality and location to those noted in the growing trees. As the experimental field was situated quite near the processing plants, the damage occured naturally rapid and heavy. Artificial protection, snow and the shelter of other trees provided protection against damage on the experimental field as in natural conditions. At the beginning of May 1970 the needles of the November trees had all turned brown with the exceptions of those under the snow, which were still partly green at that time. At the same time about half of the needles in the protected trees were brown.

It is natural that chlorophyll is destroyed when visible damage appears. This is shown, for example, by the following statistics on the decline of the chlorophyll content in the needles of the unprotected November test trees:

14. 1. 1970	chlorophyll	content	was	17.3	mg/g	of	dry	matter
23. 1. 1970	*	*	*	14.9	*		*	*
9.3.1970	*	*	*	6.1	*		*	*
14. 4. 1970	*	*	*	1.0	*		*	*

Shoots of the test trees were brought to the laboratory during the winter, to allow closer observation of the progress of damage. Brown colour was first noted in the middle of December 1969, when the needles of the November trees brought into the laboratory turned completely brown within a week. Later in the spring the needles turned brown in an even shorter period. These observations show that the process which finally leads to total damage begins as early as midwinter, although the exceedingly slow wintertime

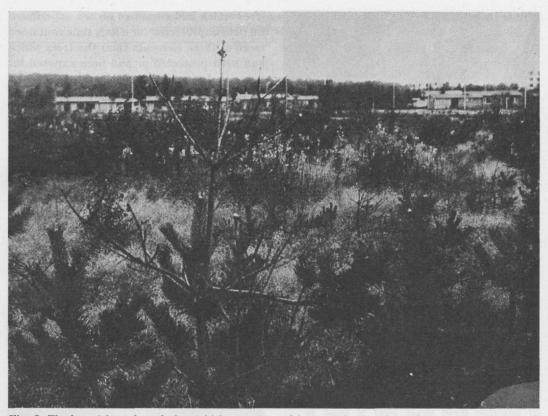


Fig. 2. The lowest branches of pine, which are convered by snow in the winter, have not been damaged. Note also the large proportion of *Deschampsia flexuosa* and *Chamaenerion angustifolium* in the field layer, which is due to the effect of fertilizer.

metabolism of the pines prevents the injuries from becoming visible until the warmer spring. The observations further show that a very short time at midwinter was sufficient for the experimental trees to become damaged. This fact is of great interest, as it is generally assumed that the toxic effect is strongest in the summer, when abundant gas exchange takes place through the stomata of plants.

### 32. Fertilizer Dust

Only one month after the test trees were brought to the field, exceedingly fine, grey dust could be seen on the surface of their needles (fig. 3). Observations in the field revealed a clear correlation between the directions into which the dust spreads and the brown colour noted in previously healthy trees. The branches pointing towards Typpi Oy, particularly the needles of the branch tops, were generally the dustiest. Table 1 gives an indication of the amounts of dust recorded. The dust content was high in the trees near the site of Typpi Oy, but decreased sharply as the distance became greater.

A grey (because of the dust) layer of ice was also noted on the needles of the test trees on several occasions.

Microscopy revealed that most of the dust had accumulated in the stomatal grooves of the needles (fig. 4). Large amounts of dust were also found in the stomatal depressions. Near Typpi Oy approximately half of the stomatal depressions of green needles even were covered and filled with dust.

The dust accumulated in the November trees by the turn of the year proved to contain large amounts of fluorine  $(F^-)$ . The deposit washed away from the surface of the



Fig. 3. Pine shoots coloured grey with fertilizer dust. (On the right an unpolluted shoot for comparison).

needles contained as much as 2 % of fluorine. The total sulphur content of the dust deposit was 0.5—1 %. Apparently, however, most of the dust covering the needles is insoluble in water. Table 2 shows roughly the amounts of fluorine and sulphur that may dissolve in water from the dust.

In the case of table 2, it should be noted that the amounts of fluorine, sulphur and nitrogen naturally depend on the amount of water used and the effectiveness of washing; despite this, the values presented are probably more or less comparable with each other. The trees which had remained on the experimental field unprotected for a long time contained more of these elements than the trees which had been protected or had been exposed for only a short period, not to mention the samples obtained from further away (Kuivasjärvi).

The analyses of the dust (assays of potassium, phosphor and calcium were also made) show that its composition resembles relatively closely the compositions of the fertilizers produced at Typpi Oy. Thus the dust deposit on the needles can be recognized as fertilizer dust from the operations producing compound fertilizers. The pH measurements made support this notion: the dust is nearly neutral. It therefore should have no acid effect on the needles.

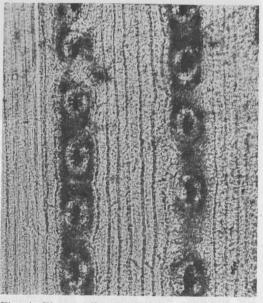


Fig. 4. Pine needle stomatal cavities filled with fertilizer dust. (The dust can be seen as black in the picture).

Table 1. The amount of dust on the shoot samples of the November test trees. Samples collected on Dec.17th 1969. The amount of dust = the deposit settled at the bottom of the washing basin, expressed as<br/>percents of the dry weight of the shoots.

	The amount of dust (%)
The youngest shoots on the side of the test trees facing Typpi Oy	6.0
Dider parts of the shoots on the side of the test trees facing Typpi Ov	51
The youngest shoots on the side opposite to Typpi Ov	3.8
older parts of the shoots on the side opposite to Typpi Oy	3.5

reus the fillering content of prop de-cone is the star	F mg/l	SO <sub>4</sub> mg/l	$\Sigma - N mg/l$
Unprotected November test trees:		al and indu	ental area:
<ul> <li>shoots over 1 year old with needles</li> <li>the youngest shoots with needles</li> </ul>	$\begin{array}{c} 14.7\\21.5\end{array}$	682.4 774.9	319.2 617.6
Fully protected November test trees:		role of si	one nators
<ul> <li>shoots over 1 year old with needles</li> <li>the youngest shoots with needles</li> </ul>	$\begin{array}{c} 4.4 \\ 6.5 \end{array}$	275.7 305.0	90.0 124.4
Februar test trees:		the effect of	ertilizir due
<ul> <li>shoots over 1 year old with needles</li> <li>the youngest shoots with needles</li> </ul>	$5.6\\3.0$	$267.6 \\ 231.8$	$200.3 \\ 156.9$
Unpolluted trees (the Kuivasjärvi area):		A state in the second	sines(200rd
<ul> <li>shoots over 1 year old with needles</li> <li>the youngest shoots with needles</li> </ul>	$\begin{array}{c} 0.0\\ 0.0\end{array}$	128.4 49.8	37.2 38.4

Table 2. The analysis on the washing water <sup>1</sup> of the pine shoots (after the deposit at the bottom has been removed). Samples collected on Febr. 19. 1970.

 $^{1}25-50$  g of shoots (dry weight) were washed with 1.5 l of water. The values of the table have been calculated per 100 g shoots (dry weight) and 1 l of washing water.

### **33. Analyses on Pine Needles**

The concentrations obtained are not guite as reliable, since the pollutants covering the surface of the needles are difficult to eliminate. Yet, the results are probably comparable at least where the rates of accumulation are considered.

At it is known, fluorine in particular tends to concentrate towards the tip of the leaf over time. In pine, however, this tendency is not very strong; the needle tips may contain 2—10 times as much fluorine as the base (cf. e.g. TRESHOW, 1970, p. 268). The analyses made on the needles of the November trees (table 3) show that the present material contains no essential difference between the tip and the base as regards fluorine or any other substance determined. This is natural, for the transpiration flow is very slight under winter conditions. Thus the polluting agents remain near the point where they entered the needle.

The analyses made (cf. e.g. tables 3 and 5, and figures 5 and 7) show that old needles — at least if they have been exposed to effective pollutants for a sufficiently long period — contain more toxic and other harmful substances and also nutrients (at least N and K were analyzed) than needles under 1 year old. The differences, however, are not great.

In order to find out what the situation was in the youngest needles, some quite recently grown needles collected on June the 22nd 1970 were analyzed for F and S. The results are presented in table 4. The results show that near Typpi Oy very young needles contained in smaller quantity fluorine than did older needles. Thus it seems probable that F is not, at least in large amounts, translocated from the older parts of the tree into the

Table 3. Analyses on the base and tip parts of the washed needles of the November test trees. The samples of needles collected on Jan 7. 1970.

and the second s	F ppm	S %	$\Sigma - N \%$	К %	Р%
Needles over 1 year old, base	222	0.31	3.02	1.4	0.54
» » » » tip	204	0.28	3.10	1.4	0.49
Needles of the youngest shoots, base	147	0.33	2.59	1.3	0.51
» » » » tip	219	0.28	2.67	1.1	0.46

	F p	pm	S	%
Polluted trees:	sample		Ha dettination	
<ul><li>Needles over 1 year old</li><li>young needles</li></ul>	$\begin{array}{c} 39.2 \\ 16.6 \end{array}$	$\begin{array}{c} 28.8\\ 6.2 \end{array}$	0.18 0.13	$\begin{array}{c} 0.12\\ 0.16\end{array}$
Unpolluted trees:		a contractorio		
<ul> <li>needles over 1 year old</li> <li>young needles</li> </ul>	7. 8.		0. 0.	

Table 4. Analyses on quite young and more than 1 year old washed pine needles. The samples were collected on June 22nd 1970 at two areas on the western side of Typpi Oy (polluted trees) and at one site on the Kuivasjärvi area (unpolluted trees).

growing new needles. As for sulphur, the values recorded near Typpi Oy were low in both younger and older needles.

Tables 3 and 4 present the results of some F, S and N analyses made on the pine

needles. Extremely high values for fluorine were obtained from the November trees only one or two months after they were brought to the experimental field. As can be seen from figure 5, the peak values were more

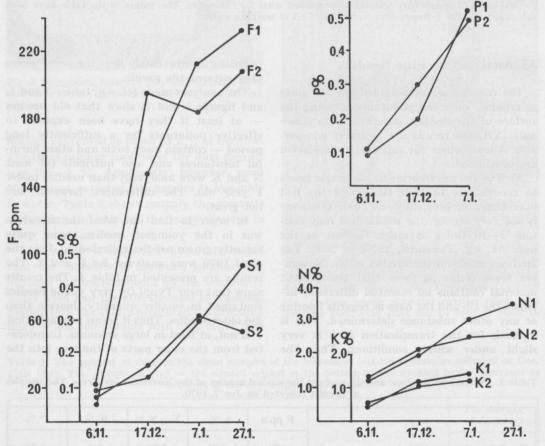


Fig. 5. Accumulation of fluorine, sulphur, phosphor, nitrogen and potassium in the needles of the test trees brought into the vicinity of Typpi Oy on Nov. 4th 1969 during the period Nov. 6th 1969 - Jan. 27th 1970. Fl = amount of fluorine in pine needles over one year old, F2 = amount of fluorine in needles less than one year old. Similarly S = sulphur, P = phosphor, N = nitrogen, and K = potassium.

than tenfold if compared with the initial values (control trees). In unindustrialized areas the fluorine content of pine needles is generally 1-10 ppm (e.g. GARBER, 1968, p. 43-45, Treshow, 1970, p. 271). Bossavy (1966) on the other hand, considers 25 ppm to be a nearly normal value for Pinus silvestris (cf. also ROBAK, 1969, p. 28). The values obtained from the analyses carried out by Typpi Oy on the unpolluted area further away fall into this order of magnitude. The highest F content recorded in green needles was 29 ppm (among the samples collected August 6th-15th 1969), but this tree was already damaged (TYPPI OY, Tutkimusosasto 1970, p. 3). The data concerning the tolerance of the pine are somewhat contradictory, but pines are generally regarded as relatively sensitive to fluorides (cf. e.g. THOMAS, 1964, p. 249-250, Rippel & Janovicová, 1968, WENTZEL, 1968, p. 367, DAINES, 1969, p. 438, TRESHOW, 1970, p. 292).

The peak fluorine values recorded for the November trees must be considered so high that they would result in the death of the needles even if coniferous trees were — as ROHMEDER & SCHÖNBORN (1968, p. 58) suggest — much more resistant in winter conditions than in the summer. As the fluorine which has penetrated into the needle remains in one place until metabolism is stimulated in the spring (as explained above), death apparently occurs then at the latest.

The initial values for sulphur were below 0.1 % (healthy November trees on Nov. 6th 1969, cf. fig. 5). By January, values 3-4 times higher were recorded on the experimental field. Pines are generally regarded as less resistant to sulphur oxide than deciduous trees (cf. e.g. THOMAS, 1964, p. 234, TRESHOW, 1970, p. 256, BJÖRKMAN, 1970 and the literature presented by him). The susceptibility of needles to the effect of sulphur depends not only on the SO<sub>4</sub> concentration and the duration of exposure, but crucially also on the stomatal conditions of the needles and the rate of metabolism. It is therefore impossible to give clear and unambiguous sulphur tolerance values for pine needles. According to KATZ (1952), healthy needles of coniferous trees normally contain 0.1 % of organic sulphur (per dry weight). Investigations by Typpi Oy revealed total sulphur contents of 0.1-0.2 % in the green

needles of the neighbourhood pines. LAAMA-NEN & LAHDES (1969, p. 41–43) noted that the sulphate concentration in the pine needles exceeded 0.5 % of sulphate in some sections of a mixed residential and industrial area.

As the investigations carried out in the neighbourhood of Typpi Oy are not yet sufficient, it is difficult to say anything definite about the possible role of sulphur in the injury of trees. It seems probable, however, that sulphur is less significant than fluorine, though it certainly has some effect as a contributary factor. The effect of fertilizer dust can also reduce the S-content of the polluted trees and help decrease the amount of damage (cf. STEFAN, 1969, BJÖRKMAN, 1970). The peak values recorded for total sulphur content (0.4-0.6%) are however sufficiently high to bring about the conditions under which damage is likely to occur.

The total nitrogen content in healthy needles of the test trees from the unfertilized area and the normal trees used as controls was normal, i.e. 1-1.5 % of dry weight (cf. e.g. AALTONEN, 1950, table 1 and 3, VIRO, 1950, p. 18-19 and 1955, p. 16-21). In the same way as F and S contents, nitrogen content also increased as a function of time in the test trees. The nitrogen content of the November trees had almost doubled by the latter of January (cf. fig. 5). According to the measurements made by Typpi Oy (Typpi Oy, Tutkimusosasto, 1970, p. 2-3), the mean nitrogen content of the needles in the healthy area was 1.1-1.2 %. Green pine needles near Typpi Oy generally contained a little over 1.5 % of nitrogen. According to TREsноw (1970, p. 224), tissues are damaged only when the leaves contain over 4 % of nitrogen.

Very little is known about the possible significance of nitrogen for oxidant toxins. The symptoms noted around Typpi Oy do not resemble the symptoms caused by e.g. ozone or the PAN compounds (cf. e.g. DARLEY, E. F., 1968). The symptoms caused by NO<sub>2</sub>, on the other hand, are similar to the SO<sub>2</sub> damage (cf. e.g. HAUT & STRATMANN, 1967, p. 54—56, TRESHOW, 1970, p. 359). As far as it is known, the concentration of NO<sub>2</sub> in the air may become relatively high on dark and foggy days. Yet its toxic effect is probably not of the same order of magnitude as that of SO<sub>2</sub>. The damage of trees around Typpi Oy is apparently not due to nitrogen, not at least to gaseous nitrogen alone. It is more likely that the high nitrogen contents are brought about by fertilizer dust, which contains both nitrate and ammonia.

It is generally known that higher than normal nitrogen concentrations in the growing base may sometimes result in a prolonged growing period in the autumn. In these cases the perennial parts may not always have enough time to adapt to winter conditions. As a result, the pines may suffer from a shortage of water due to excessive transpiration — thus especially in the early spring. As will be explained later, both needle structures that are more mesomorphic than normal and spring-time water insufficiency are apparent in the trees around Typpi Oy. The strong effect of nitrogen fertilizers may be the reason for this. The rapid destruction of the test trees, however, shows that the effect of fertilizers is not the only, nor the primary reason, even near Typpi Oy.

The potassium and phosphor contents of the needles are similarly higher than normal (fig. 5). It is apparent, however, that neither potassium nor phosphor have any actual toxic effect on the needles, particularly since the balance between the different nutrients is not badly disturbed. At the beginning of January the phosphor and potassium contents of the needles of the November trees were twice as high as the values reported by AALTONEN (1950, table 6) and VIRO (1950, table 4).

Since the spring of 1966 the content of fluorine, sulphur and nitrogen in the needles of pine surrounding Typpi Oy has been followed at 12 observation points. Figure 6 shows graphically the variations in fluorine, sulphur and nitrogen content at three ob-

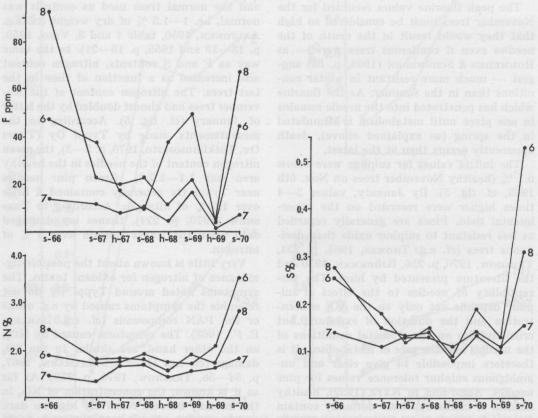


Fig. 6. Variation of fluorine, nitrogen and sulphur contents in pine needles less than two years old in the three observation points near Typpi Oy during the years 1966 - 1970. No 6 = 0.7 km NW of the compound fertilizer plant, margin of a forest, 7 = 1.5 km NW of the compound fertilizer plant, in the middle of a forest, and 8 = 0.8 km NW of the compound fertilizer plant, margin of a forest. s - 66 = spring 1966, h - 67 = late summer 1967 (and so on).

100

ogive of appoint of finorise, while the woody par	F ppm	S %	$\Sigma - N \%$
Unprotected November test trees:	1996-1976	in your	
	202	0.49	3.31
<ul> <li>needles over 1 year old</li> <li>needles of the youngest shoots</li> </ul>	165	0.45	2.70
Fully protected November test trees:	Patris Mas (. 1941 - Nas (.		gewa tadiy
- needles over 1 year old	24	0.13	1.53
- needles of the youngest shoots	23	0.11	1.43
Unpolluted trees (the Kuivasjärvi area):	out presen		points 6 a
– needles over 1 year old	0	0.09	1.25
- needles of the youngest shoots	0	0.09	1.29

Table 5. Analyses on washed pine needles. Samples collected on Febr. 19th 1970.

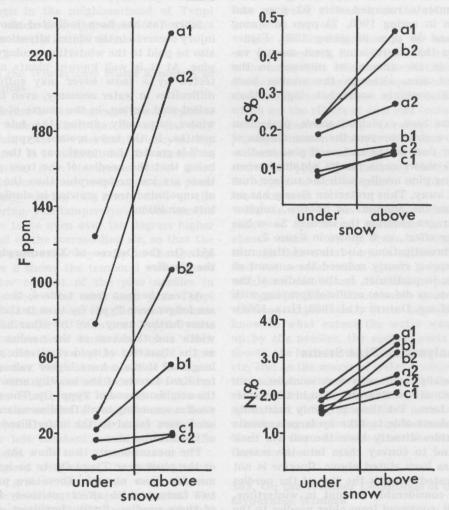


Fig. 7. Fluorine, sulphur and nitrogen contents of pine needles above and under the snow in winter 1970. al = needles over one year old of the test trees brought near Typpi Oy on Nov. 4th 1969, a2 = needles less than one year old of the same trees, bl and b2 = needles of the pines brought near Typpi Oy on Febr. 4th 1970, cf. al and a2, cl and c2 = trees growing naturally near Typpi Oy (in the W direction), cf. al and a2. The needle samples of the November trees were collected Jan. 27th 1970, the others May 5th - 12 th 1970.

servation points in the damaged area. Especially at point 6, the variation in fluorine content correlates closely with the degree of damage noted during different years (spring 1966, 1969 and 1970). The point in question is situated in the very middle of the damaged area, as is also point 8. Point 7 is situated further away in the forest, and it is therefore natural that the fluorine and nitrogen contents are smaller there. Sulphur content similarly correlates with the degree of damage at points 6 and 8. Without presenting the results obtained at the 12 different points, it may be concluded that sulphur content only once exceeded 0.50 %. The highest fluorine contents recorded were 93 ppm and 59 ppm in spring 1966, 75 ppm in spring 1968, and 50 ppm in spring 1969. Figure 6 shows that there is no great annual variation in the amount of nitrogen in the damaged area, although the needles have nitrogen contents somewhat higher than normal.

As has been explained above, protection and snow cover prevent the accumulation of fertilizer dust on the surface of pine needles. Table 5 shows some results obtained when analyzing pine needles with the surface dust washed away. Thus protection clearly has an effect on the amounts of fluorine, sulphur and nitrogen entering the needles. Snow has a similar effect, as is shown in figure 7.

The investigations also showed that rain in the spring clearly reduced the amount of fluorine, in particular, in the needles of the test trees, as did also artificial spraying with water (cf. e.g. DAINES et al. 1952, HILL, 1968).

### 34. Analyses on Pine Stems

Especially acid soils may contain quite large amounts of fluorides even in the watersoluble form. Yet there probably exist only a few plants able to take up large amounts of fluorides directly from the soil via their roots and to convey them into the leaves.

As has been stated above, fluorine is not translocated towards the tips of the needles in any considerable amount in wintertime, nor is it conveyed from older needles to the new growing needles. Without going into the detailed results of the analyses made on the stems of the trees, it may be briefly concluded that near the ground the surface (bark) part of the stem in the test trees contains a certain amount of fluorine, while the woody part contains very little. The tops of the trees have very little fluorine in both the bark and the woody parts. Thus fluorine has not reached the top of the trees at least in the woody part. It therefore seems certain that the fluorine contained in the needles comes directly from the substances covering their surface.

# 35. On the Wintertime Ecology of the Pine

Since (as has been indicated above) the injury occurred in the winter, attention should also be paid to the wintertime ecology of the pine. At it is well known, plants not protected by a snow cover may suffer from difficulties in water economy, even from socalled cold drying, in the course of the long winter, especially during the late winter months. In the trees around Typpi Oy this peril is greater than usual, one of the reasons being that the needles of the trees growing there are less xeromorphic than the needles of unpolluted trees growing in similar moisture conditions.

# 351. On the Degree of Xeromorphims of the Needles

As can be seen from table 6, the needles are longer near Typpi Oy than in the control areas further away. On the other hand, the width and thickness of the needles as well as the diameter of epidermal cells and the length of stomata have higher values in the fertilized forests of the healthy area than in the neighbourhood of Typpi Oy. The smallest needles according to all the dimensions measured were found in the unfertilized forests of the healthy area.

The measurements thus show the needles of the pines near Typpi Oy to be less xeromorphic than normal. There are probably two factors which affect positively the size of these needles: firstly, fertilizers, and secondly, the fact that part of the buds are destroyed and the remaining ones consequently grow into needles bigger than normal.

Area	Average length (mm)	Average width (mm)	Average thickness (mm)	Average diam- eter of epider- mal cells (mm)	Average length of stomata (mm)
Neighbourhood of Typpi Oy	34.93	1.564 (+ 0.039)	0.732	0.0152	0.0566
Sanginjoki, fertilized area	$(\pm 2.047) \\ 33.60 \\ (\pm 2.785)$	$(\pm 0.039)$ 1.580 $(\pm 0.034)$	$(\pm 0.016) \\ 0.753 \\ (\pm 0.013)$	$ \begin{array}{c c} (\pm 0.003) \\ 0.0156 \\ (\pm 0.0006) \end{array} $	$(\pm 0.0023)$ 0.0587 $(\pm 0.0010)$
» , unfertilized area	$(\pm 1.568)$ $(\pm 1.568)$	$(\pm 0.037)$ $(\pm 0.037)$	$(\pm 0.026)$ $(\pm 0.026)$	$\begin{array}{c c} (\pm 0.0000) \\ 0.0141 \\ (\pm 0.0002) \end{array}$	$(\pm 0.0010)$ 0.0564 $(\pm 0.0015)$

Table 6. Measurements made on healthy needles one growing season (1969) old in the neighbourhood of Typpi Oy and further away in fertilized and unfertilized areas (Sanginjoki).

It should be noted that the samples of needles were obtained from apparently healthy trees even in the neighbourhood of Typpi Oy, in which case the effect of fertilizers seems to be the crucial factor.

# 352. On the Wintertime Water Economy of the Needles

A decrease in the degree of xeromorphism has the natural effect that the needles may suffer from crises of water insufficiency during the winter. It is difficult for the needles to obtain water from the frozen soil or stem, although continuous transpiration takes place particularly during the sunny days of the early spring. The temperature of the needles may rise to be even over ten degrees higher than that of the surrounding air, so that the stomata open relatively early in the spring.

Figure 8 shows the temporal variation of the water content of the pine needles in branches above the snow both near Typpi Oy and farther away in the controll area (Oulu, Kuivasjärvi). Even near Typpi Oy the measurements were made from trees which were still relatively healthy-looking, i.e. had green needles. The water content of the needles above the snow naturally decreases throughout the winter until the end of March, whereas the needles covered by snow both near Typpi Oy and farther away remain in more or less constant conditions until the snow melts.

The green needles of the polluted area show far greater variation in water content than the trees in the unpolluted area. The situation in the polluted area became highly critical in March, when the water content of the old needles became extremely low. During the first half of April the situation returned almost to normal. A period of thaw occurred at that time, and the needles could take up water at least from the stem if not even from the soil.

Since the investigations so far made are not yet adequate, it is difficult to say what is the immediate effect of the fertilizer dust discharged from the process operations on the stomata and temperature of the needles, and which are the effects of dust and toxic agents on transpiration (cf. e.g. NAVARA, 1968). The principal reason for the disorders in the wintertime water economy of the trees surrounding Typpi Oy is probably the fact that the degree of xeromorphism of the needles has decreased.

HAVAS (1971) has suggested that some plants of the boreal coniferous zone may absorb water directly from melting snow through their aerial parts in winter. We carried out several water uptake experiments with pine shoots, one of which is presented in figure 9. Since shoots were used in the watering experiments, it is impossible to know to what extent the water was taken up by the needles, the surface parts of the shoots, the buds, or the scales of short shoots, etc. But as the assays of water content show, the water content of the needles also increased clearly during the experiment. It therefore seems that during humid thaw periods in the winter, pine needles take up water not only from the stem but also from the water on their surface.

### 353. On the Weather Conditions of the Unfavourable Winters

On the basis of what has been presented above, the most unfavourable winters seem to be the ones with long sunny periods of

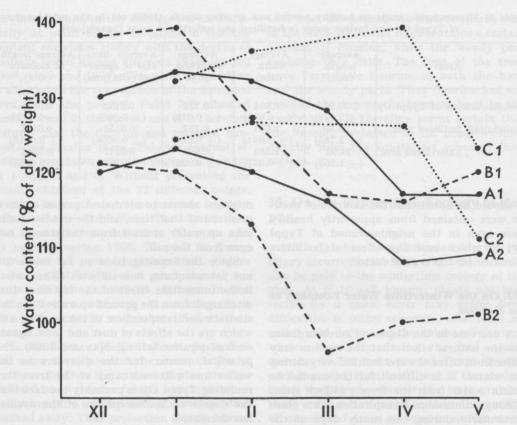


Fig. 8. Variation of water content in pine needles during the winter 1969-1970, the monthly mean values. Al = needles less than one year old in the unpolluted area (Kuivasjärvi), A2 = needles over one year old of the same trees, B1 and B2 = green needles of the trees near Typpi Oy, cf. A1 and A2, C1 and C2 = needles near Typpi Oy covered by snow during the winter, cf. A1 and A2.

cold and drought, but no rainfall. Under such conditions the pines around Typpi Oy may suffer from quite severe crises in their water economy. Meterological observations made at Oulu show that all the years during which damage occurred (winters 1965-66, 1968-69 and 1969-70) were unusual in that they had long periods of abnormally cold weather. The first of these winters was particularly difficult, as the weather was much colder than usual from November up till April. Similarly, during the other two winters the temperature remained below the normal, values for 3-4 months on end. It should be noted, however, that the winter 1962-63 was cold for a considerable length of time, but no great damage occurred then.

It seems obvious that the winter weather conditions have played a role in the damage of pines around Typpi Oy. Yet it is very difficult to estimate the effect of weather conditions on the damage caused by toxic agents of fertilizer dust.

### 354. Assimilation of the Needles

In connection with different fumigation treatments, plants have often also been subjected to experiment on respiration and photosynthesis (see e.g. THOMAS, 1964, p. 236—237 and 254—259, TRESHOW, 1970, p. 258—260, 284—287, 315—316 and 325— 327). Changes have been noted in the physiological activity of plants even when no visible damage has yet appeared. Generally fumigation with either fluorine or sulphur results in depressed assimilation (if the toxic effect is sufficiently great), but the assimilation values later approach the normal level

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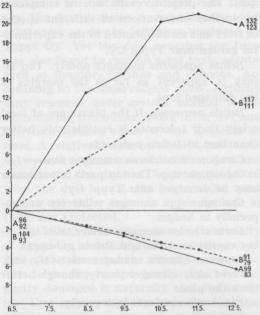


Fig. 9. Watering and transpiration experiment of pine shoots during May 6th — 13th 1970. Changes of weight (%) as compared with the initial weight (marked as 0) of the shoots. Each point represents the mean of two shoots and two measurements carried out during one day. A = shoots of the unpolluted area (Kuivasjärvi), B = shoots near Typpi Oy (with green needles). The figures express the water content (% of dry weight) of the shoots at the beginning and at the end of the experiment (the upper figure = part of shoots less tan one year old, the lower figure = older parts of shoots).

again, provided the fumigation is discontinued in time. Recovery is generally more rapid after sulphur treatment than after exposure to fluorine, for the fluorine must be conveyed to the tips and margins of the leaves before the situation returns to the normal. Several studies have been made on the inhibiting effect of severe fluorine poisoning on the respiratory enzymes. There are also observations on how fumigation with fluorine mighs even stimulate respiration before visible damage appears.

The present experiments on photosynthesis were mainly designed to find out whether the needles collected from around Typpi Oy show physiological changes before any visible damage occurs. The experiments were therefore made on seemingly healthy-looking shoots. At the same time an attempt was made to follow the chlorophyll content of the needles. Because of the abundant material, only some typical cases can be presented here (fig. 10). The results show a relatively great dispersion, which is probably due to the fact that different trees with possibly different physiological conditions were used in each experiment. Moreover, the situation naturally changes over time. Despite this, the results show that the net assimilation of the trees growing in the polluted area was on average weaker than that of the trees in the unpolluted area. Artificial treatment with fertilizer dust also clearly reduces the net assimilation of initially healthy needles.

It is difficult to estimate to what extent the changes noted in assimilation are due to

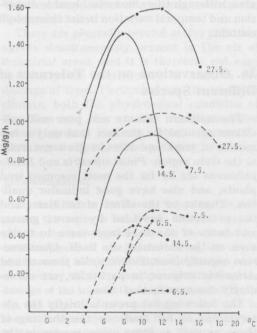


Fig. 10. Net assimilation curves of green pine shoots in the spring 1970. Continuous line = assimilation of unpolluted trees, dotted line = assimilation of polluted trees. The dates of the experiments are given: 6.5. upper curve = pine shoots under the snow near Typpi Oy, lower curve = pine shoots above the snow near Typpi Oy. 7.5. = pine shoots collected from an unpolluted area; part of them (dotted line!) have been treated with the water used for rinsing the test shoots containing fertilizer dust. 14.5. =the March test trees on the experimental field (dotted line!) and on an unpolluted area. 27.5 = shoots collected near the Oulanka Biological Station at Kuusamo; part of them (dotted line!) have been treated with the water used for rinsing the polluted shoots. Experimental conditions: light 20 000 lux, flow of air 52 l/h.

the blocking of the stomata by dust. As has already been mentioned, the dust is particularly likely to enter the stomatal depressions. Respiration curves (not presented in Fig. 10), however, seem to suggest that the stomata are not completely prevented from carrying out gas exchange, for respiration is lively both in the trees of the polluted area and in pines treated artificially with water containing fertilizer dust.

The experiments thus support the notion that the trees of the damaged area show changes in their physiological condition and activity even before visible changes appear.

The chlorophyll content revealed no clear difference between the green pine needles of the damaged area and those of the unpolluted area, although there was considerable dispersion and temporal variation in the chlorophyll content.

# 36. Observations on the Tolerance of Different Species

The northern climate and poor soil conditions account for the fact that only a few species of trees and schrubs grow naturally in the Oulu region. *Pinus silvestris* and *Betula pubescens* are by far the most common seed plants, and also have good indicator qualities. Thanks to the effect of fertilizers and the protection provided by snow, grasses and herbs of the field layer seem to thrive even on the industrial site itself. *Chamaenerion angustifolium, Deschampsia flexuosa* and *Artemisia vulgaris*, in particular, are exceedingly common in that area.

The following list presents briefly the observations so far made on the thriving of those as well as some other species in the neighbourhood of Typpi Oy. The list includes several species used as garden plants in this area. The present results will be completed by further observations of different species of trees and shrubs planted in the experimental garden near Typpi Oy.

Betula pubescens manages poorly. Top injury is apparent as far as the margins of the damaged area.

Betula verrucosa. If the plants are of local origin their tolerance is considerably better than that of Betula pubescens.

Caragana arborescens manages well.

*Cotoneaster* spp. The top parts in particular may be damaged near Typpi Oy.

*Crataegus* spp. manages relatively well especially in hedges.

Larix sibirica manages better than the other coniferous trees and *Betula pubescens*.

Lonicera tatarica manages relatively well. Picea abies manages poorly, though better than the pine.

Pinus silvestris manages poorly.

Populus tremula manages relatively poorly. Prunus padus manages moderately well.

Ribes nigrum manages relatively poorly.

*Ribes spicatum* manages better than *R. nigrum.* 

*Rosa rugosa* is the most thriving rose species, but this species is mostly covered by snow in the winter.

Salix spp. manages moderately well.

Sorbus aucuparia manages best of all the trees growing naturally in the area, though in the most damaged area the tops may be injured or even die.

Syringa josikaea manages well.

Syringa vulgaris manages moderately well.

It can be concluded that southern tree and shrub species generally manage more poorly when exposed to the effects of Typpi Oy than the species which even otherwise grow well, either planted or naturally, in the Oulu region.

### 4. SUMMARY

The symptoms of the pine — and also of the birch, though the results are not presented here — show that the damage is initiated in winter conditions. Although the visible damage does not appear until the spring in natural conditions, laboratory experiments show that it has already occured in the winter. Most of the literature on this topic contains information of toxic damage brought about by different gases in the summer. This phenomenon can also be seen at Oulu, where several species near Typpi Oy develop further injuries in the summer.

Large amounts of fine fertilizer dust discharged from the compound fertilizer process during the winter accumulated in the needles of the test trees brought near Typpi Ov. The dust contains more or less the same substances as do the fertilizers produced by Typpi Oy. Yet the dust covering the trees was also found to contain some toxic agents. such as fluorine, sulphur and chlorine. Accordning to the observations made, fertilizer dust descends under certain weather conditions during the winter, being often wet and therefore sticking easily to the branches of trees. A relatively clear correlation was found between the occurrence of fertilizer dust and damage to the trees. The trees protected from the dust — e.g. the parts under snow did not suffer from as much damage as those not protected.

The fallout of fertilizer dust in the vicinity of Typpi Oy has clearly increased the growth rate of the pines and the size of their needles. The large size of the needles in the trees partly damaged is naturally also due to the fact that only part of the buds develop. One results from this is that the needles become more susceptible to possible drying during the winter. It was also noted that the trees in the polluted area suffer from occasional crises in their water economy especially during the late winter. On the other hand, the metabolism, of the needles is disturbed by the physical effects of the deposit of fertilizer dust covering the surface. The fine dust blocks the stomatal grooves and depressions, thus disturbing the gas exchange in the spring and even in the summer, unless rain has access to the needles and washes away the dust. Yet when we are looking for the reason of the damage, neither the fertilizing effect, the disorders in water economy, nor the blocking of the stomata can be the major injurious factor; the crucial role is apparently played by the toxic effect.

The amount of fluoride, in particular, may be so great in the trees surrounding Typpi Oy that the needles are destroyed. During one or two midwinter months the needles of the test trees accumulated fluorine about 10 times as much as they normally tolerate. It also seems that under winter conditions F is not translocated to the tip part of the needle, and consequently almost the whole needle is damaged. The trees did not take up any noticeable amounts of fluorides from the soil, nor were fluorides conveyed from older needles to the new ones. When the needles are suffering from a shortage of water in the late winter, the toxic water solution on their surface may penetrate into them more easily than at times when they are not suffering from water deficiency. The sulphur contents of the needles were not so alarming, though in some cases even they had reached the critical level. Unfortunately, the needles of the trees in this area have been inadequately analyzed for e.g. chlorine. Chlorine may, after all, play a role in the damage of trees.

There are generally several active polluting agents simultaneously present in the air of industrial areas, and it is therefore not easy to specify the contribution of each to the damage of trees. Particularly in the northern climate, both the physiological condition of the trees and the weather conditions show several special features which apparently have some effect in the polluting situation.

#### Acknowledgements

My best thanks are due to Typpi Oy for suggesting this topic to me, for the financial support given to the investigations, and for the laboratory, facilities placed at my disposal during the course of this work. Maist. P. Meriläinen, who has acted as secretary to the committee investigating the damage of the trees in the neighbourhood of Typpi Oy, has helped me in many ways. I also wish to thank the other members of the committee, Prof. N. Söyrinki, and Forest Officer E. Estama, who have all been most helpful for my work. LuK Satu Huttunen, LuK Elsa Kangas and yo. Markku Henttonen acted as my research assistans. My warm thanks are due to them all. I also wish to thank docent A. Laamanen, who kindly read through the manuscript and made many valuable comments.

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### MÄNNYN VAURIOISTA ERÄÄN POHJOIS-SUOMEN KEMIALLISEN TEHTAAN LÄHEISYYDESSÄ

#### SELOSTE:

Keväällä 1966 ilmeni Oulussa Typpitehtaan lähistöllä ensimmäisen kerran suurehkoja puustovaurioita varsinkin männyissä (*Pinus silvestris*) ja koivuissa (*Betula pubescens*). Vuonna 1970 käsitti vaurioitunut alue noin 250 ha. Tässä tutkimuksessa pohditaan lähinnä mäntyjen vaurioitumisen syitä. Havainnot osoittavat, että männyn neulasten tuhoutuminen saa yleensä alkunsa talviolosuhteissa, vaikkakin näkyyät vauriot ilmenevät vasta keväällä. Yleensähän on muualla tehdyissä tutkimuksissa osoitettu, että esim. myrkylliset kaasut vaurioittavat puita tehokkaimmin kesäolosuhteissa.

Typpitehtaan lähistöllä todettiin sinne tuotujen terveiden mäntyjen neulasten pinnalle keräytyvän talvella lannoitepölyä. Analyysit osoittavat, että lannoitepölyyn on absorboitunut myöskin eräitä kasveille myrkillisiä aineita, kuten fluoria, rikkiä ja klooria. Varsinkin tietyissä sääolosuhteissa laskeutui männyn neulasten pintaan runsaasti tällaista märkää ja myrkyllistä pölyä. Tarkasteltaessa lannoitepölyn esiintymistä tehtaan ympäristössä ja puiden vaurioitumista todettiin näiden välillä vallitsevan jokseenkin selvän korrelaation. Esim. sellaiset puut, jotka ovat rakennusten suojassa tai lumipeitteen alla talvella, säästyivät pahimmilta vaurioilta. Sensijaan vaurioita oli eniten yksin kasvavissa puissa samoinkuin esim. metsän laitamilla.

Toisaalta voitiin todeta lannoitepölyn selvästi lisänneen puiden kasvua tehtaan lähistöllä. Tämä ilmenee mm. männyn neulasten koon mittauksista. Neulaset ovat tehtaan lähistön puissa vähemmän kseromorfisia kuin yleensä vastaavanlaisilla kasvupaikoilla kauempana. Osittain tähän vaikuttaa myös tietenkin se, että kun osa neulasista kuolee, kasvavat jäljellejääneet tavallista suuremmiksi. Neulasten normaalia mesomorfisempi rakenne johtaa siihen, että puiden vesitaloudessa voi tulla kriisejä varsinkin myöhään kevättalvella, jolloin haihduttaminen tällaisista neulasista on normaalia suurempaa. Näin varsinkin silloin, kun sääolosuhteet ovat pitkään olleet epäsuotuisia (pitkiä pakkasjaksoja). Toisaalta myöskin neulasten metabolia kärsii jonkin verran ilmeisesti siitä, että lannoitepöly tukkii mm. ilmaraot.

Lannoitevaikutus tai siitä johtuvat mahdolliset kriisit puiden vesitaloudessa tai ilmarakojen tukkeutuminen eivät kuitenkaan yksin riitä ilmeisestikään selittämään Typpitehtaan ympäristön vaurioita. Tärkeä osuus on myöskin em. myrkyillä.

Varsinkin fluorin määrä voi olla tehtaan ympäristön puissa niin suuri, että se yksin riittää tappamaan neulaset. Yhden tai kahden talvikuukauden aikana keräytyi koepuiden neulasiin fluoria noin kymmenen kertaa enemmän, kuin mitä ne normaalisti sisältävät. Talviolosuhteissa fluori ei näytä siirtyvän neulasten kärkiin, mistä seurauksena on yleensä koko neulasen tuhoutuminen. Männyt eivät ota fluoria sanottavasti maasta, eikä sitä myöskään paljonkaan siirry vanhoista neulasista uusiin. Näyttää siis todennäköiseltä, että fluori kulkeutuu neulasiin suoraa niiden pinnalta ja ilmeisesti parhaiten silloin, kun puut kevättalvella kärsivät veden puutetta, jolloin myrkyllistä vesiliuosta pääsee neulasiin. Neulasanalyysit osoittivat, että rikkipitoisuus ei ole yhtä hälyttävä, vaikka eräissä tapauksissa sekin saavutti kriittisen tason. On mahdollista, että eräät myrkylliset aineet, kuten mm. kloori, jota toistaiseksi ei ole riittävästi analysoitu, ovat myös-

Tutkimus osoittaa sen, että teollisuusalueella ilmenee samanaikaisesti useita eri tekijöitä, jotka tavalla tai toisella toisiinsa kytkeytyneenä aiheuttavat puiden vaurioitumista. Eri tekijöitä on vaikea ja mahdotontakin pitää täysin erillään toisistaan ja arvioida niiden kunkin osuutta vaurioihin. Todennäköiseltä kuitenkin näyttää, että pohjoisen havumetsävyöhykkeen alueella on ilman saastumisen vaikutuksia kasveihin selvitettäessä otettava huomioon myöskin sellaisia ekologisia tekijöitä, jotka ovat vähemmän merkityksellisiä etelämpänä.

kin osaltaan vaikuttamassa puiden tuhoutumiseen.

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MÄNNYN VAURIGISTA ERAAN POHJOIS-SUOMEN KEMIALLISEI TERTAAN LÄHEISYYDESSÄ

Kevällilä 1966 Inner Oshiosa Typpinidaan jähitoila andunakken korren saurohitija pisutovan toila viminida mänoylon (Pinus sidenpio) ja kok oilesa (Jotale puterozo), Vuenda 1970 hänitä yrintoiliinut aine mois 250 ha. Tääsä jutkintokense ainditaen idiuma mäntylen rennollinnisen veitä Tyvolloisi osoilta yri, elsi tukunysiomiatteritehon milinen on yteensk aikuma lehvoloimitteisen, viite milinen on yteensk aikuma lehvoloimitteisen, viite

Morradala, en somalla tracpisat unklentitalade bonbeta, etta esto, regenetikes, sancid samiatite ret oute telentitationist beskelasobietsa.

Pypalishtaan ühistolle anleittin anne usobiyen terviden mäntyjer metetter prinale kerkviralis, tatonite karastepillyk. Aneiryst miditanst, että hansitepiisyö on attorteritanni myöskin että kasvelle hyyskillisiä aineita, keren fuerta, näsön y ja aluorta, verseken tirtyskä sokolasultalasa ker

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HAVAS, PAAVO 0.D.C. 425.2	HAVAS, PAAVO 0.D.C. 425.2
1971. Injury to Pines in the Vicinity of a Chemical Processing Plant in Northern Finland. – ACTA FORESTALIA FENNICA 121. 21 p. Helsinki.	1971. Injury to Pines in the Vicinity of a Chemical Processing Plant in Northern Finland. – ACTA FORESTALIA FENNICA 121. 21 p. Helsinki.
The present investigation shows that injury to pines ( <i>Pinus silvestris</i> ) in the boreal coniferous zone ( $65 ^{\circ}N$ ) occurs in winter conditions in the vicinity of a fertilizer processing plant, unless they are covered by snow. This kind of injury has multiple causes. Firstly, fertilizer dust discharged from the process operations may reduce the degree of xeromorphism of the needles,	The present investigation shows that injury to pines ( <i>Pinus silvestris</i> ) in the boreal coniferous zone (65 °N) occurs in winter conditions in the vicinity of a fertilizer processing plant, unless they are covered by snow. This kind of injury has multiple causes. Firstly, fertilizer dust discharged from the process operations may reduce the degree of xeromorphism of the needles,
which further results in disorders of the water economy. Secondly, along with the wet fertilizer dust the needles absorb toxic substances, especially, fluorides and certain sulphur compounds. The amounts of fluorides, in particular, are large enough to bring about damage. The combined effect of these factors causes trees to die during winters with long periods of intense cold. It seems, therefore, that in the northern conditions pollution may have effects not observable in more southern regions.	which further results in disorders of the water economy. Secondly, along with the wet fertilizer dust the needles absorb toxic substances, especially, fluorides and certain sulphur compounds. The amounts of fluorides, in particular, are large enough to bring about damage. The combined effect of these factors causes trees to die during winters with long periods of intense cold. It seems, therefore, that in the northern conditions pollution may have effects not observable in more southern regions.
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19/1. Injury to Fines in the vicinity of a Chemical Frocessing Plant in Northern Finland, – ACTA FORESTALIA FENNICA 121. 21 p. Helsinki.	Plant in Northern Finland. – ACTA FORESTALIA FENNICA 121. 21 p. Helsinki.
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