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The bark of Scots pine (*Pinus sylvestris* L.) as a biological indicator of atmospheric air pollution

MARIA ŚWIEBODA AND ANDRZEJ KALEMBA

Research Centre for the Protection of Nature and Natural Resources,
Polish Academy of Sciences, Lubicz 46, 31-512 Kraków.

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Abstract

In dead bark layer of Scots pines (*Pinus sylvestris* L.) exposed to the emission of aluminium works and the electric power plant "Skawina" as well as of chemical plants in Cracow, pH was determined and the content of F, S, Mg, Na, P, K, and Ca. The F content in the dead pine bark varied from 0.018 to 0.386 mg/g, and that of sulphur from 1.00 to 3.83 mg/g. A significant correlation was demonstrated between pH, and fluorine and calcium content in the bark. On the other hand, sulphur content was dependent on the amount of Na, K and Ca. For demonstrating contamination of the surface bark layer a test was applied consisting in evaluation of the germinating and development capacity of seeds. Fluorine content in pine dead bark layer reflects the F concentration in atmospheric air, it may, therefore, serve as indicator of the degree of air pollution with fluorine compounds.

INTRODUCTION

Tree bark has recently been used as a biological indicator of atmospheric air pollution. Such investigations were started in Sweden (Skye, 1968; Staxäng, 1969) and continued by other authors (Grodzińska, 1971, 1977; Lötschert, Köhm, 1973, 1976; Zdanowska, 1976). Generally deciduous trees were chosen for these studies, with a thick cracked bark, such as *Quercus robur*, *Fraxinus excelsior*, *Tilia cordata*, *Alnus incana*, and among conifers — *Pinus sylvestris*.

In the present investigations on the influence of emission from the aluminium works "Skawina" on the vegetation, chemical analysis was performed of dead bark layer from Scots pines persisting still on several localities at various distances from the industrial plants. Beside the pH, the content of elements characteristic for the emission of the aluminium works and electric power plant "Skawina" and of chemical plants situated in the southern part of the city of Cracow was determined.

MATERIAL AND METHODS

Dead bark layer (cracked) was collected from the trunks of living pine specimens on 13 localities situated within a radius of 12 km from the aluminium works and electric power plant "Skawina" (Fig. 1). Additionally bark samples were collected in the Ojców National Park (30 km to the North) and on a second control site 30 km distant in south-eastern direction from the industrial plants.

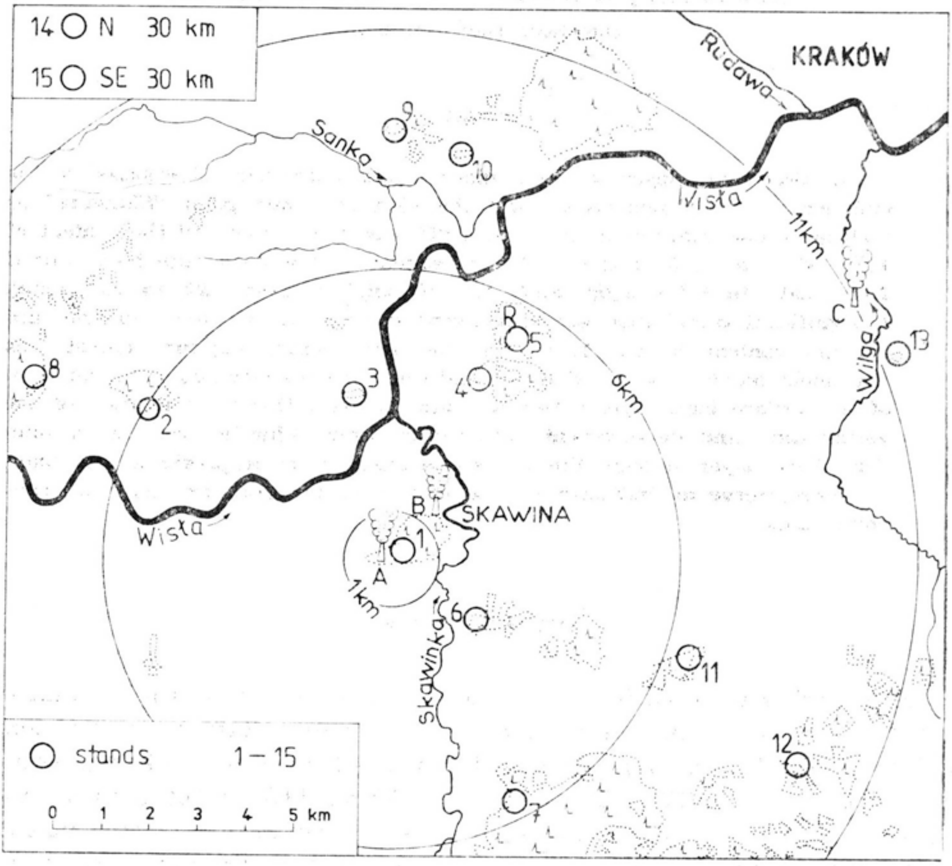

Fig. 1. Distribution of *Pinus sylvestris* L. localities on the area investigated
A — aluminium works "Skawina", B —electric power plant "Skawina", C — soda factory, R — floristic reserve "Skołczanka"

At each site the external bark layer was cut on the trunks from 10-15 pines aged about 80 years at a height of 1.5 m from the ground. The material was dried and ground to fine powder, then, after mixing, samples were taken for analysis. In these samples the following determinations were performed;

a) pH in H_2O — potentiometrically,

b) total fluorine — colorimetrically by Belcher's method with an alizarin complexing agent and lanthanum nitrate, photometry was run at wavelength 620 μm,

c) total sulphur, magnesium, sodium, phosphorus, potassium and calcium were determined by the methods described by Nowosielski (1974).

The contamination level of pine bark by toxic substances from industrial emission was determined in a biological test consisting in evaluation of the seed germination capacity (Świeboda, 1976). For this purpose seeds of radish (*Raphanus sativus* L.) highly sensitive to fluorine (Navara et al., 1966) were taken. The seeds were germinated on Petri dishes. As substrate served finely comminuted dead bark imbided with distilled water. On each dish 25 seeds were sown in 4 replications for each site. The seeds germinated under daylight and at room temperature. The course of germination was recorded after 24, 48 and 96 h.

The relation between the analysed elements and bark pH as well as their influence on radish seed germination were expressed as the linear correlation coefficient.

CHARACTERISTIC OF *PINUS SYLVESTRIS* L. LOCALITIES

The Scots pine localities on the territory examined are exposed to air pollution of various intensity, therefore, on the basis of changes in certain morphological features of the trees, the latter could be classified to 4 zones denoted by the symbols A, B, C, and D.

In the zone A situated closest to the aluminium works "Skawina" there is one site with pines. The specimens growing here show poor viability as indicated by the clear crowns and intensive dying back of branches. The trunks of the trees are covered with a dark dust layer.

In the zone B reaching to a distance of 6 km from the metallurgic works the negative influence of the industrial emission on *Pinus sylvestris* appears in the deformation of the tree habitus, drying up of the tree tops and earlier needle shedding. The growth of the pines in this zone is inhibited, thus the crowns take on an umbrella shape.

To the zone C the sites were classified with pines growing at the edge of the territory under study. The degree of damage to the pines in this region depends on the situation of the given locality in relation to the source of air-polluting emission. Most exposed to the noxious influences were the pines growing in the Vistula valley, since the strong southern winds carry the gaseous emissions from the industrial plants. When the noxious substances reach with the air the natural barrier formed by the Bielany hills, their concentration increases (Świeboda, Kalemba, 1978). Therefore, the degree of damage to the pines on localities 9 and 10 is close to that observed in zone B. On the other hand, localities 8, 11 and 12 lie beyond the zone of the most frequent winds. Scots pine on these sites is healthier since the air pollution concentration is much lower. As compared with trees on the control sites, however, the annual increments of the shoots are smaller and the branches possess only a two-year set of needles. Some scarce lichens appear on their bark.

The small group of highly damaged pines growing close to the chemical plants in Cracow was classified to zone D. The number of pine specimens on this locality diminishes drastically from year to year, this being also connected with the change in the chemical properties of the soil. The trunks are not covered with lichen, only in the crevices of the bark a green alga appears, *Pleurococcus viridis*.

Symbol K denotes two control sites. On both these localities the pine exhibits no noticeable developmental disturbances.

RESULTS

1. pH and content of the elements analysed in dead bark

The pH of the analysed pine dead bark samples varied from 2.9 to 4.1 (Table 1), the lowest value referring to the control site situated at a distance of 30 km south-east of the aluminium works "Skawina". The highest pH was found in dead pine bark from sites close to the industrial plants.

Fluorine content in the pine bark reached 0.386 mg/g in locality 1 (Table 1), most exposed to fluorine emission. On the other hand, the lowest fluorine amounts were noted in the bark from the two control sites in the Ojców National Park in the Wyspowy Beskid Mountains.

Total sulphur in the bark samples varied from 1.00 to 3.83 mg/g (Table 1), the highest values were noted on locality 3 in the Vistula valley straight opposite the electric power plant chimneys. This valley is protected from the western side by hills. Similarly as in the case of fluorine, the lowest sulphur content was found in bark from the control sites and the locality 12 situated in south-east direction from the electric power plant.

Table 1

pH and content of some elements in *Pinus sylvestris* L. bark exposed to noxious emissions of aluminium works "Skawina" and from control sites

Site no.	Distance from emission source km	Direction from emission source	Zone	pH	F	S	Mg	Na	P	K	Ca
					mg/g						
1	0.3	NE	A	4.1	0.386	1.44	0	1.20	0.268	0.850	9.33
2	5.8	WNW		3.75	0.035	1.37	0.0070	1.55	0.120	1.000	9.06
3	3.4	NNW		3.3	0.051	3.83	0.0468	2.23	0.232	1.230	14.97
4	4.1	NNE	B	3.4	0.071	2.20	0.0103	1.32	0.445	0.925	14.66
5	5.4	NNE		3.6	0.060	1.37	0.0250	1.22	0.393	0.770	9.00
6	2.2	SE		3.25	0.030	1.55	0.0250	1.87	0.358	1.000	6.93
7	5.7	SSE		3.0	0.043	1.27	0.0275	1.26	0.255	0.770	2.70
8	8.3	WNW		3.2	0.035	1.12	0.0185	1.57	0.286	0.695	3.50
9	8.8	N		3.3	0.053	2.54	0.0268	1.08	0.197	0.850	8.83
10	8.5	N	C	3.3	0.077	2.89	0.0088	1.74	0.322	1.308	14.86
11	6.6	ESE		3.2	0.028	1.16	0.0268	0.93	0.214	0.770	4.30
12	9.5	ESE		3.25	0.034	1.00	0.0348	1.20	0.269	0.925	3.03
13	11.4	ENE	D	4.1	0.059	1.12	0.0335	1.32	0.286	1.080	15.40
14	30	N	K	3.4	0.019	1.04	0.0135	1.70	0.197	0.615	3.46
15	30	SE		2.9	0.018	1.07	0.0220	1.16	0.120	1.150	2.63

Magnesium was not detected in the surface pine bark layer from trees growing next to the aluminium works. In the remaining samples its amount varied from 0.0070 to 0.468 mg/g (Table 1).

Sodium content in dead bark layer varied from 0.93 to 2.23 mg/g phosphorus was present within the limits of 0.120-0.445 mg/g, and potassium content was within the range 0.615 (Ojców National Park) to 1.308 mg/g (Table 1). The above enumerated elements show no dependence on the situation of the sampling sites in relation to the industrial establishments.

Calcium in the pine dead bark layer varied from 2.63 to 15.40 mg/g (Table 1). Its highest content was found in bark collected from site 13 close to the soda plant and on localities 3, 4 and 10 lying on calcium substrate.

The first place among the elements analysed is occupied, as regards quantity contained in pine dead bark layer, by calcium, next comes sulphur and sodium. On site 15 least contaminated with SO_2 the situation changes and sulphur ranks as fourth after Ca, Na, and K (Table 2). The rating of fluorine also changes with the highest content on site 1. Most of the bark samples have a very low magnesium content, so that it usually occupies in the quantitative ordering of element content the last place and precedes fluorine only on localities 12 and 15.

Table 2

Sequence of quantitative occurance of analysed elements in the bark of *Pinus sylvestris* L. on the examined sites

Site	Elements
1	Ca > S > Na > K > F > P > —
3, 4, 5, 7, 9, 10, 11	Ca > S > Na > K > P > F > Mg
2, 6, 8, 13, 14	Ca > Na > S > K > P > F > Mg
12	Ca > Na > S > K > P > Mg > F
15	Ca > Na > K > S > P > Mg > F

The calculated linear correlation coefficients indicate that there is a positive significant correlation between: a) pH, and fluorine and calcium, b) sulphur and sodium, potassium and calcium, c) potassium and sodium, d) calcium and potassium (Table 3).

2. Evaluation of the degree of pine bark contamination by the biological test

The course of *Raphanus sativus* L. seed germination is shown in Fig. 2 in which the fluorine content in dead bark layer serving as substrate for the seedlings is also plotted.

Table 3

Linear correlation coefficient values between pH and analysed elements content in *Pinus sylvestris* L. bark

F	0.603***						
S	−0.078	0.065					
Mg	−0.022	0.073	0.0225				
Na	−0.195	−0.022	0.475*	0.157			
P	0.167	0.131	0.144	−0.290	0.076		
K	−0.005	−0.033	0.574**	0.183	0.522**	−0.359	
Ca	0.521**	0.226	0.678****	0.007	0.236	0.420	0.601***
	pH	F	S	Mg	Na	P	K

$P_{0.10}$=0.441* $P_{0.02}$=0.592***
$P_{0.05}$=0.514** $P_{0.01}$=0.641****

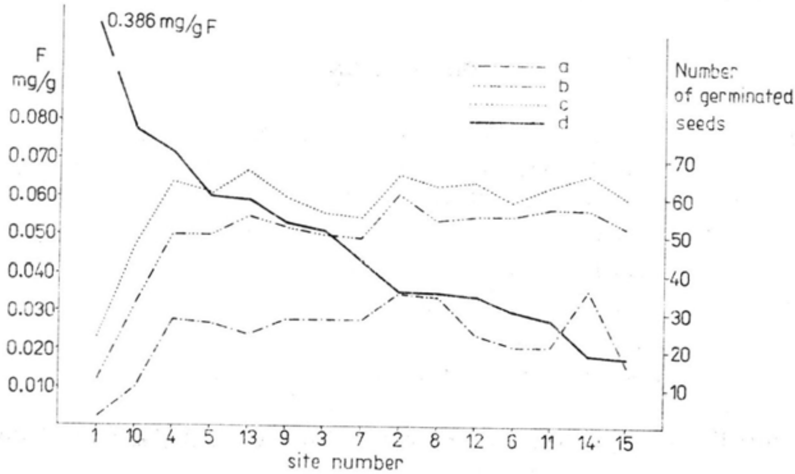

Fig. 2. Relation between number of germinating *Raphanus sativus* L. seeds and fluorine content in bark substrate from the particular sites

a — number of seeds germinating after 24 h, b — after 48 h, c — after 96 h, d — fluorine content

Germination of seeds was scarcest after 24, 48 and 96 h on bark substrate from locality 1. In this case the germination capacity of the seeds was lowered by 62 per cent as compared with that on bark from the control site. Germination was depressed less, by 22 per cent, on bark from site 10, and by 7-8 per cent on that from sites 3 and 7. In the remaining cases, particularly as regards localities 2, 4, 12, 13 and 14 the chemism of dead bark layer was even stimulatory for seed germination.

It results from the calculated correlation coefficients that there is a highly significant negative correlation between fluorine content in the bark and the number of radish germinating seeds (Table 4). No statistical correlation has, however, been demonstrated between bark pH and sulphur content, and the number of germinating seeds. As regards the remaining elements, a positive correlation was noted at a significance level of $P_{0.10}$ for magnesium.

Table 4

Linear correlation coefficient values between number of germinated seeds, and pH and content of some elements in bark of *Pinus sylvestris* L.

	pH	F	S	Mg	Na	P	K	Ca
No. of germinated seeds	−0.075	−0.817*****	−0.189	0.513*	0.050	−0.011	−0.022	0.045

$P_{0.10}=0.441$* $P_{0.001}=0.760$*****

DISCUSSION

The pH bark from Scots pine growing in this country under natural conditions is acid, with certain variations in dependence on the sampling site. For instance pine bark from the Białowieża Primeval Forest has pH 2.8 to 3.4 (Grodzińska, 1977), and in the Biała Forest complex pH is 3.12 to 3.20 (Zdanowska, 1976). For dead bark layer Filipek and Drogosz (1977) report values of 3.8 to 4.0. In the present study the pH of pine dead bark layer from the natural site situated at a 30 km distance south-east of the aluminium works "Skawina" was 2.9. On most localities exposed to industrial air pollution the bark pH was within the limits reported by Grodzińska (l.c.) for the Białowieża Primeval Forest or even higher. The rise of pH of dead bark layer on sites within the range of noxious emission of the aluminium works and electric power plant "Skawina" and the Cracow chemical plants is no doubt due to the chemical composition of the polluting substances emitted. Hence, in the analysed pine bark samples, beside an increased fluorine and sulphur content, an increase in calcium content raising the pH value was noted.

Fluorine content in dead pine bark varies and shows a dependence on the F content in the air. This element is not utilised by plants for synthesis of other compounds, therefore it is stored in the tissues. Moreover, the experiment demonstrated (Świeboda, Kalemba, 1978) that dead bark absorbs fluorine from the air. The sulphur content in the pine bark usually increases with the SO_2 concentration in the air. Contrary, however, to fluorine, this element is a component of various organic compounds in plants. Hence, under the conditions specified, particularly at lower SO_2 concentrations or a limited time of its action, significant changes in S do not occur in the living plant tissues, allowing unequivocal determination of the range of the noxious action of air pollution.

In the dead bark layer of *Pinus sylvestris* L. sulphur is usually positively correlated with sodium, potassium and calcium. The relation between S and K was demonstrated by Laaksovirta and Olkonen (1977) for Scots pine needles in industrial areas. The rise of the potassium level in plants is, as shown by Materna (1961), due to the action of sulphur dioxide present in polluted atmospheric air.

In the pine bark samples analysed the calcium content increased with the rising sulphur level. A similar dependence was demonstrated by Lötschert and Köhm (1977) in the bark of leafy trees. These authors suppose that calcium ions can be taken up together with sulphur or separately, but in similar amounts. Therefore, the content of calcium in the bark of trees growing in industrial areas may serve as indicator of air pollution.

In the chemical composition of dead bark layer from *Pinus sylvestris* L. calcium prevails (1%) (Filipek and Drogosz, 1977), while magnesium, potassium, sodium and phosphorus occur in much lower quantities. The present results demonstrated that in the dead bark layer of pines exposed to polluted air calcium also occupies the first place as regards the quantitative occurrence of the elements analysed, whereas magnesium is last on the list preceded by potassium, sodium and phosphorus. Moreover, in all bark samples sodium comes before potassium. The germination capacity of *Raphanus sativus* seeds on a powdered dead pine bark substrate reaches 67 per cent. This specific substrate no doubt depressed the germination process (Aaron, 1976), nevertheles, there was a marked difference between the results for sites situated close to the aluminium works and those for the control sites. Among the analysed elements, fluorine exerts an inhibitory effect on radish seed germination, as indicated by the highly significant correlation coefficient.

The present investigations lead to the following conclusions:

1. Scots pine bark is capable of absorbing chemically active compounds, therefore, it may serve as a biological indicator of the degree of air pollution both with gaseous substances and dusts which are mechanically deposited.

2. The chemical characteristic of this pine bark allows to establish the range of long lasting industrial emissions, whereas air analysis or plant tests reflect only the state of pollution in the period of measurement or exposure.

3. The bark pH of pines growing on industrial areas depends on the chemical composition of the polluted air. When emission is of complex character, the acidifying influence of SO_2 is neutralised, therefore the

pH and sulphur content in pine bark cannot be the only measure of the air pollution degree.

4. The fluorine content in dead bark layer of *Pinus sylvestris* and in transplanted bark sections from control areas is a very good bioindicator of air pollution with fluorine compounds and may serve for diagnostic purposes.

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Kora sosny zwyczajnej (Pinus sylvestris L.) jako biologiczny wskaźnik stopnia zanieczyszczenia powietrza atmosferycznego

Streszczenie

W korze martwicowej sosny zwyczajnej (*Pinus sylvestris* L.) zebranej na stanowiskach znajdujących się w promieniu 12 km od huty aluminium „Skawina" (ryc. 1) oraz na dwóch stanowiskach kontrolnych oznaczono pH oraz zawartość F, S, Mg, Na, P, K i Ca (tab. 1).

Na objętym badaniami terenie, oprócz huty aluminium znajduje się także elektrownia oraz zakłady sodowe, stąd też emitowane zanieczyszczenia mają złożony charakter chemiczny.

Odczyn analizowanych prób jest kwaśny (2,9-4,1 pH), wyraźnie wzrastający w pobliżu huty aluminium i zakładów sodowych. Zawartość fluoru w korze sosnowej waha się od 0,018 do 0,386 mg/g, a siarki od 1,00 do 3,83 mg/g. W próbach o zwiększonej zawartości siarki wzrasta także ilość wapnia.

W analizowanej korze martwicowej sosny zwyczajnej pierwsze miejsce pod względem ilościowym zajmuje wapń, a ostatnie zwykle magnez. Natomiast pozycja siarki i fluoru ulega zmianie w zależności od położenia danego stanowiska w stosunku do źródeł emisji (tab. 2).

Zależności zachodzące między odczynem kory martwicowej a zawartością poszczególnych pierwiastków przedstawiono w tabeli 3. Dodatni wpływ na wartość pH wywiera fluor i wapń.

Zdolność kiełkowania nasion rzodkwi zwyczajnej (*Raphanus sativus* L.) na podłożu sproszkowanej i odpowiednio nasączonej kory martwicowej sosny zwyczajnej była uzależniona od zawartości fluoru (tab. 4) i w przypadku stanowiska położonego najbliżej huty aluminium została obniżona o ponad 50% w stosunku do próby kontrolnej (ryc. 2).