

## EFFECTS OF CHEMICAL AIR POLLUTION ON FORESTS AND OTHER VEGETATION

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### 1. INTRODUCTION OR WHAT IS AT STAKE?

What is at stake at present? Much more than we imagine. Chemical air pollution can no longer be ignored. Its detrimental effects on vegetation can be observed all over Europe and in large parts of North America (Plate 1-4). Sick trees may be found also around the industrial areas in Asia, Africa and Latin America, and until now nobody has really discovered the change in soil types and ground vegetation in the tropics.

So, what is at stake?

Autotrophic vegetation as a whole has rendered possible the development of animals on earth, of consumers and predators at the higher levels in the food chains of ecosystems. More than 100,000 species of animals, most of them insects, depend on the producers' life. Mankind cannot survive without green plants and their capability of building up carbohydrates from just water and carbon dioxide in the chloroplasts of leaves by the use of solar energy. Agriculture and horticulture have made use of this capacity since prehistoric ages and forestry has developed from the uncontrolled use of firewood and timber for houses, ships, tools, and furniture to a planned production of renewable raw materials.

Vegetation has covered the ground except where it was prevented by permanent frost or dryness. It has enabled and enforced soil formation and prevented or reduced soil erosion by running water or deflation by wind. From many points of view, forests are the most developed form of vegetation. Their dominant plants, the trees, extend their roots deeper into the ground than herbs, grasses or shrubs; they grow much

higher, thus creating especially in the tropics several storeys, one above the other which are the habitat of innumerable insects, birds, and other animals. They have the greatest capacity to produce their own climate, which is less windy and more balanced than that of open plains. Forests reduce the extremes in water flow, both dryness and floods.

They prevent avalanches in the mountains, and within generations people in Austria and Switzerland have protected their "Bannwald" against any destruction. Finally, forests and other vegetation are indispensable for our pleasure, for outdoor recreation, and for our health by filtering poisonous gases and cancerogenic dust from the atmosphere. Now, that is at stake by chemical air pollution. Why and how?

## 2. THE DOUBLE ATTACK OF AIR POLLUTION ON VEGETATION

Vegetation is affected by air pollution on at least two different pathways:

- Direct effects on overground plant parts
- Indirect effects on plants by soil changes and by impact on roots.

One might add a third way:

- Changed disposition of host plants to parasites and lowered resistance to abiotic stress — frost, heat, wind, e.g.

### 2.1. *Direct effects of air pollutants on overground plant parts*

Plant leaves are surrounded by their skin, which consists of epidermic cells, mostly without chloroplasts, and a thin cork layer, the cuticula, which protects them against water loss and contact with toxic dust and gases (Fig. 1). Only marginal amounts of air pollutants can penetrate an intact leaf surface; however, acid droplets, for instance, below pH 3.4 or certain heavy metals like cadmium can cause stipples after having overcome the barrier. Most gases find their way into the plant through small natural openings, the stomata which usually are located on the lower leaf surface. Stomata open in the morning and usually close at night thus controlling CO<sub>2</sub> uptake and the rate of photosynthesis as well as stomatal transpiration, that is, water loss. Toxic gases have a chance to enter the leaf via the same stomata which are opened for

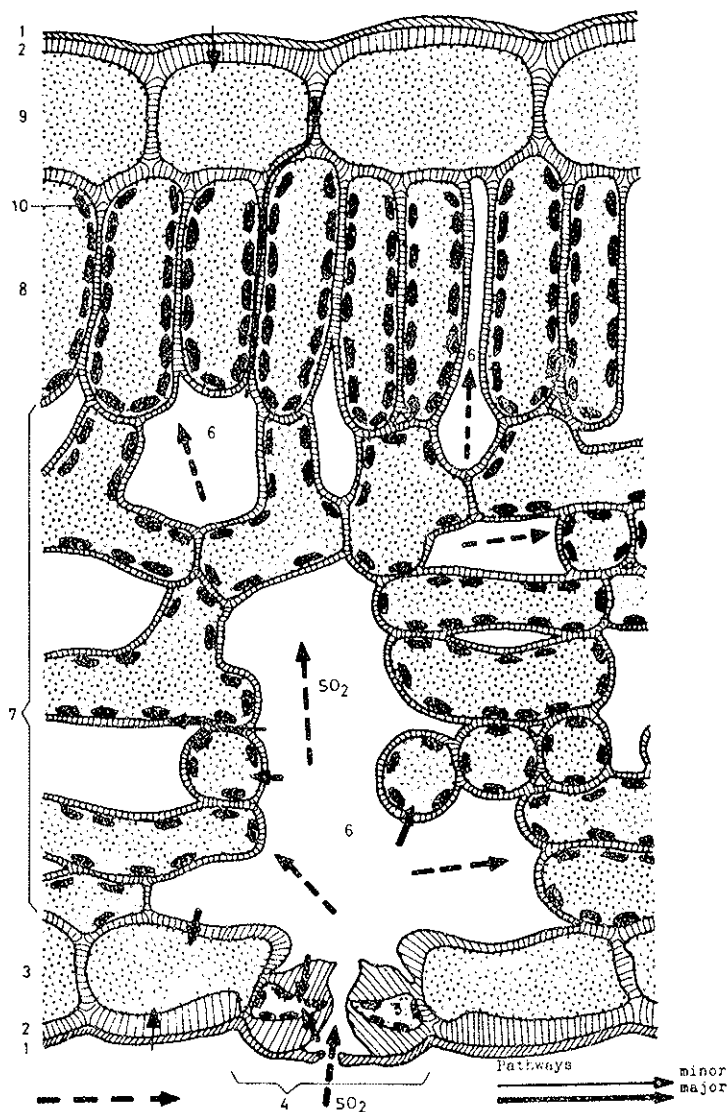


FIG. 1. Simplified scheme of uptake and distribution of  $\text{SO}_2$  in a green plant leaf. Broken arrow: Distribution of gaseous  $\text{SO}_2$ . Solid arrow: Diffusion of dissolved  $\text{H}^+$ ,  $\text{HSO}_3^-$ ,  $\text{SO}_3^{2-}$  and  $\text{SO}_4^{3-}$  (from Knabe, 1976. Design of cells redrawn from Nultsch, 1971).

- |                    |  |
|--------------------|--|
| 1) Cuticle         | 6) Intercellular space                         |
| 2) Cell wall       | 7) Spongy parenchyma                           |
| 3) Lower epidermis | 8) Palisade parenchyma                         |
| 4) Stomata         | 9) Upper epidermis                             |
| 5) Guard cell      | 10) Chloroplasts (other cell contents omitted) |

the necessary gas exchange. Water soluble gases like  $\text{SO}_2$  are then dissolved in the water-soaked cell walls and transported by the flowing water or just by diffusion to the cell interior, reaching the protoplasm and the green chloroplasts.  $\text{SO}_2$  and other pollutants interfere with plant metabolism in various aspects, the results perhaps being reversible depression of photosynthesis or irreversible necrosis of tissue. If larger parts of the plant are killed, the plant may die. Another possible effect is the contamination of fruits and fodder, which makes it hazardous for consumption by animals and men.

## *2.2. Indirect effects on plants by soil changes and by impact on roots*

Plants usually root in those soils which are appropriate to their demands with regard to soil reaction, nutrient, water, and air content. These conditions can be changed by air pollution. The soil may either become more acidic by deposition of acid gases, dust, and rain or be made more alkaline by basic dust around lime quarries or cement plants. It can also be contaminated by toxic substances, heavy metals, chlorinated hydrocarbons or radioactive particles. The change in soil conditions might reach an extent which will cause the dying of fine roots, leading to the degeneration of larger roots (Wieler, 1905, 1933; Ulrich *et al.*, 1979, 1983). Windfall of trees, withering of plants or changes in plant associations may be the result.

## *2.3. Changed disposition and resistance*

The ability of a plant to withstand the attack of parasites and plant diseases is strongly dependent on its vigour. Weakened trees are more successfully attacked by bark beetles and pathogenic fungi like *Armillaria mellea* (Schnaider and Sierpinsky, 1968). Also trees that have been exposed to  $\text{SO}_2$  are less tolerant to frost or low temperatures (Materna, 1974).

# 3. DIFFERENT WAYS TO LOOK AT THE PROBLEM

There are various ways to look at the problem. Let us discuss three of them:

- (I) Cause-effect studies
- (II) Effect-cause studies
- (III) Ecosystem analyses.

### 3.1. Cause-effect studies

Cause-effect studies follow a line described in Table 1. They always start with a given toxicant and look for its effect on plants or biocoenoses.

#### 3.1.1. Fumigation experiments

The first simple approach used in the last century by Stöckhardt (1871), consisted in applying a certain amount of gas to a chamber in which plants were grown. Stöckhardt proved the phytotoxic quality of sulphur dioxide because the fumigated plants showed leaf necroses or died under excess  $\text{SO}_2$  concentrations. However, as great parts of the gas were absorbed by the chamber wall, the toxicity levels were very much underestimated by this method.

Correct fumigation experiments therefore require the continuous

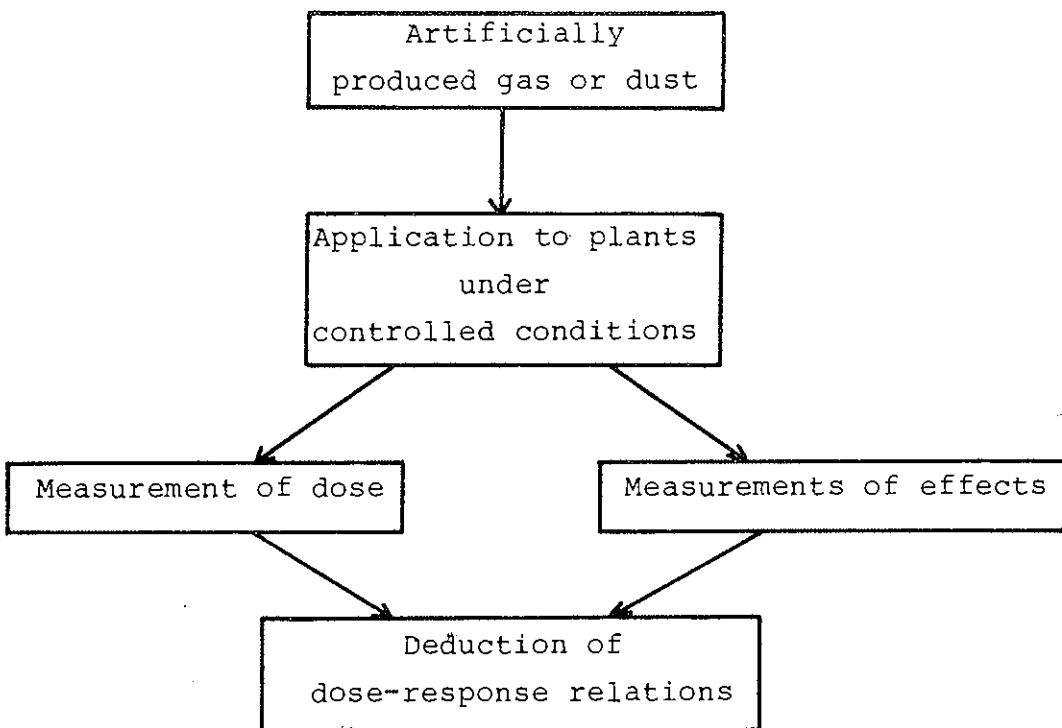


TABLE 1 - Cause-effect studies in the laboratory or greenhouse.

measurements of the toxicant within the chamber close to the plant and a steady flow of air and gas which have to be mixed properly.

Such studies were carried out by Thomas and Hill (1937).

However, critical concentrations were still underestimated because a greenhouse deprives plants of wind, dew, rain and natural climate. So open-top chambers, invented in the U.S. and used by Heagle *et al.* (1973), were a real improvement. They allowed a much more realistic view of the situation. Such laboratory experiments are the only way to get experimentally correct information on various questions such as, whether two components act additively, synergistically or antagonistically. Long-term effects and those which include the soil pathway are nevertheless hard to perform. Neither can large trees be investigated in such a way.

#### *Example 1 (unpublished data):*

Vegetative and generative progenies of two mother trees were fumigated in greenhouses with  $4 \mu\text{g} \cdot \text{m}^{-3}$  HF at three different times of the year in order to find out how much the seasonal variation of weather and plant development will affect the tolerance of Norway spruce. Another concentration of HF, either higher or lower, and a control without fumigation completed the experiment. One treatment at one period always contained 4 graftings and 8 seedlings of each mother tree. The fumigation method itself was developed by Guderian *et al.* (1969).

Parts of the results are shown in figure 2. The mean percentage of injured shoots per plant ( $\bar{y}$ ) is related to the time of exposure ( $x$ ). It is obvious that all progenies reacted much more sensitively in spring after flushing than in summer. Visible reaction in late fall was still less pronounced (not shown in figure 2). Typical symptoms of acute injury in early June can be seen in plate 5.

#### *Conclusion*

Air quality standards have to take into account the seasonal fluctuation of tolerance. The level below which no effect will be seen on Norway spruce under field conditions is, however, much lower than that found in this fumigation experiment. The observed differences between graftings and seedlings and between both progenies (see Fig. 2) require greater numbers of replication for verification and interpretation because environmental stress and genetic disposition have to be separated from each other.

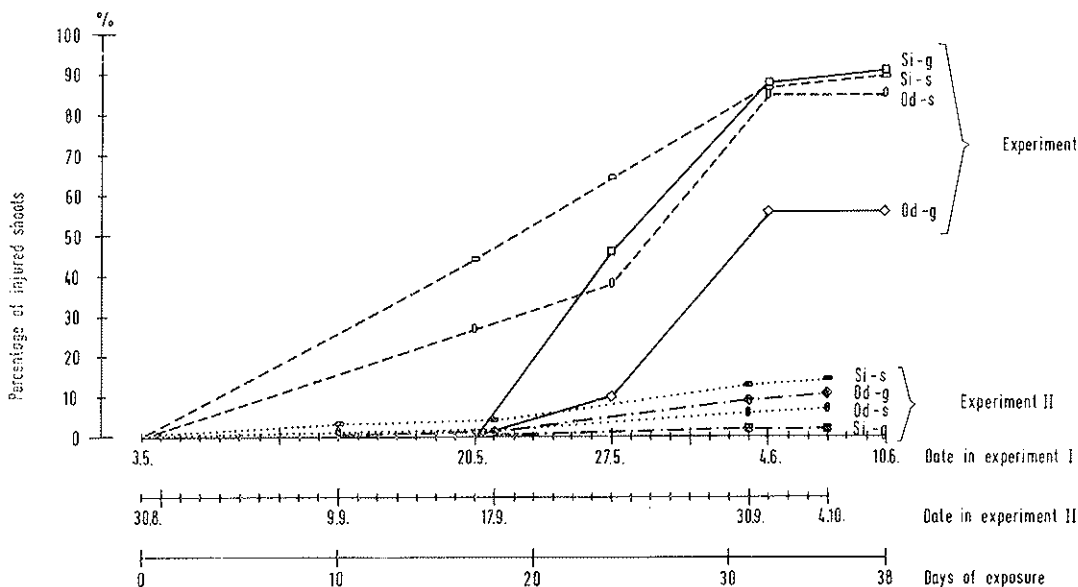


FIG. 2. Percentage of injured shoots of Norway spruce (*Picea abies* Karst) after fumigation with  $4 \mu\text{g HF} \cdot \text{m}^{-3}$  in May/June versus September. Means of 4 graftings and 8 seedlings, respectively. Fumigation from October to early December without hardly any difference to the control (not shown in figure 2).

Si-g: Graftings of clone Sieber  
 Si-s: Seedlings of mother tree Sieber  
 Od-g: Graftings of clone Oderhaus  
 Od-s: Seedlings of mother tree Oderhaus

### 3.1.2. Cause-effect studies in field experiments

Field experiments include all the variations of weather and ambient concentration of pollutants. Constant fumigation can be achieved neither by small artificial sources nor by exposing plant populations close to an industrial source. Hence the simultaneous measurements of air pollution and its effects on the exposed plants are a must.

#### *Example 2 (according to Guderian and Stratmann, 1968)*

The authors had exposed grains, vegetables, brushes, and tree seedlings in pots to the fumes of an iron smelter at Biersdorf, Germany, at increasing distance from the source which is shown in plate 1.

Station I was located close to the smelter,

Station VI, which served as a control, was situated in a neighbouring valley.

Plate 6 gives an impression of growing damage with decreasing distance from the source. The red currant bushes (*Ribes sanguineum*) at station III looked better than those at station II but they were still more affected than those at the control site VI. The authors carefully measured growth rates of all exposed plants and ambient  $\text{SO}_2$  concentrations.

### *Conclusion*

Buck (1970) concluded from the Biersdorf experiment that Norway spruce, Scotch pine, English oak, and European beech show growth depression at a mean concentration of  $0,08 \text{ mg SO}_2 \cdot \text{m}^{-3}$  if 2,5% of the measured values exceeds  $0,98 \text{ mg SO}_2 \cdot \text{m}^{-3}$  whereas no effect was found at a mean of  $0,07 \text{ mg SO}_2 \cdot \text{m}^{-3}$  and a 2,5 percentile of  $0,59 \text{ mg SO}_2 \cdot \text{m}^{-3}$ . The importance of the ratio of mean to peak concentrations in risk studies has been discussed by Buck (1970) and Knabe (1971).

The maximum immission values for  $\text{SO}_2$  as suggested by the German Association of Engineers (VDI) were strongly influenced by such experiments (Table 2). The even lower and stricter "air quality standards to protect forests" proposed by the International Union of Forest Research Organization (IUFRO) relate, on the contrary, to dose-response studies in injured forests, also including unfavourable site conditions (Table 3).

### *3.2. Effect-cause studies*

#### *3.2.1. General considerations*

Experts are asked to explain why certain plants are suffering or dying away and what toxic air pollutant or what source may be responsible for this. The general procedure of an expert can be taken from table 4. The expert should not restrict his job to just looking at the reported plant damage. Instead, he should include other plant species, the distribution of injured plants, and their relations to industrial or other sources. He should also collect plant samples to find out whether certain elements have accumulated in excess or are below the physiologically necessary minimum content which requires strict observation of certain prescrip-



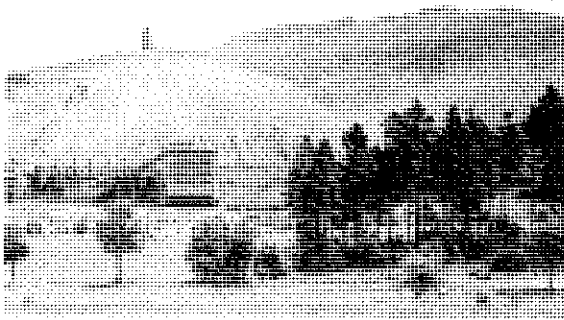


Plate 1. Denuded zone around a small iron smelter without cleaning of stack gases as a result of high ambient concentrations of  $\text{SO}_2$ . Heavy soil erosion after loss of protective vegetation could not be prevented by terraces. Typical for careless industry in the Fifties. Biersdorf, W. Germany, 1959 (Photograph by courtesy of H. van Haut).



Plate 2. Denuded hills around a copper smelter. Trees, shrubs and herbs were killed. Gully erosion can also be seen. Water courses may be blocked by eroded materials. Ogden, Utah U.S.A., 1969.

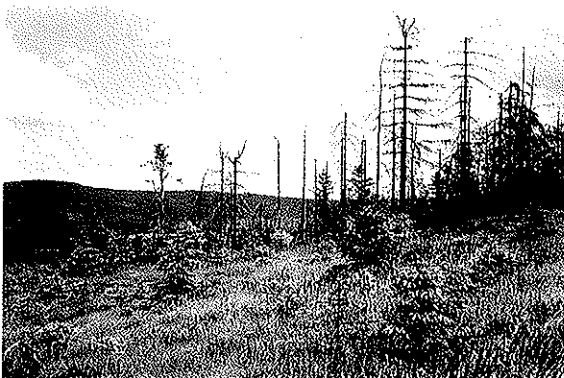


Plate 3. Forest destruction on the top of mountains in the Erzgebirge, Krusny hory, situated between the German Democratic Republic and Czechoslovakia caused by  $\text{SO}_2$ -emissions from lignite fired power plants 25 km away. Old trees, meanwhile cut, died first, medium aged trees followed. Young plants which should replace dead stand, have no future. North Bohemia, June 1970.

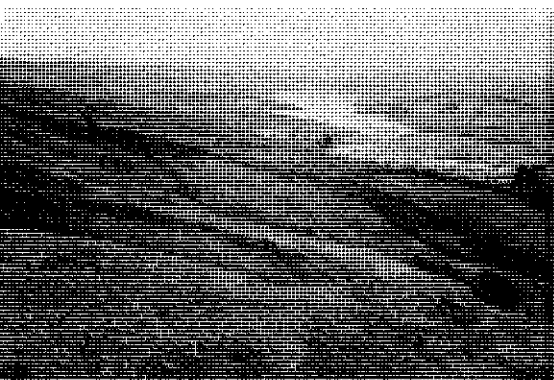


Plate 4. Large scale destruction of land in the Copper Valley, Tennessee. Attempts of revegetation after reduction of  $\text{SO}_2$  emissions showed little success because of soil degradation. New Coppertown, Tennessee, U.S.A., 1975.



Plate 5. Typical symptoms of injury caused by hydrogen fluoride during the most sensitive period of Norway spruce (*Picea abies* Karst). Single needles or parts of shoots appear burned. Duisburg, W. Germany, June 1976.

TABLE 2 - Proposed maximum concentrations of SO<sub>2</sub> (MIK-Werte) for the protection of plants in Germany. Values in mg SO<sub>2</sub> · m<sup>-3</sup> (according to VDI, 1976 \* and 1978 \*\*).

	Year	Mean concentration		
		Growth Season	30 min. not exceeding . . .	
			percent of time	
			2.5%	5.0%
	*	**	**	*
1. MOST SENSITIVE PLANTS:				
<i>Species of the genera</i>				
Abies, Picea, Juglans, Ribes, Trifolium, Medicago	0.06	0.05	0.25	0.22
2. SENSITIVE PLANTS:				
<i>Species of the genera</i>				
Picea, Pinus, Larix, Tilia, Fagus, Malus, Hordeum, Avena, Secale, Triticum	0.09	0.08	0.40	0.35
3. LESS SENSITIVE PLANTS:				
<i>Species of the genera</i>				
Acer, Alnus, Populus, Quercus, Prunus, Rosa, Solanum, Zea, Vitis, Beta, Brassica, and species of the family of the Liliaceae	0.13	0.12	0.60	0.53

tions (Knabe, 1982). The expert also needs the knowledge or assistance of entomologists and phytopathologists to clearly distinguish between the effects of insects, diseases, and abiotic factors, including pollutants.

Several pictorial presentations may be of great value to the identification of the classic air pollutant symptoms, but they do not help very much to find the causes of the so-called "new" types of injury to trees. The following publications can be recommended: Lacasse and Moroz (1969), van Haut and Stratmann (1970), Jacobson and Hill (1970), Malhorta and Blauel (1980), and Farbbildheft der Allgemeinen Forstzeitschrift zum Erkennen von Immissionsschäden an Waldbäumen (1983).

### 3.2.2. Practical procedure for an expertise

At first, the expert has to check the order for the expertise and to

TABLE 3 - International Union of Forestry Research Organizations, Subject Group Air Pollution: Maximal concentrations of air pollutants for the protection of forests (in  $\mu\text{g}/\text{m}^3$  air).

Kind of Protection	Component	Annual mean	Mean of 24 hours	97.5 percentile of the 30' values
1. For the protection of full productivity on most sites	$\text{SO}_2$ $\text{HF}$	50 0,3	not more than 12 times per year exceeded 100	150 0,9
2. Also for the maintenance of protective and social functions of forests on critical or extreme sites	$\text{SO}_2$ $\text{HF}$	25 still to be clarified	50	75

Ljubljana, September 27<sup>th</sup> 1978 (IUFRO 1979).

clarify his own expert status. Then the work begins, always remembering the general considerations listed above by first looking at all parts of the affected plants.

### *Crown and leaves*

Are there any changes in colour of leaves or needles? Can there be seen signs of necrosis? If so, are they irregularly distributed, which often is considered as typical for ozone injury, for example, in tobacco or pinto beans (Plate 7 and 8)? Or are they concentrated on leaf margins and tips, which often is observed after the slow accumulation of chloride (Halbwachs, 1963) or fluoride (Guderian *et al.*, 1969)? Plate 9 shows typical marginal necrosis of an elm leaf, plate 10 tip necrosis of tulip leaves, whereas the flowers themselves are not injured. Acute injury by  $\text{SO}_2$  often appears as interveinal chlorosis followed by necrosis which can be seen at an elm leaf in plate 11. On the contrary, ozone injury mostly consists of small chlorotic or necrotic spots. Pine needles (plate 12) and

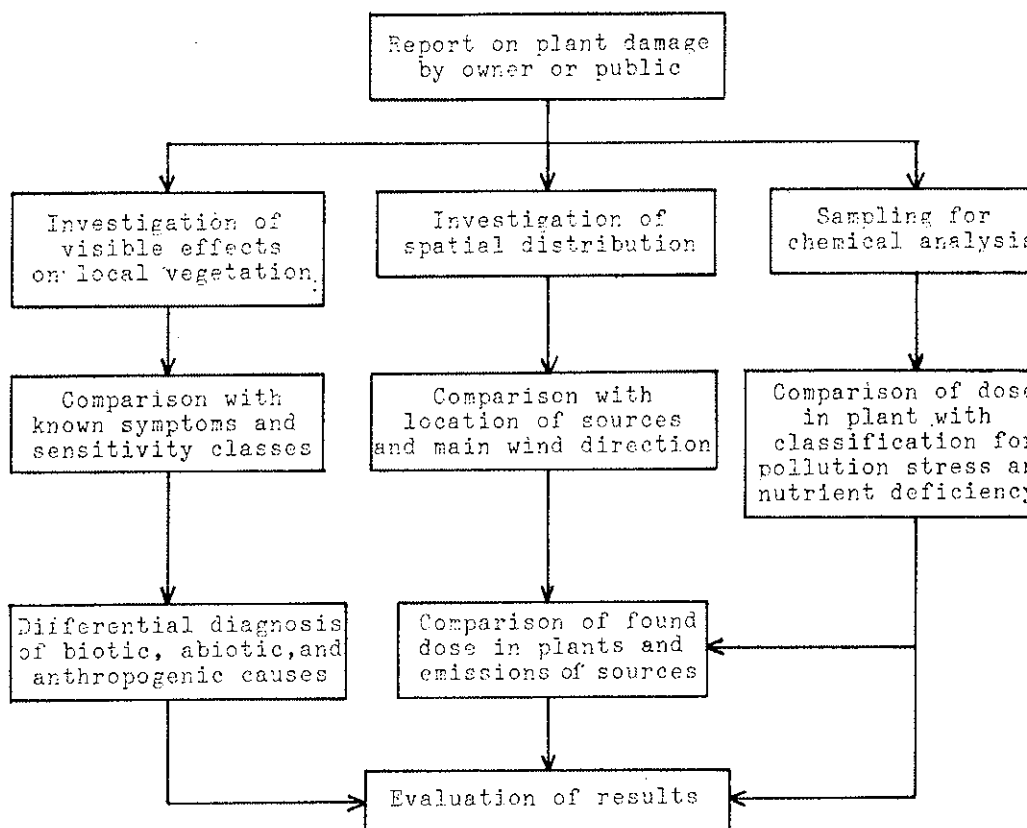


TABLE 4 - Effect-cause study. Search for causes or sources of observed plant damage.

tobacco leaves (plate 7) may serve as an example for this mottling syndrome. Bleaching of upper needles which are exposed to sunshine and dew (plate 15) may be the result of photochemical reactions within the plant cells.

#### *Premature senescence and abscission*

Are the leaf retention and colour as usually developed in other years or do the crowns appear unnaturally thin? A special sign of pollutant impact on leaves is premature senescence due either to the induction of ethylene biosynthesis by stress (Ziegler and Berndt, 1983) or just to



Plate 6. Effects of  $\text{SO}_2$  on red current brushes (*Ribes rubrum* L.) in the second year of exposure to fumes of an iron smelter. Sparse foliage, small leaf size, premature leaf abscission, and little or no fruit could be seen at a distance of 600 m from the source (left). The damaging effects were less expressed at a distance of 750 m. Healthy looking brushes could be observed at the control site situated 6000 m from the source. Biersdorf, W. Germany, 1960 (right). Photograph by courtesy of H. van Haut.



Plate 7. Sensitive tobacco plants (*Nicotina tabacum* L., strain Bel W 3) with necrotic spots on leaves caused by ambient ozone, light brown spots having been induced by ozone injury 24 to 48 hrs ago. After longer time of exposure, dead tissue will be bleached. Phytotoxic concentrations of ozone in W. Germany have been demonstrated for the first time by the use of tobacco as indicator plant (Knabe et al., 1973). Kettwig, W. Germany, 1967.



Plate 8. Necrotic leaf tissue of young bean plants (*Phaseolus vulgaris* L.) reveals the impact of oxidants, mainly of ozone. This plants is used as bioindicator in the U. S. to control oxidant air pollution. Cincinnati, Ohio, U.S.A., 1967.

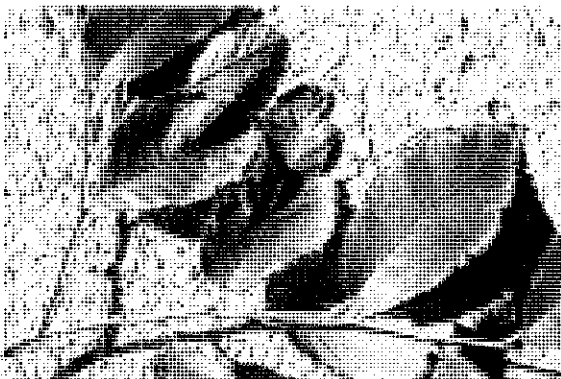


Plate 9. Marginal yellow leaf chlorosis and brown necrosis as a result of the accumulation of fluoride in leaf margins of elm

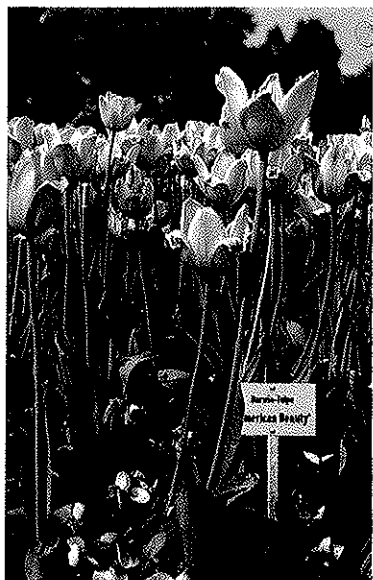


Plate 10. Spoiled «American Beauty». Leaf tips of this beautiful tulip strain (*Tulipa gesneriana* L.) burnt by accumulation of air born fluoride. Cologne, W. Germany, April 20, 1971.

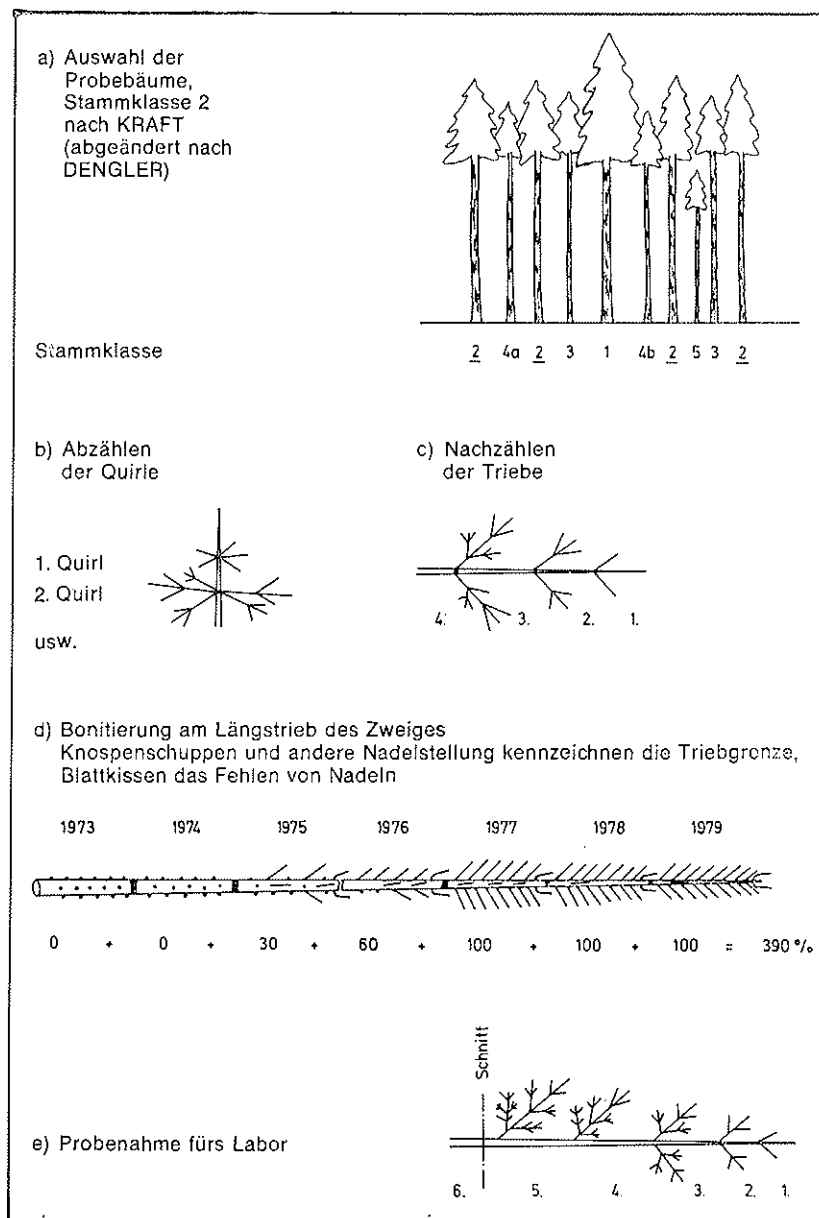


FIG. 3. Survey in spruce forest. a) Selection of sample trees class 2 (dominant); b) Counting of whorls from top to seventh whorl; c) Re-counting of annual shoots from top to seventh shoot; d) Assessment of needle retention at main lateral shoot; e) Sampling for later chemical analysis.

the accumulation of pollutants. The physiological aberration results in leaf drop in August or in the reduction of living needle years from 6 or 7 (plate 13) to 4, 3 or even less (plate 14). A strong correlation of needle loss with the accumulation of F, Cl and S in needles of Norway spruce was demonstrated by Knabe (1983). The correlation coefficient ( $r$ ) ranged from 0.7 to 0.8.

### *Twigs and stem bark*

Are twigs and the bark all right? For example, are there dead secondary branches on living main branches? A unique symptom is green leaves or needles and branches of Norway spruce lying on the ground apparently fallen down from their original position (plate 16). The cause is not known. Bark wounds without any mechanical impact are also to be looked for. Their position on the lower side of horizontal branches hints at poisonous droplets hanging there after light rainfall (plate 17). Necrotic bark spots along the stem can also be observed. They may be dry (plate 18), resinified (plate 17) or wet with stem sap oak. At present, it cannot be decided whether the fungus *Pezicula cinnamomea* acts as primary pathogen in the bark disease of red oak (Butin, 1983) or occasionally invades the plant as secondary pest after the impact of acid stem flow or acidification of the soil.

### *Roots*

Are the roots healthy and well distributed?

The distribution of fine roots in the various soil horizons can be used as a good indicator of root health. Root development is considered as very poor in soil layers containing 1 to 2 fine roots per  $\text{dm}^2$ . 5 to 10 fine roots per  $\text{dm}^2$  or 10 to 20 fine roots per  $\text{dm}^2$  are regarded as a mean or strong root development respectively. Dying root tips, for example at the border between humus layer and mineral soil hint at toxic soil conditions in the lower horizon (see Fig. 4).

### *Soil*

And what could have happened to the soil?

Ulrich *et al.*, (1984 a) have given a series of criteria for interpreting the results of soil analyses.

Soil investigations start with digging a hole with three clean sides

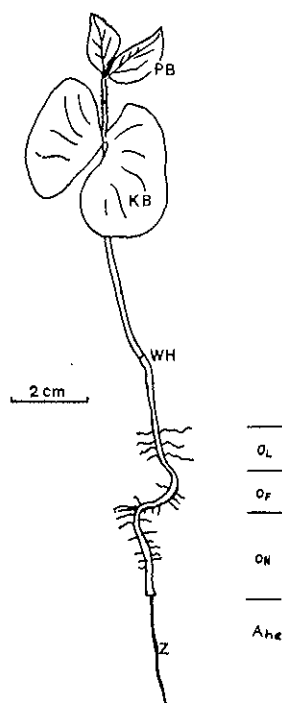


FIG. 4. Distribution of fine roots of European beech seedlings in various soil horizons (according to Becker 1982).

- $O_L$  — Litter layer
- $O_F$  — Fermentation layer
- $O_N$  — Humus layer
- $A_{he}$  — Humus containing mineral layer (eluvial horizon)

and with then describing the humus conditions and soil horizons. In the mountains of Northrhine-Westphalia, the beginning of podsolization with high contents of free or exchangeable iron and aluminium can often be observed.  $Fe^{3+}$  can easily be determined by the use of a 10% solution of KSCN by forming the red complex of  $Fe(SCN)_3$  which is then made more visible in butanole (Knabe and Bartels, in prep.).

### Example 3 (unpublished data)

A plantation of sycamore (*Acer pseudoplatanus*) on sandy soil to the north of Recklinghausen in the Ruhr district showed extended marginal



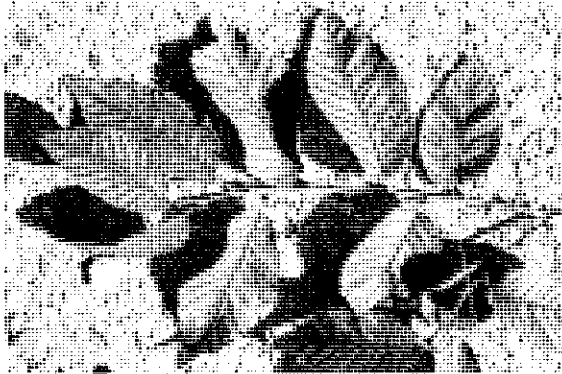


Plate 11. Intervetinal chlorosis of elm (*Ulmus pumila* L.) which is regarded as typical symptom of acute impact of  $\text{SO}_2$ . Compare difference to plate 9. Ogden, Utah, U.S.A., 1969.

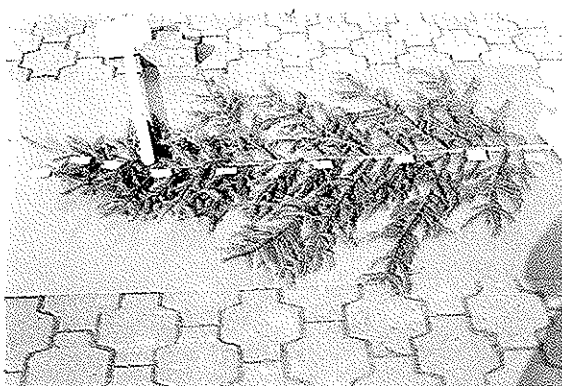


Plate 13. Six to seven living needle years on branch of Norway spruce (*Picea abies* Karst) are only to be found in areas with clean air. This example of low land spruce in Lower Saxony, W. Germany, even shows some needles from 1964. Cloppenburg, W. Germany, March 1973.



Plate 14. Only one living needle year on twig of young Norway spruce (*Picea abies* Karst) reveals high chronic air pollution on top of mountains in North Bohemia. The photograph was taken close to plant shown on plate 3. This tree cannot survive because young shoots are missing supply from older needles. North Bohemia, June 1970.



Plate 12. Mottling of one year old needles of Eastern white pine (*Pinus strobus* L.). Place unknown. Photograph courtesy of Arthur Costonis, July 8, 1966.

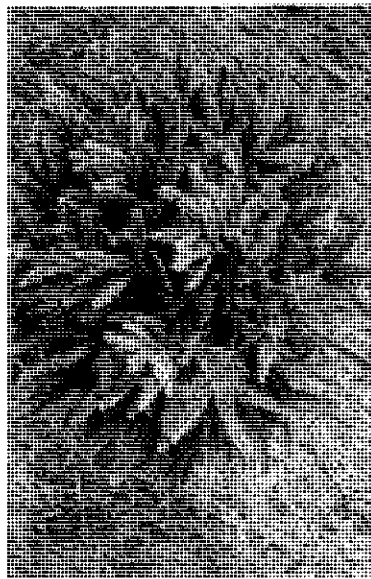


Plate 15. Chlorotic plants of Norway spruce (*Picea abies* Karst) in polluted area. Lower branches are still green. Cause may be due to photochemical reactions within the needles or on the needle surface. Kliny, North Bohemia, April 1983.

leaf necroses (see plate 19). The symptoms on leaves were similar to those observed after fluoride fumigation. However, analyses of leaves did not show an increase in fluoride, chloride or sulphur when healthy looking plant leaves were compared with sick ones (table 5). The sick looking plant leaves contained even less F as well as Ca and Mg so that nutrient deficiency seemed possible. Slightly higher contents of Cu and Si as well as a 2.5% higher content of Mn were determined. Excavated plants showed the almost complete absence of fine roots in the upper soil horizon below the humus layer. No roots had found their way from the planting hole to the neighbouring soil stratum (plate 20).

When tested with KSCN as described above, the rootless horizons showed high concentrations of free  $\text{Fe}^{3+}$  despite the poor sandy soil. (Table 6).

TABLE 5 - Element content in leaves of sycamore (*Acer pseudoplatanus* L.). Arithmetic mean and standard deviation of 8 sick and 8 healthy looking plants. All values in  $\mu\text{g} \cdot \text{g}^{-1}$  dry material (= ppm).

Component	sick		healthy		t-value ( $n_1=n_2=8$ )
	mean	stand dev.	mean	stand dev.	
a) No significant difference between sick and healthy leaves					
Pb	6.88	2.47	5.88	1.55	0.97
Zn	79.25	19.26	91.63	42.84	0.75
K	11400.00	2038.91	10887.50	1399.42	0.59
Cl	2312.50	830.55	2550.00	845.15	0.57
S	3812.50	622.06	3462.50	821.04	0.96
P	1312.50	180.77	1237.50	184.68	0.82
b) Contents in sick leaves lower than in healthy ones					
Ca	3950.00	772.75	8537.50	1572.02	7.41**
Mg	887.50	195.94	2457.00	328.42	11.74**
F	19.22	5.46	40.57	10.28	5.19**
c) Contents in sick leaves higher than in healthy ones					
Cu	9.75	1.39	6.00	1.51	5.18**
Mn	1345.00	297.75	512.50	164.56	6.92**
Si	14412.50	2687.23	10375.00	2489.42	3.12***

\*\* significant at the 1% level,

\*\*\* significant at the 0.1% level.

TABLE 6 - Soil profile beside a sycamore tree (*Acer pseudoplatanus* L.) in the Haard forest, north of Recklinghausen, Germany.

Depth [cm]	Horizon	Test on FeIII	Remarks
+ 3 to 0	H	3	Humus layer at the stem foot of <i>Pteridium aquilinum</i>
- 1 to - 5	A <sub>e</sub>	2	Lower stolon region of <i>Pteridium aquilinum</i>
- 11 to - 16	A <sub>e</sub>	4	Almost rootless
- 16 to - 20	B	5	Rootless or dead roots
- 22 to - 25	B	5	Rootless, only very few sycamore roots
- 35 to - 40	C	1	Nearly rootless, yellow sand
- 60	C	1	Living roots, light yellow sand
- 90	C	1	Light yellow sand

Evidence for the toxic acidification of soil by the use of the test on soluble iron (according to Knabe and Bartels, in prep.).

Range 1: no FeIII detectable, range 5: 100 µg FeIII/0.2 g soil, approx.

Mineral soil: Rootless within 0-15 cm, approx.

Border line between humus layer and mineral soil = 0 cm.

### Conclusion

The withering of sycamore plants on sandy soil seems to be more likely the result of soil acidification and nutrient deficiency than that of direct impact of air pollutants. Final confirmation of this hypothesis will only be provided by more detailed soil and root investigations as well as by fertilizing experiments.

### 3.3. Ecosystem analysis

Ecosystem analysis has become a major scientific approach which was very much enforced by the International Biological Program (IBP) the aim of which being the evaluation of biomass production in different climates and vegetation types and the quantitative determination of the flow of energy, water, and chemical components in order to lead to a better understanding of food chains, natural successions, and interactions of biota with abiotic environment.

Originally, air pollution was not part of the concept. The request for also doing immission measurements in those places where intensive

biological research is carried out — as had been suggested by Knabe (1971), at the IUFRO Congress in Gainesville, Florida — was not realized by the research groups at that time. But as soon as experimental results of deposition measurements revealed an astonishing and unexpected input of air pollutants into forest ecosystems, the interest grew rapidly. Now every book on air pollution contains chapters dealing with the ecosystem approach (Craig *et al.*, 1980; Smith, 1981; Moriarty, 1983 and Treshow, 1984) and handbooks of ecology can no longer neglect the problems of air pollution. Ulrich in Germany as well as Likens and Borman in the United States belong to the group of those scientists who promoted very much such studies of environmental interactions.

*Example 4 (adapted from Ulrich et al., 1984 and VDI, 1983)*

Ulrich and co-workers determined the precipitation ( $r$ ) and element input ( $e$ ) above a meadow ( $F_{1r}$  and  $F_{1e}$ ) and below a tree stand ( $F_{2r}$  and  $F_{2e}$ ) of both Norway spruce and European beech, as well as the element output by drainage and percolation below the root horizon ( $F_{3r}$  and  $F_{3e}$ ) in the Solling mountains, Germany (Fig. 5a). With the exception of heavy fog days,  $F_{1r}$  is always greater than  $F_{2r}$  because some water does not reach the ground. The difference in rainfall ( $F_{1r} - F_{2r}$ ) has been called "interception" which is the sum of evaporation and transpiration.

$F_{1e}$  is mostly smaller than  $F_{2e}$  because dust, certain gases, and dissolved elements of fog droplets are filtered out from the atmosphere by the forest canopy. The difference in element flow ( $F_{2e} - F_{1e}$ ) can therefore be regarded as a first but rough quantitative assessment of the atmospheric input to the canopy ( $F_{7e}$ ) (Knabe, 1977). The difference  $F_{2r} - F_{3r}$  gives the total water consumption of the forest stand, the difference  $F_{2e} - F_{3e}$  the amount of element accumulation in the soil or element loss, if negative, within a given period.

It appears as  $F_{10e}$  in Fig. 5b.

A more detailed model is shown in this Fig. 5b. The throughfall  $F_2$  of Fig. 5a is replaced by the partial fluxes stem flow ( $F_{11}$ ) and canopy drip ( $F_{12}$ ). There are also included additional fluxes as litter fall  $F_{4e}$ , dry deposition to soil  $F_{5e}$  and internal fluxes of the system as uptake by roots and flow to the crown  $F_{6e}$ , leaching of leaves  $F_{8e}$  and flow from roots to soil  $F_{9e}$ .

In this respect, the VDI (1983) gives quite an understandable calculation (pp. 179-185).

According to the results of Ulrich, there is to be considered an input

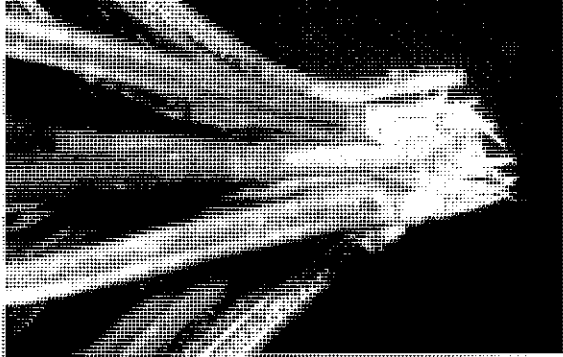


Plate 16. End of shoot of Norway spruce (*Picea abies* Karst) found on the forest floor. Shoot was torn off probably after its consistence had been weakened by environmental stress or lack of nutrients. Witten, W. Germany, March 1983. Photograph courtesy of Ms. Ebel.



Plate 17. Bark wounds from Norway spruce (*Picea abies* Karst) partly covered by raisin on lower side of branch photograph taken from below and on upper stem, here in original position. Hardeggen, Solling, W. Germany, 1982.



Plate 18. Bark disease of Northern red oak (*Quercus rubra*) in area with high air pollution. This disease is not found in areas with clean air and good soil conditions. The fungus *Pezicula cinnamomea* may be involved. Haard Forest, North to Recklinghausen, W. Germany, 1977.



Plate 20. Soil profile with roots of injured sycamore maple tree (*Acer pseudoplatanus* L.) from plate 19. Roots could not leave the plant hole but only rooted into deeper soil parts which are not as acidified as the upper soil horizons. Haard Forest, North to Recklinghausen, W. Germany, 1982.



Plate 19. Heavy injury on leaves of sycamore (*Acer pseudoplatanus* L.) maple showing marginal necroses which are not due to fluoride pollution because of low fluoride contents in leaves. Lack of magnesium as a result of soil acidification by acid rain may be the reason. Haard Forest, North to Recklinghausen, W. Germany, 1982.

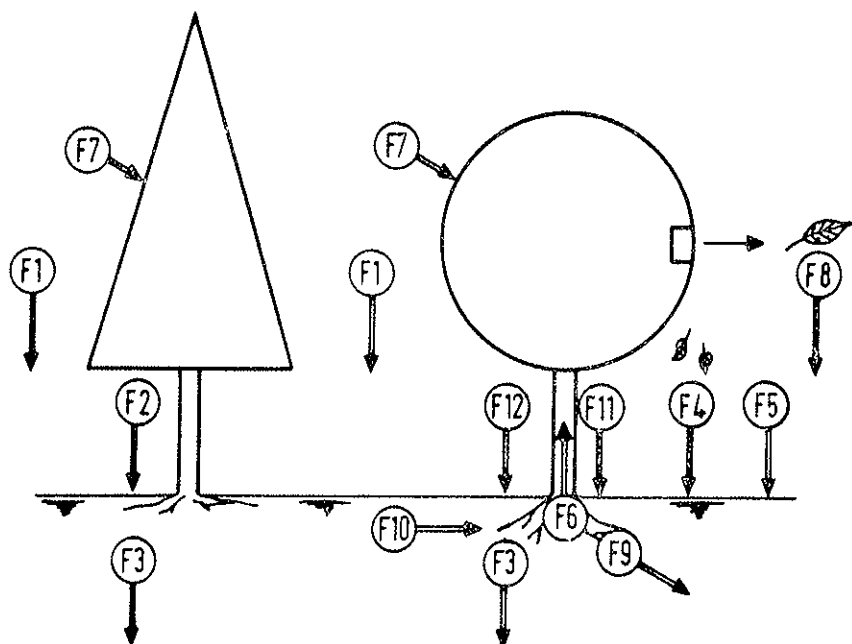


FIG. 5. Element fluxes in a forest ecosystem (Changed from VDI 1983).

Fig. 5a. "Black box"-model of main fluxes.

Fig. 5a. "Black box"-model of main fluxes.

F 2 Throughfall below forest stand

F 3 Percolation to ground water

F 7 Interception deposition to canopy

Fig. 5b. More detailed model of fluxes.

F 1, F 3, F 7 see Fig. 5a.

F 2 replaced by F 11 and F 12

F 4 litter fall

F 5 dry deposition to soil

F 6 uptake by roots and flow to crowns

F 8 leaching of leaves

F 9 flow from roots to soil

F 10 flow into soil storage

F 11 stem flow

F 12 canopy drip

of acids into the Solling from 1966 to 1973 as follows (values given in  $\text{kmol H}^+ \cdot \text{ha}^{-1} \cdot \text{a}^{-1}$ ):

wet deposition = 0.80

interception beech = 0.84

interception spruce = 2.61

production in soil = 2.50

Thus, deposition of air pollutants may be the minor or major source

of man-made soil acidification.  $1 \text{ kmol H}^+ \cdot \text{ha}^{-1} \cdot \text{a}^{-1}$  is equivalent to the annual lime requirements of  $100 \text{ kg CaCO}_3$  per ha.

Because natural weathering of minerals in the upper soil horizons is not sufficient to neutralize that amount of acid, a gradual acidification takes place so that even calcareous soils can be affected, especially at the stem base of beech and red oak, both possessing high stem flow rates. In silicate soils, clay minerals can be destroyed, releasing  $\text{Al}^{3+}$  and  $\text{Fe}^{3+}$ , by replacing  $\text{mol Ca}^{2+}$

$\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  at the exchange sites of minerals. If the ratio  $\frac{\text{mol Al}^{3+}}{\text{mol Ca}^{2+}}$  is lower than 0.1 in an extract of  $2 \text{ ml H}_2\text{O} \cdot \text{g}^{-1}$  soil, aluminium is regarded as being toxic for the roots of trees and other soil plants (Ulrich *et al.*, 1984). Roots of Norway spruce may already suffer at values below 0.1. Besides that, the authors still recommend other criteria.

### Conclusion

Acid rain, or in more scientific terms, acidic deposition, has to be regarded as a real menace to wide areas in the Northern Hemisphere. There are plenty of sites, for instance in Northrhine-Westphalia, where roots remain in the humus layer and do not grow into the upper mineral soil horizon. The acids remove cations which are indispensable for plant nutrition and dissolve aluminium and heavy metals, both of geogenic and atmospheric, that is, anthropogenic origin. This results not only in root damage as mentioned above, but also in disturbed mycorrhiza and a reduced number of soil animals, as shown in fig. 6. The depletion of nutrients by leaching and the reduced circulation within the ecosystem is affecting the nutrition, resistance and growth of plants. The loss of sensitive plants and soil animals will further reduce the richness in species of a forest or lead to a final tree die-back, eventually to non forest successions which cannot fulfill all welfare functions of woods. A simplified model of these indirect effects of acid deposition is shown in fig. 6.

### 3.4. Climatic risk factors

Climatic observations can help to understand plant damage caused by air pollution better than just being focused to its concentration, because combined effects may play an important role. Climatic maps have to be read therefore from another point of view than before. Three examples are mentioned.

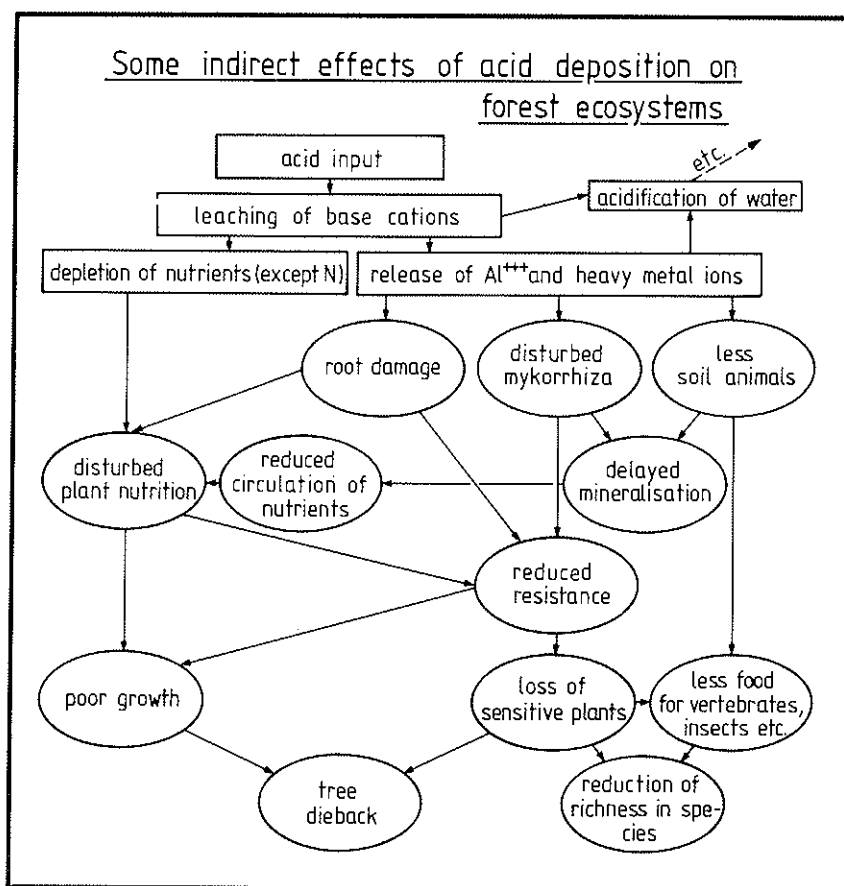


FIG. 6. Results of acid deposition and soil acidification.



*Example 1: Height of precipitation*

The amount of acidic deposition depends very much on the amount of precipitation. Hence, the risk from acidic deposition is usually increasing with rainfall.

This development is changing the importance of rainfall to forests from a very positive factor to an eventually negative factor for plant growth.

Fig. 7 shows a map of Northrhine-Westphalia describing this risk under the supposition of equal concentration and soil susceptibility. This map will have to be modified by completing it with concentration values and the results of soil investigations. The present "pilot project" of the State Institute of Ecology of Northrhine-Westphalia will fill up this gap (Block and Bartels, 1983). It should be mentioned that "high risk" in Northrhine-Westphalia may be regarded as medium risk in countries with rainfall of up to 2000 mm.

*Example 2: Combination of frost and pollution*

The map of mean temperatures in January of Northrhine-Westphalia (Fig. 8) shows those areas below a mean of 0°C where a combination of frost and SO<sub>2</sub> can lead to plant damage. Norway spruce, e.g., can tolerate winter temperatures of about -40°C in healthy conditions, but -10°C might be too much for a plant weakened by preceding or corresponding pollution impact. Forests close to the national borders of distribution are obviously especially endangered.

The combination of ammonia (NH<sub>3</sub>) and frost has also detrimental effects on coniferous trees. The needles of young fir plants (*Abies nordmanniana* Spach) in the surrounding of a chicken farm near Hamburg turned brown after cold winters, whereas they remained green after mild winters as observed by the author in the sixties.

Climatic maps of greater periods are better for risk analysis, actual maps of the climatic situation in individual years would be better for the interpretation after the damage had occurred.

*Example 3: Poisonous fog*

Fog is usually regarded as a positive factor for a forest stand because trees are filtering out additional moisture. Some forest societies in the subtropic or tropic mountains depend very much on this kind of precipitation as on the island of Tenerife or on Mount Kenya. Strongly acidified

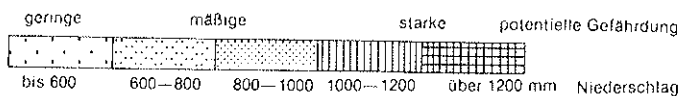
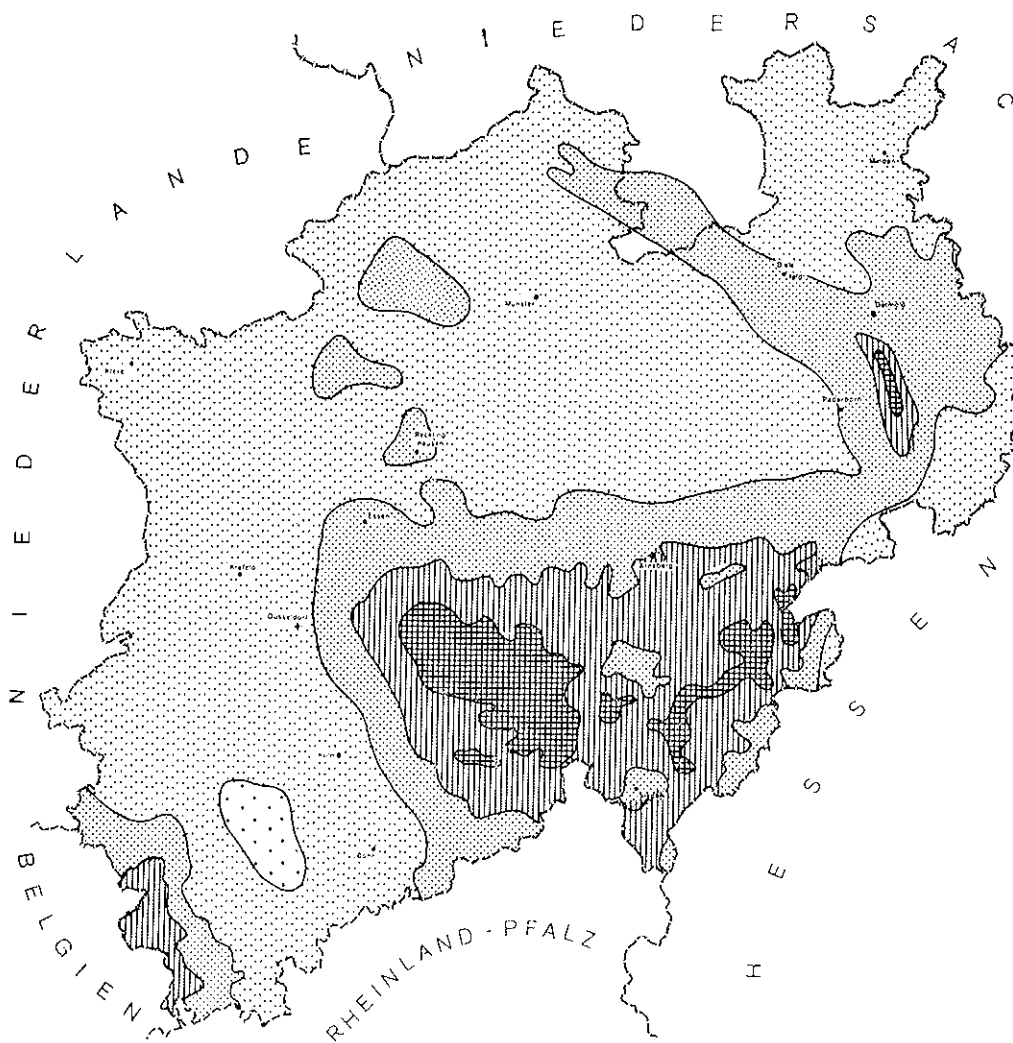


FIG. 7. Mean annual precipitation in mm per year in Northrhine-Westphalia as indication for potential total input of acids by wet deposition. Data from Deutschen Wetterdienst Offenbach for the period from 1931 to 1960. Scale 1:2,000,000.

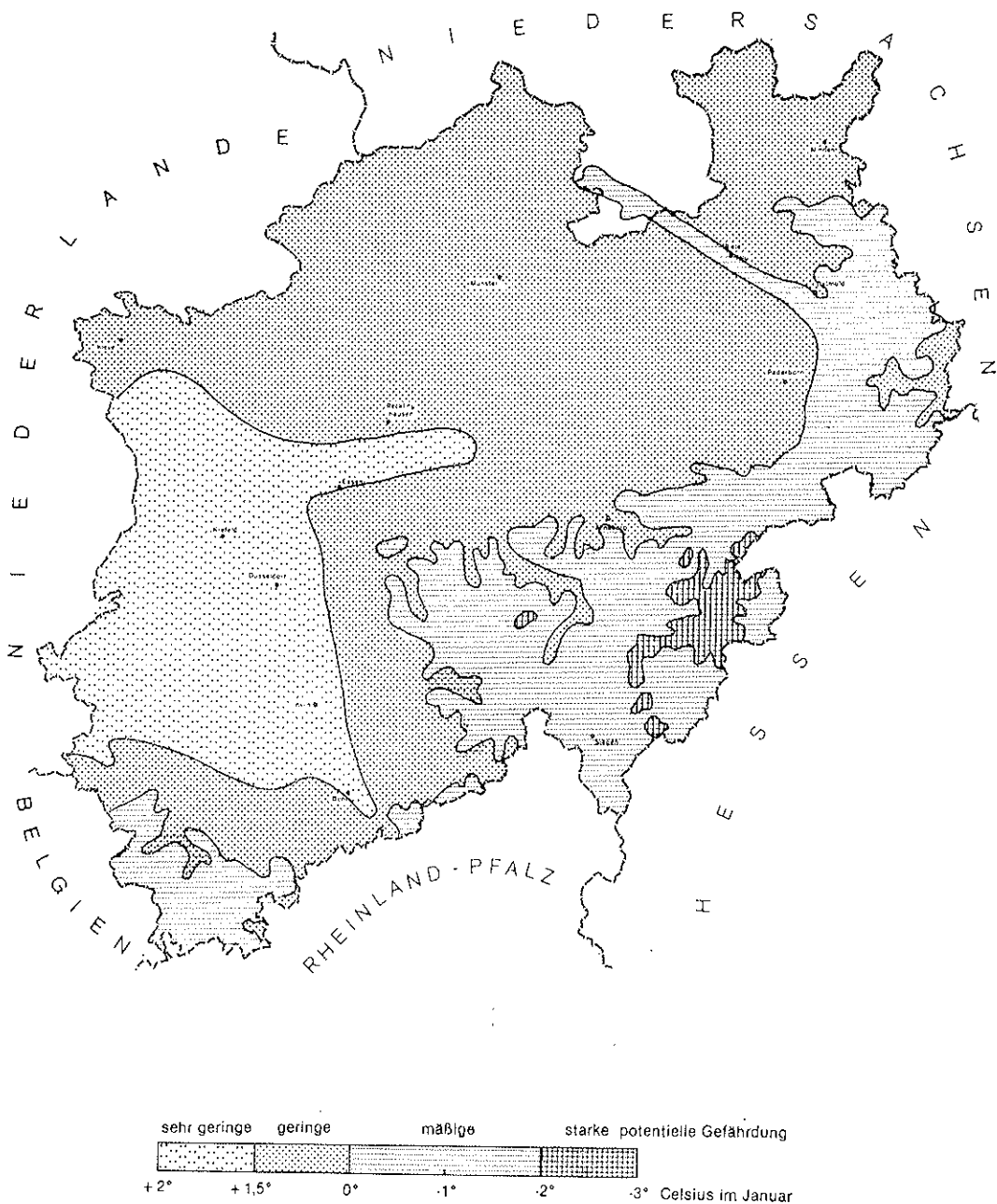


FIG. 8. Mean air temperature in January in °C in Northrhine-Westphalia as indication for potential detrimental combination of frost and air pollution. Data from Deutschen Wetterdienst Offenbach for the period from 1931 to 1960. Scale 1:2,000,000.

or oxidizing fog, containing aggressive radicals, however, which is usually called "smog" is everything else than good for a stand (cf. VDI, 1983). So foggy climate must be regarded as an additional hazard in areas with polluted air.

Fig. 9 shows a map of Northrhine-Westphalia with the number of fog days, differing between valley fog (Talnebel) and low altitude cloud-fog (Wolkenoder Hochnebel). It is obvious that 100 fog days represent a greater hazard than less than 50 days because the prospects of having some fog days with corresponding high pollution levels will be doubled.

#### 4. HIGH STACKS - MAN'S TRICK HOW TO AVOID LIABILITY

High smoke stacks have changed the situation very much. When the effects of sulphur dioxide emissions in the surroundings of metal smelters or coal burning power stations became too deleterious to the biotic environment, man invented the "high stack". At first sight, this solution seemed to be convincing in the assumption that he could produce as much as he wanted without getting complaints from his neighbours. So dilution was considered as being the solution of the pollution problem. The average stack height of power stations in the U.S. built in 1960 was 73 m, but in 1969 already 183 m (TVA, 1970, cited by Munn and Bolin, 1971). Nowadays, stacks of 300 m are not uncommon.

##### *Example 5*

The technical prescriptions for the construction and operation of industrial plants in the Federal Republic of Germany (TA Luft, 1974) still allowed almost unlimited emissions in as much as the impact on the near neighbourhood could be excluded by high chimneys. Thus, the coal-fired power plant of Dinslaken-Voerde could reach emissions values of about 23,500 tons of  $\text{SO}_2$  and almost 10,000 tons of  $\text{NO}_x$  since 1975, whereas the ill-famed smelters in the Sudbury region of Ontario, Canada, even emitted about 1,000.000 tons of  $\text{SO}_2$  per year (Linzon, 1958) which was more than the emission rate of the whole Ruhr district, Germany, in 1947.

##### *Conclusion*

Usually, high chimneys prevent liable operators or sources for damage in distant vegetation from being identified, their license depending on the



maximum and average increase of concentration in  $4 \times 4$  km areas around the industrial plant. The emission being mixed with a greater air mass, the residence time of pollutants in the atmosphere is extended and the possibility of photochemical reactions increased. However, detrimental effects are by no means excluded. They usually occur in greater distance, that is, in areas where vegetation may be more sensitive because of climatic stress or poor nutrient conditions. They also occur via other pathways by soil acidification or in combination with other pollutants from different sources. Being the token of uncontrolled economic growth, high stacks have certainly contributed to the "global air pollution" as described by Munn and Bolin in 1971. From a more general point of view, high stacks have prevented a decentralized economy from being developed and adapted to its environment by favouring large installations and hiding the ill-effects.

## 5. FOREST DIE BACK - A WORLD WIDE PROBLEM

### 5.1. *From fume damage to forest decline: Affected Species*

Forests which have survived all political, economic, and social changes in the industrialized countries of the world during the last centuries are now seriously endangered.

There was a steady development from localized but very severe cases of mortality and growth loss due to oxides of sulfur or to fluoride, first to regional destruction as in the Ruhr district or the "Erzgebirge", later to a statewide and finally continental dimension of forest decline.

The first damages outside the industrial areas were observed on *Silver fir* (*Abies alba* Mill). That species had already become extinct in the Erzgebirge in the Thirties. Forty years later the decline was obvious from Poland to France, that means all over most natural stands and artificial plantations in Europe.

Today the die-back of silver fir is regarded as a real menace to the further existence of this species which reacts more sensitive to dilute concentrations of air pollution than any other tree of forest importance. Its disappearance causes great silvicultural problems because firs help to stabilize mixed forest stands in the mountains against wind fall by rooting much deeper than Norway spruce.

*Norway spruce* (*Picea alba* Karst) was the second species which showed obvious changes in health.

The localized fume damage in the "Erzgebirge" spread to regional die-

back on the top of this mountain ridge between Czechoslovakia and the German Democratic Republic from 1950 to 1980.

In 1979 latent damage could be proved for half of the spruce stands in Northrhine-Westphalia by careful counting of needle retention on felled trees still before the outbreak of clearly visible symptoms appeared in 1982 (Knabe, 1983).

Widespread damage of Norway spruce was observed also in Southern Germany after 1981 and later on in Switzerland and Eastern France while it could be seen in South East Poland also within and outside the industrial region in Upper Silesia. Extended areas were affected in South West Sweden but not as severe as in Central Europe (Scholz, 1984).

There was no clear relation between degree of damage and soil conditions but a positive correlation to wind exposure which leads to a greater flux of air contaminants. High altitudes of central highlands were affected most.

*Red spruce* (*Picea rubens* Sarg.) has shown an unexplained die-back in the coastal mountains of North East America which is supposed to be a result of regional soil acidification caused by air pollution (Tomlinson, 1983).

*Scotch pine* (*Pinus sylvestris* L.), the main forest species on sandy soils or dry sites in Europe did not escape the present decay. The reduction of needle years and needle length and sparse foliage create the impression of thin crowns and sickness. Poor sandy soils in the plains were affected most which might be due to nutrient imbalances because of high input of ammonia from increased life stock.

*Eastern White pine* (*Pinus strobus* L.) and *Ponderosa pine* (*Pinus ponderosa* Dougl.) were affected in Eastern North America and California respectively. The disease was attributed to elevated concentrations of photochemical oxidants and partially of sulfur dioxide (Hepting, 1964; Linzon, 1958).

Deciduous trees which have been regarded as more resistant to air pollution for almost a century show now also symptoms of decline. This was first observed on *European beech* in Southern Germany after 1980, later also an *English oak* in Northrhine-Westphalia which had proved as fairly resistant both to SO<sub>2</sub> and HF. A change in pollution patterns may be the reason, eventually the combination with photochemical oxidants or with ammonia.

*Sugar maple* (*Acer saccharum* Marsh.) has been affected in Ontario, Canada. The reason of this die-back is also not clearly known.

### 5.2. *Present estimation of affected area*

Scholz (1984) has reported on the present extent of forest damage due to air pollution in Europe and North-America for the UN Economic Commission for Europe (ECE). This report is based on official sources of the affected countries. This official estimate of damaged area is listed in table 7.

Most states appear with less than 3 per cent of the forested area damaged. A greater percentage has been reported by the government of Norway and Sweden (5%), Austria (6%), Poland (7%), Federal Republic of Germany (8%), Czechoslovakia (10%), and German Democratic Republic (12%).

These estimations do not reflect the actual situation however. They are much too low for what can be proved by more recent surveys, e.g., in the Federal Republic of Germany.

Table 8 brings a comparison of the surveys from 1982, 1983 and 1984 which shows a dramatic increase of damaged area. The difference between 1982 und 1983 can partly be explained by methodical difficulties in assessing slight damage by untrained personnel in 1982. The surveys of 1983 and 1984 however were made by trained foresters using the same method so that the total increase of 16 per cent of the total forest area in one year must be seen as fact and a very alarming signal.

### 5.3. *Causes for forest decline*

The wide-spread forest decline in Europe and North-America is regarded from scientists of different schools as caused predominantly by the impact of air pollution. "Without anthropogenic air pollution there would not be forest damages to the present extent" (Scholz, 1984).

As outlined in the first chapters air pollution acts in various ways. High concentrations of a single component can lead to the same degree of damage as the combination of two or more pollutants at lower concentrations or as the enforcement of natural stress which would not kill or harm a plant as severely in clean air.

Direct effects of either SO<sub>2</sub>, ozone, HF and HCl have been proved in the history of fume damages research. The steady increase in knowledge is well documented in various reviews. Scurfield (1960), Keller (1964)



TABLE 7 - Statistical data on forest damages in ECE countries (Adapted from "Länderberichte der EFC/FAO, ausgewertet vom Bundesamt für Ernährung und Forstwirtschaft, 1983", Federal Republic of Germany) - Scholz, 1984.

Country	Total forested area		Area of forest damages		Forest damages	
	Land area 1979 1000 ha	1979 1000 ha	% of land area	% of forested area	Regional centres	Main tree species
<i>Northern Europe</i>						
Denmark	4,237	493	11.6	<3	North-western Jutland	Pine
Finland	30,545	23,321	76.3	<3	Local sources	Pine
Norway	30,787	8,330	27.1	<5	South-eastern Norway	Spruce, pine
Sweden	41,162	26,424	64.2	<5	South-western Sweden	Spruce
<i>Western Europe</i>						
France	54,567	14,543	26.7	<3	Lower Seine valley, Lorraine, Rhône valley	Pine, spruce, beech, oak
Ireland	6,889	317	4.6	<3	Local sources	—
United Kingdom	22,820	2,018	8.8	<3	Local sources	—
<i>Southern Europe</i>						
Spain	49,954	15,260	30.5	<3	Local sources	Pine
<i>Central Europe</i>						
Austria	8,273	3,282	39.7	6	Upper Austria, Styria, Tirol	Fir, spruce
Czechoslovakia	12,551	4,535	36.1	10	Ridges of Ore mountains, Iser mountains and Beskydy mountains	Fir, spruce, pine
German Democratic Republic	10,610	2,951	27.8	12	Districts of Cottbus, Halle, Leipzig	Spruce, pine
Germany, Federal Republic of	24,440	7,318	29.9	8	Agglomeration areas, ridges of central highlands, impact areas of western slopes	Fir, spruce, pine, beech, oak
Hungary	9,234	1,594	17.3	<3	Local sources	Black pine
Netherlands	3,395	291	8.6	<3	Local sources	Spruce, fir
Poland	30,456	8,677	28.5	7	Industrial region of Upper Silesia	Coniferous forest, especially pine
Switzerland	3,977	1,052	26.5	<3	Northern cantons, Valais	Fir, spruce, beech, pine
<i>Non-European countries</i>						
United States	912,680	290,760	31.9	<3	North-eastern USA, Los Angeles basin	Pine, yellow pine, Jeffrey pine
Israel	2,033	116	5.7	<3	Local sources, traffic roads	Aleppo-pine, pine species

TABLE 8 - Increase of forest damage in the Federal Republic of Germany from 1982 to 1984 (Adapted from Bundesminister für Ernährung, Landwirtschaft und Forsten, 1984).

Tree species	Area of tree species		Damaged area (damage classes* 1+2+3)		
	Millions ha	per cent of all species	1982	per cent of tree species 1983	1984
Spruce	2.950	40	9	41	51
Pine	1.464	20	5	44	59
Fir	0.179	2	60	75	87
Beech	1.262	17	4	26	50
Oak	0.613	8	4	15	43
Others	0.940	13	4	17	31
All species	7.408	100	8	34	50

\* class 1 = slightly damaged

class 2 = damaged

class 3 = severely damaged

describe the state of knowledge at that time. Knabe (1966, 1972) adds proposals for future forest research which does Robinson (1970) for the long-term effects of air pollution in general.

The textbooks of Garber (1967), Guderian (1977), Dässler (1981) and Halbwachs (1984) summarize the so-called classical pollution effects quite well.

Prinz *et al.* (1982) see ozone as main cause for the forest decline in Germany, however they admit the need for further clarification.

Acidification of soil combined with lack of nutrients and release of aluminium ions which act toxic to plant roots was the other line of explanation. Wieler (1905, 1933), Ulrich *et al.* (1979, 1983 a, b) and Drablos and Tollan (1980) can be used to support this theory, while it is more critically discussed in VDI (1983).

Chlorinated hydrocarbons, radioactive pollutants and other kinds of radiation (like radar), which are also mentioned as possible causes for the present die-back, need still clarification by experimental work.

The chemical industry has declared 35,000 chemicals as being produced before establishing toxicity examination for "new" chemicals. What do we know about the risks of them?

Schütt (1984) gives quite an interesting list of arguments for and against all these hypotheses while Smith (1981) points at the complexity of interactions between air pollution and forest ecosystems which should always be kept in mind.

A tree has only limited ways of response. Any damage of the root system will have an effect to the leaf mass and any reduction of photosynthesis by direct impact of air pollutants to leaves will reversely reduce the number of feeder roots and start a negative feed-back (Bossel *et al.*, 1984).

Natural causes for the far-spread die-back can be ruled out as main force (Waldschäden ... 1983; Schütt, 1984), because the damage occurs on different soils, in the plains as well as in the mountains, but with forest edges and exposed sites suffering more. Sick trees are found in natural mixed forests as well as in even aged monocultures with pronounced injury in older stands. Various tree species are affected which is unlikely for any biotic parasite as fungi, insects or viruses.

From the author's point of view there is no single cause for the present decline except you take the overall input of various chemicals into the system which is by far higher than the buffer capacity of soil and forest. As stated in the "Bundestag" at an expert hearing in October 1983, the present crisis is caused by a varying combination of pollutants with varying weight depending on site, species and climate.

#### 5.4. *Consequences of forest decline*

The consequences of forest decline are numerous and can hardly be fully understood at present. A socioeconomic evaluation has been tried in Acid rain (1984).

We know that the economic role of forests would fade out. Forestry in Europe was based on a balance between annual growth and annual cuttings. This balance is disturbed first; later annual growth will become less and less while cost will increase because a single tree management costs more than clear cuts or periodic thinning and weeds impede re-planting. Hence forest land owners will not get any return, while wood consumers will not get the steady supply which they need. So many jobs will be lost.

We also know that several welfare functions of forests will be affected. Wood covered land has a leveling effect on climate, snow smelt and run off. Bare land shows much greater peaks of wind velocity, run off and in

consequence greater erosion, while dry periods are not balanced any more by retained ground water in mountainous land. Soil erosion can lead to permanent losses in fertility. The eroded soil then will be deposited somewhere, will fill lakes and reservoirs and block water courses. Leached aluminium and heavy metals spoil water quality and endanger human health.

In Alpine areas, where forests had been an indispensable protection against avalanches, villages will have to be evacuated, roads to be closed in winter time which surely will reduce or prevent winter sport.

The recreational value of forests will be affected also, slightly at the beginning, strongly if everybody can see what happens. Such a place does not promise as good a climate as assured in the prospects. This reduction in recreational value will also be felt in and around urbanized areas.

The genetic drain will be enormous. Trees sensitive to air pollution might contain genes with great importance for the population in relation to its ability to cope with other stress factors. The loss of whole species cannot be excluded as can be seen in the increasing extinction of Silver fir in various forest areas. The loss of flowers, birds, insects and other living beings takes away what has been here before man appeared.

This brings us to think about further consequences. Does not mankind just represent one single species? Why should this species escape the bad fortune it has brought to others? Biotic systems of such high stability as forests need really a heavy chemical attack to be killed. The same chemicals enter our lungs, nutrients and water and will cause long-term changes in our body or genes which we do not notice in time. The increasing numbers of cancer in various organs all over the world speak their own language.

Or if we look from another direction. Was not all this nature delivered to man by the Lord to be cared for and well treated? Its rude exploitation, its reckless destruction was not told to man. So Christians should be especially aware of what is going on and do their best to come out of this situation.

## CONCLUSION - WHAT CAN BE DONE?

The present forest decline cannot be explained by natural factors but has to be seen as the result of increasing chemical impact. These chemicals are produced by burning fossil fuels, intentionally brought into the environment as pest control e.g., released as by-product, or left as waste somewhere.

So what can be done?

This is not the place to give detailed answers. Only some fundamental thoughts will be pointed out.

(1) Everybody who reads this paper should recognize his own responsibility. So do not read it or do something! Our actions will depend on our position in life.

(2) Everybody is a consumer, so change in consuming habits from "wastemakers" to thoughtful housekeepers who re-use goods or take care for their duration will reduce the use of energy and the release of chemicals to the environment. He can insulate his home, buy renewable goods, eat less meat and reduce driving cars or stay within speed limits.

(3) Almost everybody is a citizen with rights and duties. So he can influence the politicians of his district, political parties, authorities and press in his region or even at higher levels. We should use this power in favour of nature conservation.

(4) Some are teachers, priests, nurses or other educating people. Many of us have children who are observing our behaviour. Setting standards with our own life can influence them. A tender care for flowers, animals or the green in our surrounding will certainly have an effect on our own life and on theirs too. Informing them on the interrelations between man and nature, between chemical input and biologic response is the second way of changing the situation. Only people aware of the situation can act. So one has to learn more and more.

(5) Some of us are engineers, technicians, chemists or inventors. Many of us work in a factory, a bank or an administration. There are innumerable occasions to do something for the environment in our jobs, much more than we have thought before understanding the problem ourselves. It is time to make use of these possibilities.

(6) Only a small minority does belong to the leading circles in administration, government, church, industry, army, banks, media etc. At present they are asked to use their influence towards a better understanding of natural cycles, aiming at a survival programme for our forests as well as for ourselves.

(7) We should be well aware that the present arms race is the greatest waste of raw materials, energy, labour and intelligence we can think of, as was outlined by Pope John Paul in November 1983 analogously. This intelligence would be urgently needed to solve the real problems: hunger, social development and environmental protection.

## SUMMARY

Effects of air pollutants on vegetation, especially forests.

Air pollutants are affecting terrestrial vegetation in various ways. Direct effect of gaseous pollutants as  $\text{SO}_2$  are well known since the middle of the last century. Other phytotoxic gases as HF, HCl were detected later, photochemical oxidants not before the middle of this century. Recent findings revealed synergism between different gases as  $\text{SO}_2$  and  $\text{NO}_2$  or ozone which causes much greater effects than originally expected.

Acid mist and dew is another cause for damage to vegetation. Research has been intensified over the last years to a great degree.

The mechanism of dry deposition of acid gases on plant surfaces and their results on plants are hard to simulate and are not fully understood.

Indirect effects of air pollution on vegetation result either from a general weakening or preconditioning of plants which make them a better host for pathogenes or parasites or reduce their resistance against frost und drought. Examples are reported.

Or they are the result of changes in soil chemistry and texture.

Acids may leach nutrients and make aluminium and iron or other heavy metals soluble, so that roots find poorer growth conditions or die.

The present decay of forests in parts of North America and in whole middle Europe which is extending to France, Eastern Europe and South Sweden is regarded as a very alarming signal to mankind to change its living conditions in a way which will not cause such detrimental effects.

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## DISCUSSION

MARINI-BETTÒLO

Thank you very much, Dr. Knabe. It is very interesting and this is quite clear evidence of the impact of the chemical events in the atmosphere on the biosphere. I would like to ask Dr. Knabe if he has seen a recent report in *Environmental Science and Technology* saying that it is not always possible to correlate air pollution in general with forest decay.

KNABE

I have not seen that special publication but there are similar publications or similar arguments. The problem is not so easy. First you have to take into account the accumulation of air pollutants within the forest ecosystem. This accumulation occurs either in the soil by a steady increase of acidity and heavy metals or within living beings themselves. Increase of acidity means the loss of other nutrients, of basic cations like calcium, magnesium, potassium. Accumulation of acidity can also occur in the form of aluminum ions attached to the buffering soil colloids as a result of the disintegration of clay minerals, if the soil pH in water falls below 4.2. Below pH 3.8 also iron becomes soluble and is then adsorbed by the surface of clay minerals and other soil colloids, too. That is one topic.

Another topic: heavy metals are accumulating also within trees, within roots, within needles and other parts of the stem; so you have a constant weakening of the tree. You cannot relate this content to the present ambient concentration of the pollutant in the air, because it has been accumulated in the past. However ambient concentrations of ozone and SO<sub>2</sub> and NO<sub>2</sub> may also affect the tree and make the effect even worse. Let us take an example: plants have the ability to detoxify certain agents, like SO<sub>2</sub>; they can reduce it and metabolize it within the cell, and they can also deal with oxidants, like ozone. Well, ozone is breaking off double bonds within certain enzymes and other cell constituents, but it can be metabolized afterwards. For doing this, however, energy is necessary, and if energy production — that means photosynthesis — is reduced and the tree is missing the energy to do it, then this double impact may be too much for it. So if a scientist wants to correlate ambient concentrations to decay of forests, he should precisely distinguish

between very different cases. We can prove — I still have some slides which could be used as an example — that upper soil layers have reached a level of acidity at which any root growth is prevented, so that no root grows into that special layer. We can prove that the leaves are hurt at certain concentrations of sulphur dioxide or hydrogen fluoride, so that photosynthesis will be impossible or the leaves are dropped or burnt. And we can prove that there is a concentration of soluble heavy metals in the soil at which those metals will penetrate into the roots and cause damage there. We cannot explain, however, the sudden outbreak, the sudden widespread forest damage at present — that we cannot fully explain.

#### LIBERTI

I only wish to express appreciation for the fine presentation of the changes in vegetation that you have been able to show us. However, I want to make a couple of comments on your lecture, according to the following lines. The first one is that you described two procedures for the approach of this system: one starting from effects and going back to the causes, and the other one talking about causes and going to the effects. In your lecture you practically followed the first line, to show the effects and to describe the causes which might be responsible for them. Now my criticism is the following: I am wondering if you made any effort to make any correlation between ground concentration of various pollutants and the damages that you have been able to find in the plants. This is quite an important point, because in the presentation of your lecture you were talking about the effect of doses. When we are taking a plant, also if the plant is exposed to a serious pollutant because it stays always in the same position, it will get a certain dose. This is important because we are able to correlate the effect on doses. And I am insisting on this point because from a biological standpoint public administrators and technical people are trying to establish some A quality standard in terms of various pollutants. For instance, these days we are having a standard for  $\text{SO}_2$ , a standard for particulate matter, a standard for nitrous oxide, etc. Now if we can correlate doses and effect, we might be in a certain position to say that the data which have been established by public health people are valid as far as air is concerned, but not for the environment, because plants are more sensitive to this effect. I really would be very much pleased to have your comments on this point.

Another question I would like to ask you is the following. You show us some of your results in terms of concentration of various pollutants on the

plants, and you show us the difference between vegetable species which have not been exposed. Now these days we are very much worried as to whether most of the damage is coming through a reaction which takes place in the cells of the various species or is something affecting the adsorption of these species. For instance, in one of your pictures you showed the effect of fluoride. Now, being a chemist myself, I am always very much worried about the fluoride business, because we may have fluoride in the gaseous phase as hydrogen fluoride, which we believe is toxic. However, we may have fluoride as inorganic particles, which practically will belong to particulate matter, and according to our knowledge we believe these are practically harmless. So I believe that these days we have to go more deeply into this effect just because we have to be able as scientists to supply some information to technical people. And I do not think there is too much sense in talking about fluoride as such because we have gases, particles or fluorides in aerosolic species. I certainly will appreciate your comment on my questions.

KNABE

Thank you very much, Professor Liberti, for the questions. First, there are measurements between the ambient concentration of pollutants and the growth of vegetation. I have not made those studies myself, but there are such studies. There are comparisons between several locations in the northern hemisphere, where they have measured the concentration of air pollution and the growth of vegetation, and they found that the production and the growth rates decline with an increase of sulphur dioxide and an increase of hydrogen fluoride. Those figures could be obtained from the report made by the state institute for air pollution control at Essen. There was also found a correlation between the content of heavy metals in the atmospheric suspended matter and the content of heavy metals in the plants which have originally been precipitated on the surface. Well it is different — you can wash it off the surface partly, but you cannot wash it off totally. Cadmium shows a greater mobility, it is also penetrating the cuticula, going into the leaf, whereas lead is staying more or less on the surface.

LIBERTI

Sorry if I interrupt you, your answer is quite satisfactory. However, can it be expressed in numerical terms, because at the beginning you were talking about doses. Now it would be terrifically interesting to know this data because

the great effort which is made on an experimental basis is trying to establish what are the limits for different pollutants. In other words as a general attitude we do not like to stop industrial activities and we do not wish, in the defense of the environment, to make a limitation in various countries in any activities.

The aim that we wish to achieve is a certain regulation of production so that man's activities, and also living conditions of the society, can go forward in a very healthy way. Now in order to do this, we have to try to get figures which may be considered standard emission rates. It means the amount of toxic material which may be emitted into the atmosphere. Owing to the average turbulence situation we may have viable conditions so that the plant life can go ahead quite well. What worries me is the fact that with the benefit of the World Health Organization in Geneva certain standard values have been established so that we believe, for instance, that a certain concentration of  $\text{SO}_2$  in a certain range is said not to affect human life. My question is the following — I do not know if you are able to answer it, it is certainly a big wish of people engaged in this problem — is it possible to establish a certain limit for concentration of pollutants compatible with human health without causing damages to forests and the environment? In order to do this, we have to be able, to a certain extent, to establish a minimum dose on which a certain plant can flourish and can go ahead. I therefore would like to have your comment on the possibility of achieving this aim.

KNABE

I can give you two sets of figures. One was set up by the Association of German Engineers (VDI) — their committees have worked on these standards — and the other was established by IUFRO, International Union of Forest Research Organizations. Let us begin with hydrogen fluoride as you mentioned, and which has to be distinguished from other fluorides. In my paper I gave only the fluoride content of leaves or needles and so I could not say if it had been hydrogen fluoride (HF) originally, so I could just measure the content in the needles.

But if you come to ambient concentrations, we have a set of 0.3 microgram per cubic meter ( $\mu\text{g}/\text{m}^3$ ) HF as annual mean for very sensitive plants, 0.5  $\mu\text{g}/\text{m}^3$  HF for sensitive plants, and 1.4  $\mu\text{g}/\text{m}^3$  HF for less sensitive plants. For sulphur dioxide the three groups would require annual means of 60  $\mu\text{g}/\text{m}^3$ , 90  $\mu\text{g}/\text{m}^3$  and 130  $\mu\text{g}/\text{m}^3$   $\text{SO}_2$  and for hydrogen chloride 100  $\mu\text{g}/\text{m}^3$  HCl for very sensitive plants and 150  $\mu\text{g}/\text{m}^3$  HCl for sensitive plants. We are still missing a value for less sensitive plants. For nitrogen dioxide there was only

the annual mean for sensitive plants,  $350 \mu\text{g}/\text{m}^3$   $\text{NO}_2$ . The last figure might be outdated because people in the Netherlands (Jan Mooi) and also somewhere in America found a very strong synergism between  $\text{NO}_2$  and  $\text{SO}_2$ , and  $\text{NO}_2$  and ozone. So the value for  $\text{NO}_2$  might be much too high. The VDI has also established limitations to peak concentrations which are not listed here. The IUFRO standards for the protection of forests concern the following values as sufficient:  $3 \mu\text{g}/\text{m}^3$  hydrogen fluoride as annual mean, and  $0.9 \mu\text{g}/\text{m}^3$  as 97.5 percentile of all measurement values usually means over 30 minutes. A 97.5 percentile means that only 2.5% of the individual values should exceed this limit. The corresponding values for sulphur dioxide would be the annual average of  $50 \mu\text{g}/\text{m}^3$   $\text{SO}_2$  for normal conditions, and  $25 \mu\text{g}/\text{m}^3$   $\text{SO}_2$  for conditions with very poor sites or high mountains where climatic stresses are included, so very low values, and they are derived from the results we have found in Austria and the mountains in Czechoslovakia.

LIBERTI

Are the figures that you give us accepted by the German Government, or have they been only the results of your experience? There is a strong need for values which may be internationally agreed, which might be a guide for people anywhere. And this is quite an important point, because the evolution of living conditions is sometimes a critical political standpoint in specific attitudes by various governments. Now it would be necessary to point out the necessity of having emission standards which may be accepted and which might be useful for anybody by taking into account not only air conditions but also living conditions. I am just asking your cooperation, also to define if these figures have been issued on a local basis or government basis or an experimental basis.

KNABF

Well, I could only offer you to fix all the figures here at this round table. We could include some proposals for the outcome of the meeting or we could sit together and make some detailed proposals. I think the discussion is not the place of very detailed figures to solve this problem. However, IUFRO-values ought to be respected, because IUFRO represents all forest research organizations of the world which work on the problem of air pollution and its effects on forests.

LIBERTI

I agree with your proposal.

LAG

May I ask do your analytical figures, for instance of heavy metals, refer to a dry matter basis?

KNABE

They all refer to a dry matter basis.

BROSSET

I have a small remark on what Professor Liberti has said. I share totally his opinion, but the example he gave, I think, was a little unfortunate because if we have sodium fluorides in aerosols they may not be quite harmless, because the hydrofluoric acid has a pH of 3, and if we have a solution, for instance, of pH 4, 10% of the water-soluble fluorides will be in the form of hydrofluoric acid, so that will be still a harm risk.

LIBERTI

I was pointing out the difficulty of defining what is the fluoride in the air, because we have gaseous fluoride, liquid fluoride and particles fluoride; therefore usually we are always hearing about fluoride concentration, which in my opinion does not mean too much because the gaseous is definitely a more important fact than are the particles. So I agree fully with what you say, but my statement was just in the desire to learn more about a problem which is of the greatest importance.

WIESENFELD

The very first speaker at this Study Week, Professor Phillips, mentioned the goal of a government-sponsored study in the U.K., explicitly leaving out — saying these goals were specifically left out — some of the more interesting (to chemists and physicists) aspects of the problem; and those include those phenomena which can be referred to as the source-receptive relationship in acid precipitation, acid deposition. Specifically I think it is important to



remember that one, in assessing the problem, needs extraordinarily accurate emissions inventory data. For example, if one looks at the power generation in the United States over the last 20 years and attempts to draw conclusions from that, I think that one would be led down the wrong path, for while that power utilization has gone up as a result of presumably increased population, the actual amount of emissions has gone down because of legislation. Similar observations, I think, would also be true concerning nitrogen oxide emission. I think we have to look very carefully not only at temporal data but also at spatially resolved data, and that increasingly is becoming available although the collection of that data is an extraordinarily complex and expensive proposition.

Secondly I think one has to look very carefully at transport phenomena. Several speakers in this Study Week have referred to the effect of the introduction of tall stacks which permit an increased dilution of emissions from power plants. The inclusion of a tall stack at a power plant does not cut the deposition in the vicinity of that stack — and you can use the term vicinity in any way you like — to zero and the deposition at a distance of 500 kilometers to 100%. A very substantial fraction of the deposition associated with emission from a particular source, even with the tall stack, occurs relatively near to that source; and again, the question of how near is one, that is extremely difficult to get solid answers for — one simply does not have that kind of information. If you want to take an applied mathematician and send him into shock, you ask him to model the flow from a tall smoke-stack over a distance of about ten or twenty kilometers — that is just something that is not done. The overall transport phenomenon in acid rain is an extraordinarily complex multi-scale phenomenon. So far as the chemistry is concerned, I think that, too, varies dramatically with the climate, with the season, with the presence of oxidants of various types in the atmosphere, and I fear that there can be no single generalization concerning acid rain and the mitigation of acid deposition worldwide. This is something that has to be handled on a scale which in fact I think will be much smaller.

#### CRUTZEN

What I have missed so far in the discussion, is the absolute lack of discussion of meteorological phenomena, and Dr. Wiesenfeld's insertion here was very well placed. After all, the last two years in Germany, which have caused a definite decline in forest resources, were also combined with some very exceptional two years of summer weather. The spring was far too wet and

the summers were far too hot and far too dry, and I think the meteorological aspects of forest death should definitely not be neglected. It is not only a problem of chemical constituents in the leaves which we should consider but the whole physiology of the forest. I think even people, when they do not drink, are in big trouble very quickly, and so why should forests not be? Now, on top of that, the whole pattern of deposition of dangerous or haphazardous chemicals will be changed from wet deposition to dry deposition, and the whole chemistry and the intermediates in the oxidation schemes in the atmosphere will be totally changed under these different climatic conditions. I think this is a very important issue to pursue in the future and one should look at what compounds are really being deposited on the forest.

#### KNABE

I should like to answer this by showing part of what we have done. We tried to determine the amount of the potential hazards to forests in Northrhine-Westphalia by a combination of certain factors. One of these combinations would be total acidic deposition as a result of acid concentration and the amount of rainfall. The mountaineous areas usually get higher rainfall than the plains.

Here the potential input of acid may be greater than in the industrial areas in the plains. We have already proved this hypothesis by measurements from 1973 to 75. We have measured the acid input both in the industrial areas of the Ruhr and in two rural districts. We found differences. We found that the titrated acidity, total acidity, in Mülheim-Ruhr amounted to about  $120 \text{ mgH}^+ \cdot \text{m}^{-2} \cdot \text{a}^{-1}$ , 60 kilometers north about the same amount, and 50 km south of the Ruhr, with high rainfall,  $155 \text{ mgH}^+ \cdot \text{m}^{-2} \cdot \text{a}^{-1}$ . But there is also a great difference between rain outside the forest and below spruce. In the fall of Norway spruce you find about 4 times or 3 times the amount of acidity deposited measured outside the forest. At the edge of the forest we have again an addition. We find a higher acid deposition at the edge of the forest than in the middle of the forest. And if you do the same for the Federal Republic of Germany, you find areas with higher rainfall and those areas are partly very heavily attacked by the present forest decay. In the southern part of the Bavarian mountains, in the Harz mountains or in the area of the Black forest, you have higher rainfall and by this a greater impact. In another region with high rainfall, in the Alpine mountains, there are calcareous soils, which can neutralize the acids, and so the effect has not been as great, but now it is also appearing.

CRUTZEN

Are these average numbers, or are these the numbers for the last two years?

KNABE

These figures are average numbers, but you can do the same work for a more recent period.

CRUTZEN

You should take the last two years and compare it with them.

KNABE

Pardon me, you can make a map of the average over many years and you can make another map with the input of rainfall maybe in 1982.

CRUTZEN

But I did not see that map. I see the averages.

KNABE

But this is actually a matter of the meteorologists and not my task as forest scientist. It is their business to handle the climatic maps. We try to make use of them. I only came to show you the possible combination between the climatic effect, and you were asking for that. The other combination is the combination between cold temperature and emissions. The mountains generally are the coldest area in winter time and the industrial zone is the warmest area. And so the potential hazard by combination of air pollution and winter cold is greatest in the mountains and is the least in the Ruhr district.

The third combination to be mentioned is that of fog and air pollution. These areas with high frequency of fog again lie in those mountains where we have the strongest forest decay. So climate has to be taken into account, and I agree with you also that special climatic events in the last years may have affected the trees. But this research requires more time and man-power, I can only show the importance of the problem and hope that some other group will start to work on it.

## LIBERTI

Dr. Knabe, I believe that the suggestion given by Professor Wiesenfeld would be just the best, because if you follow the same system which was used by Phillips the other day, it is quite useful to present your data in terms of the emission inventory, which is already quite an approximate way of presenting the data, but it is still the figures which are more reliable. If you are setting up the data which you present to us in terms of the emission inventory in terms either of  $\text{SO}_2$  emission or of nitrogen oxide emission, you may have a certain correlation, unless you have the emission data, which are difficult to be obtained. But since we know — and this is something on which most of the chemists agree — the mechanism of oxidation of most pollutants — if we have an emission inventory, we really are able to correlate in a better way the damages we have to forestry and to vegetation in general, with the effect of pollution. Unless we have these data, it is very difficult to make a sincere and correct appraisal of the damages that we are receiving.

## KNABE

I cannot agree with this proposal. There are a number of links between the source or cause and the effect, a whole range of stages as emission, transportation, immission, deposition and dose, and they are influenced by climate, site, genetic resistance, which can alter the picture. In summary, the effect is very far apart from the source. If you measure the dose within the plant, the amount of pollutants which the plant has taken up, or the state of the soil as it has changed by air pollution, then you have a much closer relation to the effect than just by an emission survey. We have now the forest decay just in those areas which are maybe 100 kilometers away from the industrial areas and not in the industrial areas themselves. So this proposal would not work. It is, however, very useful to find out the main sources and to start measures to reduce emissions in general or for certain pollutants.

## LIBERTI

If you have emission data, you are really able to calculate the doses, because the only variable that you are meeting is the meteorological conditions. So this is a rough appraisal, but it is absolutely the best way to correlate a certain damage if this damage is to be attributed to pollution.

## PHILLIPS

May I just make a comment in reply to Professor Wiesenfeld, and without wishing to act as an apologist for the Royal Society and the Swedish Academy of Sciences and the Norwegian Academy of Sciences? The justification given for the particular choice of survey to be carried out was, I think, not being cynical economic, that was restricted in terms of economics. Perhaps I can read the reasons: This program will not be concerned with the complex processes of how industrial discharges create acid rain, nor with the degree to which productions in the United Kingdom sulphur dioxide emissions would change the acidity of rainfall or have affected those areas of Norway and Sweden. These issues are already the subject of much intensive theoretical and experimental research in many countries. I think that is probably true. So the main thrust then is the effect *in situ*. A decade ago it was considered that high acidity alone occurred either at peak values or when the snows melt or in the less severe values but over a prolonged period. This gives an explanation for the disappearance of trout in certain Scandinavian waters — you see the very specific question they are addressing. Since that time, however, it has been shown that trout can be more tolerant to acidity than was earlier thought, and current explanations relate that disappearance to the effects of acid rain and leeching aluminium into surface waters. Our present understanding does not allow us to predict the precise relationship between levels of acid rain and the composition of surface water, or its acidity after interaction with minerals in the soil or the balance of mineral elements after this process, which relates very much to things we have heard today. So they really have not addressed the question of effects on forests. It is really the fishlessness which they are addressing, and it is a very small part of what I hope, like you, will be a much wider program. I mean I personally would have liked to see the first question and the fifth question on my list of Monday being addressed, but it is for economic reasons that it is not being addressed in this way.

## KNABE

Well, may I answer a little better your question? Again, it is surely useful to have a survey of emissions; it is very very useful to reduce emissions there where the greatest emission occurs. That would be very useful. But for the direct relationship between effect and emissions I have doubts.

## CHAMEIDES

Professor Knabe is sort of saying that we need to worry about the effects (bottom of the slide); Professor Liberti is saying that we need to worry about emissions (the top of the slide). It seems fairly clear that what we are to do is to talk about all the different parts of that slide and to understand not just pollution but the link between atmospheric chemistry and the biosphere. We need to understand all of those prophecies and I think we can agree that we do not understand how all those things connect up — we have some suspicion, and we certainly should be able to agree that in the future we need to try to work together to understand from the top to the bottom and from the bottom to the top.