Vegetatio **101**: 55–63, 1992.

Are there seasonal variations of trace element concentrations (Cd, Pb, Zn) in wood of *Fagus* trees in Germany?*

J. Hagemeyer, A. Lülfsmann, M. Perk & S.-W. Breckle
Bielefeld University, Faculty of Biology, Dept of Ecology, P.O. Box 100 131, D-4800 Bielefeld 1, Germany

Accepted 19.12.1991

Keywords: Biomonitoring, Dendroanalysis, *Fagus sylvatica*, Tree rings, Xylem

Abstract

Concentrations of Cd, Pb and Zn were determined in stem wood of beech trees (*Fagus sylvatica* L.) from 3 sites in northern Germany. Distinct radial distribution patterns of the elements were observed in the xylem. Concentrations of Cd and Pb increased from the youngest, outermost annual rings towards the center of the stem. With Zn intermediate concentrations were observed in the sapwood and higher levels at the center of the stem.

Temporal and spatial stability of such distribution patterns in the trunks was investigated. Wood samples taken from the same individual tree in different months of the year were analysed. Marked seasonal variations of mineral concentrations were observed. Also the shape of the distribution patterns of the elements varied with the season. Such variations were larger than those observed with samples taken simultaneously from different sides of the trunk. Furthermore, Pb concentrations in the stem showed variations with height above ground.

The results indicate, that radial distribution patterns of Cd, Pb and Zn in xylem rings of beech are not stable. Biomonitoring trace element pollution levels by analysis of beech wood is, thus, questionable. To obtain a reliable historical record of pollution from tree rings, the distribution patterns should be stable over a long period of time. This basic requirement of the dendroanalytical method does not hold for the examined beech. Still, with other tree species and under more favourable conditions the dendroanalytical biomonitoring method may prove valuable.

Introduction

Concentration levels and distribution patterns of potentially toxic trace elements, e.g. Cd, Pb and Zn, in wood of trees have been investigated repeatedly during the last three decades (Martin & Coughtrey 1982; Burton 1985; Hagemeyer 1986; Lukaszewski *et al.* 1988). In many studies attempts were made to use mineral concentrations determined in growth rings of different age as a means of biomonitoring heavy metal pollution.

Trees of temperate regions usually form clearly visible annual growth rings, which can be dated accurately over a long period of time. Thus, the idea is tempting to extract wood samples of different age from a tree trunk and to analyse the material for its heavy metal content in order to get

* Presented as symposium paper at the V International Congress of Ecology, Yokohama, August 23–30, 1990.

a chronological record of varying trace element pollution patterns in the environment of the tree. This method was named dendroanalysis (Tout & Gilboy 1978).

Though the feasibility of the method has never been shown beyond doubt, a large number of investigations was published. In some studies correlations were reported between patterns of tree ring heavy metal contents and temporal records of pollution from other sources, e.g. data of industrial or traffic activities (Sheppard & Funk 1975; Lepp 1976; Valkovic *et al.* 1979; Baes & Ragsdale 1981; Wickern & Breckle 1983; Meisch *et al.* 1986; Ilgen & Nebe 1989). In trees alongside a busy road in New Zealand Ward *et al.* (1974) found a certain coincidence between variations in traffic densities and Pb contents of annual growth rings, apparently originating from car exhaust fumes. In Japan, Suzuki (1975) was able to construct a rough time record of the pollution history near a zinc refinery by analysing wood of *Cryptomeria* trees.

On the other hand, various studies were reported in which the attempted heavy metal pollution biomonitoring from tree rings failed (Szopa *et al.* 1973; Barnes *et al.* 1976; Hagemeyer & Breckle 1986).

Thus, an investigation was carried out to check one of the basic assumptions of the dendroanalytical method, i.e. the stability of the mineral distribution patterns. If a reliable pollution history is to be obtained from tree rings, mineral contents and distribution patterns of the elements should be stable in the trunk wood for a long period of time. This means, that between the incorporation of the metal during the formation of xylem rings and the sampling and chemical analysis, perhaps decades or even centuries later, no significant movements of the analysed minerals should occur.

In order to elucidate the stability of elemental distribution patterns, beech wood from different places in Germany was sampled and analysed for its Cd, Pb and Zn contents. Samples were extracted from the same tree several times during the course of a year. Additionally, wood samples were taken from different sides of the stem. Thus, seasonal as well as spatial variations in the mineral contents of tree rings were investigated.

Materials and methods

Localities and trees. Wood samples of mature beech (*Fagus sylvatica* L.) were collected from 3 sites in northern Germany. One tree was chosen in the vicinity of Bielefeld, on the southern slope of the Teutoburger Wald. Trace element pollution in this area is low to moderate. The other trees were growing in the Eifel area, one in Lammersdorf and the other one in Stolberg. In Lammersdorf soil samples revealed somewhat elevated trace element levels as compared to Bielefeld. In Stolberg high levels of heavy metals occur in the environment, originating from geogenic and anthropogenic sources. At each site a larger collective of beech and spruce was sampled and examined (Hagemeyer & Breckle 1992). Only a small, though representative part of the data is given here.

Sampling procedures. From the trunks of live beech wood samples were extracted horizontally with an increment borer (30 cm long, 0.5 cm core diameter, Model Suunto, teflon-coated) at about 1.5 m above ground. Cores were taken either as 4 replicates simultaneously from the same direction, all in a vertical line with ca. 2 cm distance in between or at the same time from 4 different directions. Some beech were sampled repeatedly between April 1988 and January 1989. On the later sampling dates cores were extracted about 20 cm above or below the holes of the previous borings and in about 10 cm distance in horizontal direction. This precaution was necessary to avoid the reaction zone in the stem, caused by wound effects of earlier borings. All holes in the trunks were sealed with wooden plugs and wax to minimize damage to the tree. Furthermore, before use the borer was disinfected in isopropanol. Sample cores were instantly frozen in liquid nitrogen and freeze dried in the laboratory in order to avoid lateral movement of dissolved minerals during the processing of the material. The cores were then divided into portions of 5 or 10 annual growth rings, which were used for analysis.

At the end of the sampling period trees were felled and stem sections cut. On such cross sections from different heights above ground wood samples were obtained with a mechanical drill. These samples were taken from xylem rings of different age along a radius on the stem section.

Chemical analyses. Wood samples of increment cores as well as drilled chips were digested under pressure with conc. HNO_3. Mineral concentrations were determined by atomic absorption spectrophotometry (Perkin-Elmer 5100, Überlingen, F.R.G.): Cd and Pb with a graphite furnace and Zn by the flame technique.

Statistics. For comparisons of radial distribution patterns of minerals the Wilcoxon matched pairs signed rank test was used (Sachs 1984).

Results

The radial patterns of Cd, Pb and Zn in trunk wood of a beech tree in Bielefeld show characteristic distributions of the elements (Fig. 1). With Cd and Pb lowest concentrations are found in the outermost and youngest annual rings. In the older wood concentrations increase steadily to peak values in the heartwood near the center of the stem. In beech of this age the transition from sapwood to heartwood is supposed to occur some 40 to 60 xylem rings inside the cambium (Hagemeyer & Breckle 1992). The Zn distribution is different. In the sapwood Zn concentrations show only little variation, whereas in the heartwood they increase towards the center of the trunk.

Such analytical data plotted along the time scale of wood formation may possibly display a chronological record of metals in the wood. The question is: what do these patterns tell us? Do they convey any information about heavy metal pollution in the past?

To tackle this problem the temporal stability of the distribution patterns was examined in beech from the Eifel area (Figs. 2, 3). Beech trees were

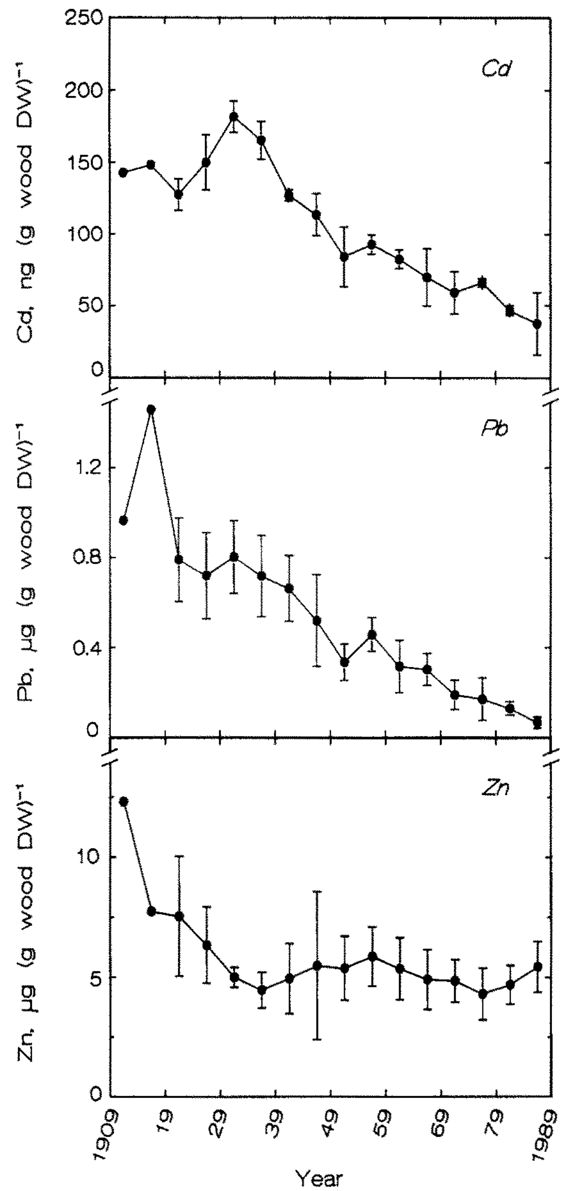

Fig. 1. Radial distribution of Cd, Pb and Zn in stem wood of a beech tree in Bielefeld in April 1989. Sampling height 1.5 m. Means ± s of 4 wood cores from the same direction. The 2 data points near the center are derived from only 2 or 1 of the cores, resp.

sampled repeatedly during and after the vegetation period and element distribution patterns were determined. For Cd the radial distribution shows marked variations with time (Fig. 2). In April lowest concentrations are found, followed by highest contents in June and intermediate levels

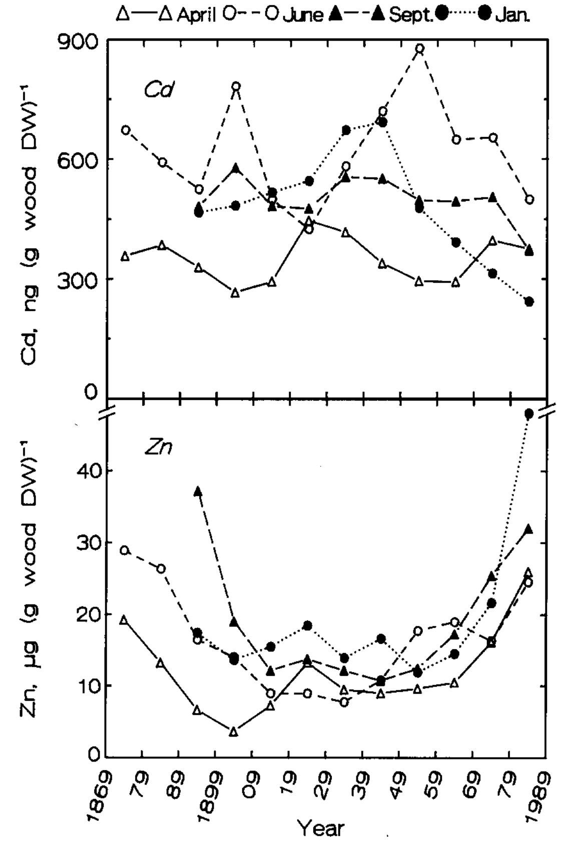

Fig. 2. Radial distribution of Cd and Zn in wood samples of a beech tree in Stolberg, taken between April 1988 and January 1989. Sampling height 1.5 m.

in September and the following January. The April values are significantly smaller than those of the samples taken in June or in September ($P<0.01$). The observed variations do not only concern concentration levels, but also the shape of the radial patterns. The Zn distribution in wood of the same beech apparently is much more stable in time, although some smaller variations are observed, as well (Fig. 2). Zink values determined in April are significantly different from those obtained in September or the next January ($P<0.01$).

The Pb distribution pattern in the same tree from Stolberg has some similarity to that of Cd (Fig. 3a). On the whole, in this tree Pb concentrations reach a maximum in the sapwood. The shape of this pattern is not stable. Lowest contents are observed in April and, again, highest in

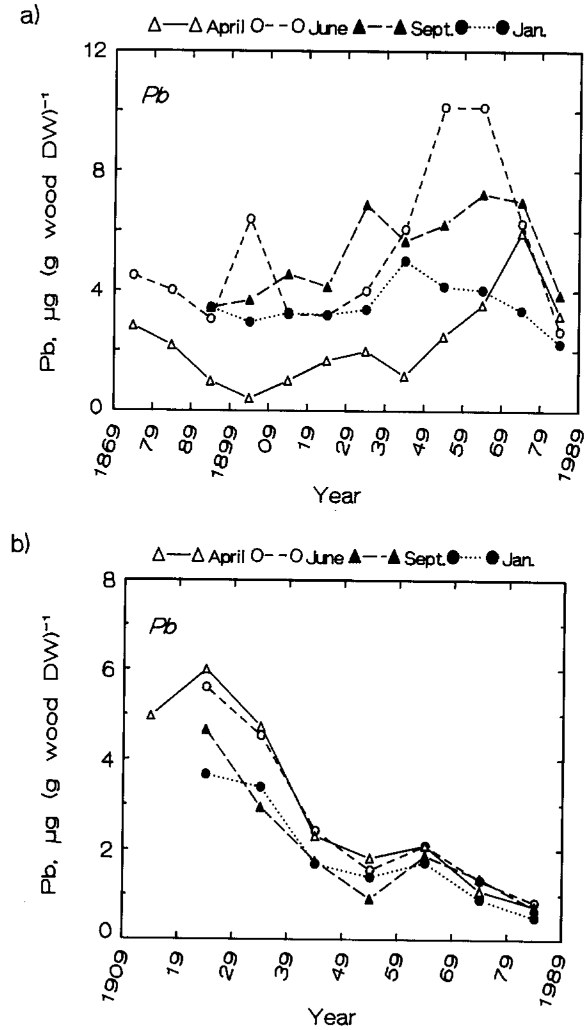

Fig. 3. Radial distribution of Pb in wood samples of beech trees in a) Stolberg (same tree as in Fig. 2) and b) Lammersdorf, taken between April '88 and January '89. Sampling height 1.5 m.

June. The difference between these two patterns is significant ($P<0.01$). As already described for Cd, also with Pb intermediate levels are encountered in September and later in January. However, one cannot generalize this tendency. In another beech from Lammersdorf, sampled in the same way, the Pb distribution pattern is quite different, showing a rather steady increase in concentrations from the outer rings towards the center of the trunk, with only a small hump in the sapwood (Fig. 3b). During the sampling period, the shape of the Pb distribution in this tree shows only little variability. The stability of the lead pattern in the tree from Lammersdorf contrasts clearly with the described variability of the data of the Stolberg beech (Fig. 3). The reason for such differences is not yet understood.

For obvious technical and biological reasons, repeated sampling of the same trunk had to be done from slightly different positions. It was necessary to avoid the region in the wood that might be affected by wound reactions around the older boring channels. Thus, repeated samples from the same tree not only differed in sampling date, but also somewhat in the location within the trunk wood. The described differences in elemental distribution patterns might not only result from seasonal but also from spatial variations of mineral contents in the wood. In order to clarify this point, wood samples were extracted from a beech trunk from 4 different directions on the same date in April and were analysed (Fig. 4). For the metals Cd and Pb radial distribution patterns show little variation with the direction in the trunk. Only minor deviations from the general patterns are observed. On the whole, there was a steady centripetal increase in concentrations of Cd and Pb from all 4 directions in this beech trunk. However, differences in concentration levels were found. Highest concentrations of Cd and Pb were encountered in the southern part of the stem and lower contents on the eastern side. Such differences tend to decline in the center, where all 4 sample cores converge at the pith of the tree. Spatial variations in Zn concentrations and distribution patterns are much smaller in this tree

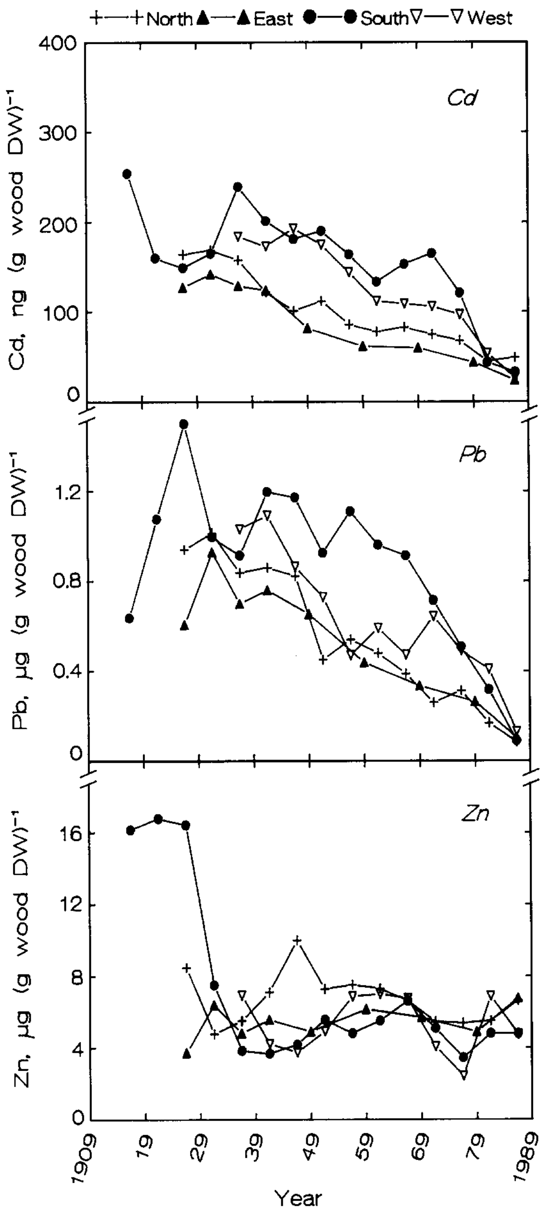

Fig. 4. Radial distribution of Cd, Pb and Zn in wood of a beech tree in Bielefeld in April 1989. Same tree as in Figure 1. Samples from 4 different directions, each 1.5 m above ground.

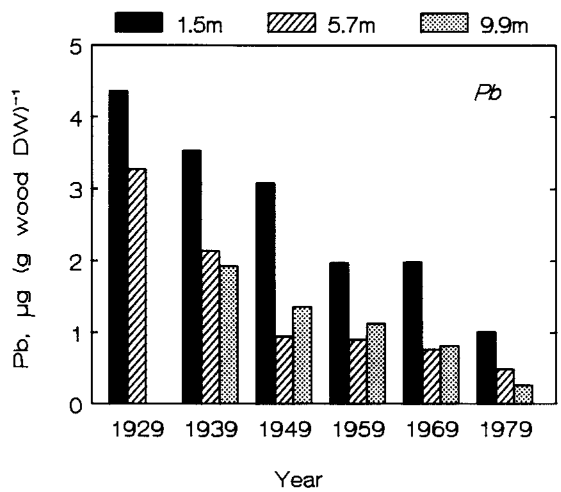

Fig. 5. Radial distribution of Pb in wood of a beech tree in Lammersdorf in January 1989. Same tree as in Figure 3b. Samples taken in eastern direction at 1.5, 5.7 and 9.9 m above ground.

(Fig. 4). Only the southern side shows a sharp increase in the inner heartwood. In the sapwood and in the outer portion of the heartwood Zn concentrations are rather stable showing only small variations on the circumference of the trunk.

The results described so far were obtained at about 1.5 m stem height. Variations of Pb concentrations and distribution patterns with stem height were investigated with wood samples from different levels above ground. In the trunk of a beech in Lammersdorf Pb concentrations tend to decline with height (Fig. 5). At all 3 examined levels radial distribution patterns are similar. From the outer part of the trunk Pb concentrations increase towards the center. However, this axial and radial pattern of Pb distribution is not typical for beech trunks in general. A survey of several trees from Lammersdorf and Stolberg revealed different distribution patterns (Hagemeyer & Breckle 1992). So far, no plausible and straightforward explanation for such differences in mineral distribution patterns among beech has been found.

Discussion

An analysis of mineral concentrations in the trunk wood of mature German beech revealed distinct radial distribution patterns of Cd, Pb and Zn. The shape of such patterns differs with the kind of element examined (Fig. 1). Wood samples taken from the same trunk on the same date, but from different directions exhibited rather similar mineral distribution patterns. However, there were some spatial variations in concentrations of the metals on the circumference of a beech trunk (Fig. 4). Thus, elemental distribution patterns show a certain variation with the location in the trunk wood. Such spatial deviations, however, were much smaller than the variations in patterns observed with samples extracted from the same tree at different times of the year. There was a marked seasonal variability of Cd and of Pb concentrations in wood of the investigated beech. Furthermore, also the distribution patterns of these two elements showed variations with the season (Figs. 2, 3). For Zn the radial distribution patterns were somewhat more stable around the year, although the determined concentrations showed variations with time. Thus, even though repeated samples of the same tree had to be taken from slightly different portions of the trunk, a conclusion concerning seasonal variability of mineral distribution patterns is possible. A comparison of the magnitude of spatial and seasonal variations shows, that changes of distribution patterns in time are much more pronounced than those with direction in the stem. Still, the applied method of sampling with an increment borer can only reveal seasonal changes, which are superimposed to apparently smaller spatial variations of mineral distribution patterns in beech wood.

What do these findings mean for the application of analytical data from tree rings as a means of biomonitoring heavy metal pollution? The presented results show, that at least for the examined beech trees the basic requirement of stability of mineral distribution patterns does not hold. Apparently, the patterns can change already in the course of one year and, thus, no reliable historical record of heavy metal pollution can be derived from such data. With the investigated beech trees from Germany dendroanalysis seems not feasible (Hagemeyer *et al.* 1989).

Comparable seasonal variations in mineral concentrations were observed not only in terms of total contents, but also in the xylem sap contents of beech. Glavac *et al.* (1990a) found highest Cd concentrations in the sap of beech in March and April and lower levels in the summer season. Marked changes were also observed with other elements, e.g. K, Ca, Mg and Mn in the xylem sap and were related to the physiological state of development of the trees during the year (Glavac *et al.* 1990b,c). In this investigation clear axial gradients of minerals in stems of beech were observed, that were also subject to seasonal variations. With Pb a decline in sap concentrations from the stem base towards the crown was observed (Glavac *et al.* 1989), that corresponds well with the presented results of our investigation (Fig. 5).

The variations in sap concentrations with time are attributed to storage and remobilization of ions, which can bind reversibly to fixed charges in cell walls or may be stored temporarily in parenchyma cells of the wood (Glavac *et al.* 1989). However, such phenomena cannot explain changes in total content of certain elements in wood as described in our investigation. Alterations in total mineral contents of the wood must result from long range transport processes in xylem vessels and in the phloem. Transport velocity in xylem of *Fagus sylvatica* was reported to reach 1 m h^{-1} (Huber & Schmidt 1936). The vessel system is, thus, capable to transport large quantities of dissolved minerals over long distances within a comparatively short period of time. Furthermore, a translocation of minerals in radial direction is possible along the rays (Ziegler 1968; Bamber 1976). Both, axial and radial transport processes may cause changes in distribution patterns of mineral elements in the course of the year. The picture occurs even more complicated as various elements are translocated with different velocities and in different quantities. This results from binding and immobilization of ions at fixed charges in the walls of xylem vessels (Biddulph *et al.* 1961; Petit & Van de Geijn 1978; Van de Geijn & Petit 1978, 1979; Wolterbeek 1987). Binding and exchange of ions depend on their electrochemical properties and cation exchange capacity of cell walls.

In order to draw a more realistic picture of mineral cycling phenomena in trees it will be necessary to distinguish clearly between those elements with a physiological role in plant metabolism, i.e. the nutrients like Zn and Cu, and potentially toxic elements without any known purpose in the organism, like Cd, Pb and others. For uptake, translocation, storage and disposal of nutrients plants have developed specially adapted biochemical and biophysical systems. As such bioelements serve distinct purposes in metabolism, e.g. as cofactors of enzymes, their translocation and distribution in the plant will be directed to the tissues were these minerals are needed. This has certainly a strong influence on the distribution of such minerals within the plant. When using a tree as source of material for biomonitoring one should not overlook the fact, that even a very old trunk is still part of a living organism with the ability to control transport and allocation of nutrients effectively.

Our presented results demonstrate the variability of radial mineral distribution patterns in the xylem of beech trunks. At least for this species the use of such data for biomonitoring trace element environmental pollution is doubtful. Nevertheless, several apparently successful dendroanalytical studies show, that this method may be useful, if the conditions are properly chosen and the results are carefully interpreted. Particularly events of exceedingly high trace element emissions may leave some distinct concentration peaks in the xylem rings that may be dated with reasonable accuracy. Also the choice of tree species seems important. In ring-porous wood, e.g. of *Quercus* or of *Ulmus*, xylem rings loose the ability to conduct water already a few years after their formation (Huber 1935; Ellmore & Ewers 1986). Thus, the axial translocation of minerals may be restricted to the outer sapwood. In the heartwood of such trees patterns of high trace element pollution may be somewhat more stable.

Future research should focus on the mobility of minerals within the plant as well as on chemical transport forms of minerals in xylem and phloem. A promising approach seems to be the analysis of tracheal sap (Bollard 1953; Glavac *et al.* 1989, 1990a; Okada *et al.* 1990), which yields information about the mobile fraction of minerals. This will provide a chance to investigate the chemical transport forms of elements in the xylem of trees (Clark *et al.* 1986). Eventually, flux models of the circulation of certain minerals within the tree may be derived as have been proposed for herbaceous plants (Oghoghorie & Pate 1972; Armstrong & Kirkby 1979; Jeschke *et al.* 1987).

Acknowledgements

Some of the data are taken from a research project funded by the Ministry of the Environment of the federal state of Nordrhein-Westfalen, F.R.G. (Hagemeyer & Breckle 1992). We gratefully acknowledge the support of the Forest Service in Bielefeld, Stolberg and Monschau (F.R.G.) during investigations carried out in their forests. Our thanks are extended to A. Stockey for critical remarks on the manuscript.

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