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Holocene raised bog deposits in the Netherlands as geochemical archives of prehistoric aerosols

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SUMMARY

High resolution records of lead and some other elements in Holocene raised bog deposits from the eastern Netherlands were compared with corresponding pollen records for the period 1000 BC to AD 1000. Trends in the curves of herbaceous human influence indicators parallel the recorded fluctuations of the aerosol input in the deposit. The chemical records may, in all probability, provide detailed information regarding the combined effects of soil erosion by agriculture (dust emission) and domestic fires in the regions around the bogs. Iron smelting operations may also have been responsible for the emission of aerosols into the atmosphere. The deposition of lead apparently corresponds to periods of agricultural and industrial expansion and depression. Already during periods of high population densities in prehistoric times some 'anthropogenic' aerosols dominated over 'natural' fractions. Relatively high lead levels in excavated prehistoric bones from agricultural societies may be explained by an increased uptake of airborne lead (in soil dust and smoke) via the lungs. Geochemical analysis can provide a more complete historical/prehistorical perspective of anthropogenic influence, especially in combination with palynological records.

Key-words: raised bogs, anthropogenic aerosols, lead, geochemical monitoring, prehistory, The Netherlands.

INTRODUCTION

Pollen analytical studies of the effect of man's impact on the vegetation after the introduction of agriculture were initiated by Iversen (1941). The analysis of pollen of apophytes and anthropochors in raised bog sections, such as *Artemisia* spp., *Plantago lanceolata, P. major* and *Rumex acetosella* especially shows the fluctuations of human impact over wider areas (Van Zeist 1955; Kubitzki 1961; Berglund 1969; Burrichter 1969; Overbeck 1975; Van Geel 1978; Dupont 1986; Middeldorp 1986). The extremely nutrient-deficient ombrotrophic bogs are important archives for the study of past deposition rates of aerosols, as precipitation and atmospheric dust provide the only source of supply (Livett, 1988). Comparisons between the trends in the curves of some pollen types and

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dust content of the peat may give an indication of the source area for the dust (Kramm 1978; Aaby 1986).

The exceptional ability of *Sphagnum* spp. to accumulate heavy metals is largely attributable to their high content of polygalacturonic acids (Clymo 1963; Clymo & Hayward 1982; Andrus 1986). According to Malmer (1988), *Sphagnum* plants accumulate inorganic constituents by: (1) direct influx of elements mainly to the chlorophyllous cells, (2) the adsorption of ions on the exchange sites on cell walls, and (3) a less specific accumulation of the elements, partly as particles, on the surface of the plant. Malmer (1988) showed that nearly all of the N, P, heavy metals, Al and probably also the constituents of the 'acid insoluble ash' (AIA; Clymo 1983), remained in the moss litter during the process of peat formation; the mosses do not use the inorganic nutrients of the underlying peat and the heavy metal concentration values increase in older parts of the moss plants, indicating the domination of passive accumulation instead of metabolic activity.

Aaby *et al.* (1979) have shown that the atmospheric input of Pb will be almost totally (99%) fixed in the surface layer of a raised bog. According to Aaby & Jacobsen (1979) Ca, Cd, Cu, Fe, Ni and V are also retained in the surface layer in quantities reflecting the actual deposition. The mobility of Zn and especially Mg can be considerable but nevertheless Aaby and Jacobsen's comparison between the deposition of these elements in the peat surface layer and their concentration in bulk precipitation showed a striking similarity. Na, K and Mn are translocated so that the concentration in peat profiles probably does not reflect the original atmospheric input. Translocation of elements was also shown by Malmer (1962), Clymo (1983) and Damman (1986). With some reservations it is possible to detect generalized patterns in element distribution in the peat profile. The elements K and Na occur in high concentration in the living *Sphagnum* surface; below the actual living parts, the concentrations drop sharply. Redox-sensitive elements like Fe, Zn, Mn and Pb are dissolved in the surface layers and accumulate at the mean water table. This observation ties in with Malmer (1988), who ranks Fe, Zn and Pb among those elements with the lowest retention values, but Mn, however, is ranked among the elements with the highest retention values. A less clear but somewhat similar behaviour is registered for Al, Mg and Ca. No obvious pattern was detected for Cu and Cd.

According to Guicherit (1974) the dominant natural source for the atmospheric input of lead is the dispersion of siliceous (soil) dusts. Other less important natural sources are sea spray, volcanic and cosmic dust and forest fires. Most geochemical pollution studies are concerned with recent (about the last 200 years) deposits (Lee & Tallis 1973; Menke 1987; Murozumi *et al.* 1969; Oldfield *et al.* 1977; Pakarinen 1978). An excellent review is given by Livett (1988). The analysis of older peat holds considerable potential for monitoring of aerosol deposition in the past (Ernst *et al.* 1974). The present study shows the results of some geochemical analyses of prehistoric samples from the Dutch raised bogs Engbertsdijksveen and Meerstalblok. Detailed analyses of pollen, other microfossils and macrofossils of 1-cm thick samples from both sites were available (Van der Molen & Hoekstra 1988; Dupont 1986) and material from the original peat monoliths was still available for the geochemical analysis.

In archaeology, lead concentrations in skeletal remains are traditionally explained in relation to dietry uptake. Uncritical use of historical sources, the erroneous belief that lead glazes were common before the middle ages and that lead water pipes contribute a potential health hazard (Hodge 1981), have resulted in a number of widely quoted studies concerning lead poisoning in Roman times (Gilfillan 1965; Nriagu 1983), which have tended to mask the wider issue of lead levels in both human and animal prehistoric bone (Runia 1987). The analysis of human bone from sites in northern Roman provinces, dating mainly to the 3–4th centuries AD, does reveal high lead concentrations (Waldron *et al.* 1976; Waldron 1983; Drasch 1982; Runia 1987) and despite the likelihood of post-depositional uptake (Waldron 1983) this must indicate increased exposure of the populations concerned. However, apart from the intake of lead via the food, airborne lead is also a factor, particularly because environmental lead is absorbed more readily. While only some 3–10% of ingested lead is retained, over 80% of the lead passing through the lungs is absorbed (Runia 1987). Airborne lead will therefore have had a more pronounced effect on the individual's lead burden than an equivalent amount taken in the food. Geochemical monitoring of raised bog deposits may give indications about lead aerosols in the past and in this way relatively high lead levels in prehistoric human and animal bone material (dating from periods during which lead was not yet used as a metal) may be explained (Runia 1987).

MATERIALS AND METHODS

Samples were taken in galvanized boxes (100 × 10 × 5 cm) which were pushed in a vertical peat face after peat digging. Initially chemical analysis was not foreseen and the possibility of some contamination cannot be excluded completely. However, the subsamples used for the chemical analysis were taken as close to the centre of the peat monolith as possible. Of the sections Engbertsdijksveen-IB and Meerstalblok-K2, subsamples, each 1-cm thick, were analysed. For the chemical analysis of the section Meerstalblok only samples from 40 to 60 cm depth were available.

For the Meerstalblok samples the element analysis for Co, Ba, I, Br, Cl, Se, Cr and Sr was worked out at the Netherlands Energy Research Foundation (ECN) by means of neutron activation analysis. After freeze-drying at −40°C the samples were exposed to a neutron flux of 5×10^{12} cm^{-2} s^{-1} for 2 h and the measurements were carried out after 16 days with a Ge(Li) detector.

The analysis of Pb, Fe, Zn, Mn, Mg, Na, Cu, Ca, K, and Al of the Meerstalblok samples and the Pb analyses of Engbertsdijksveen were carried out at the Hugo de Vries-Laboratory. Preceding the chemical analysis, the samples were dried at 105°C for 16 h and wet-ashed using nitric acid: approximately 30 mg of dried sample and 2 ml HNO_3 70% (Baker, 'Ultrex'-quality) were mixed in a pre-cleaned plastic tube. The mixture was dried in an oven at 80°C. Four millilitres of demineralized water were added and the tube was vigorously shaken.

Analyses of Pb were carried out with graphite furnace atomic absorption spectroscopy (AAS: Perkin Elmer 1100 B spectrometer and Perkin Elmer HGA 400 graphite furnace). The remaining elements were determined with the same AAS equipment using an air–acetylene or a nitrous oxide–acetylene flame after a × 10 dilution of the original sample. All analyses were carried out in duplicate.

RESULTS

The pollen diagram Engbertsdijksveen-IB represents the period of *c.* 1000 BC–*c.* AD 1000. In Fig. 1 a selection of palynological indicators for human influence on the vegetation is shown next to the Pb records. For several reasons (a.o. ^{14}C dating) not all the samples were available for the lead analysis. The local peat-forming taxa during the period concerned

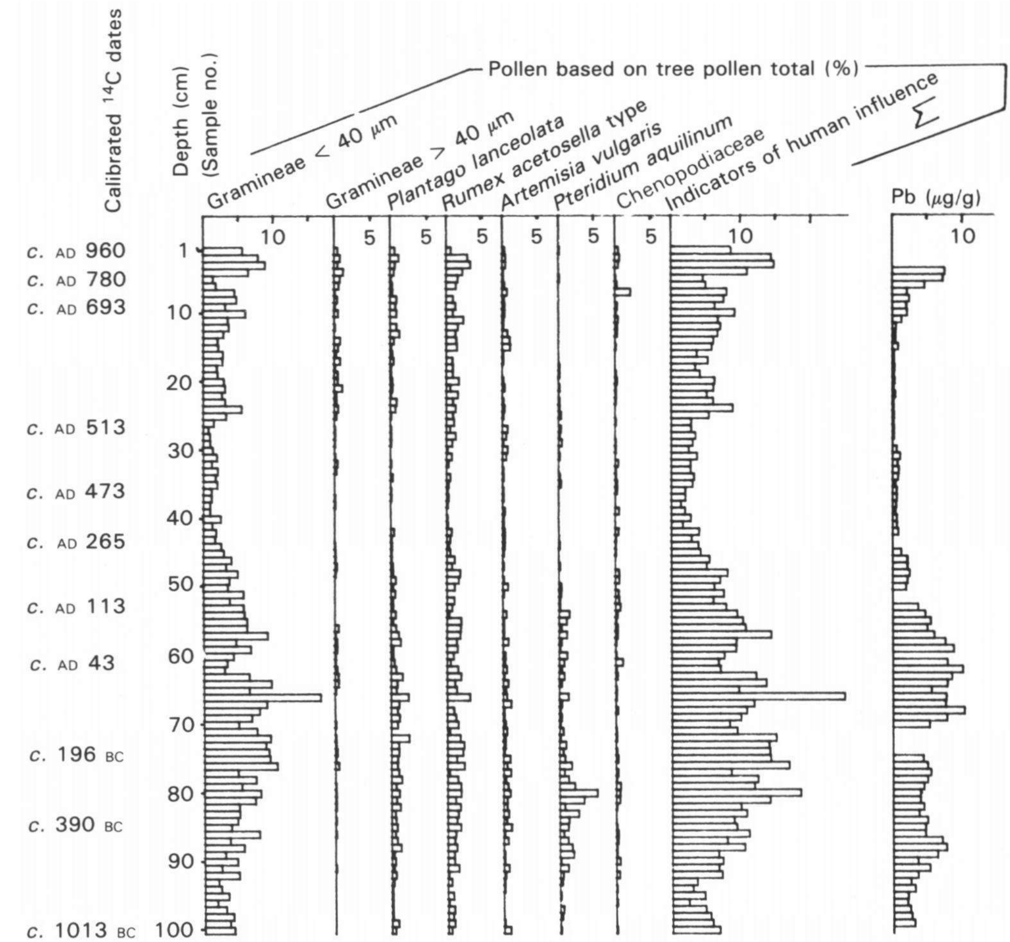

Fig. 1. Engbertsdijksveen-IB. Selection of palynological indicators for human influence on the regional vegetation, and Pb records of the peat deposit. The levels, 1–3, 29, 43, 44, 51, 52, 60, 71–74 and 100 cm were not available for lead analysis. Conventional radiocarbon dates (Van der Molen & Hoekstra 1988) were calibrated following Klein *et al.* (1982).

were *Eriophorum vaginatum, Rhynchospora alba, Scheuchzeria palustris, Sphagnum imbricatum* and *S. papillosum.* For detailed records concerning the local taxa we refer to Van der Molen & Hoekstra (1988).

A relatively high population density during the Late Bronze Age and especially during the Iron Age is evident from the relevant pollen curves (sample 100–*c.* sample 46). This phenomenon is characteristic for NW Europe (Kubitzki 1961; Burrichter 1969). The post-Roman population density decrease (Migration Period, see Kubitzki 1961; Van Geel 1978; Näsman & Lund 1988) is shown from sample 45 to sample 41. The upper 23 cm of the deposit represents the Early Mediaeval Period and the palynological records show a gradual increase of indicators of human impact. The Pb records show an overall parallel with the relatively high values in the lower 50 cm of the sample series. A considerable increase of palynological indicators of human influence in the upper 5 cm is paralleled by a rise in Pb values.

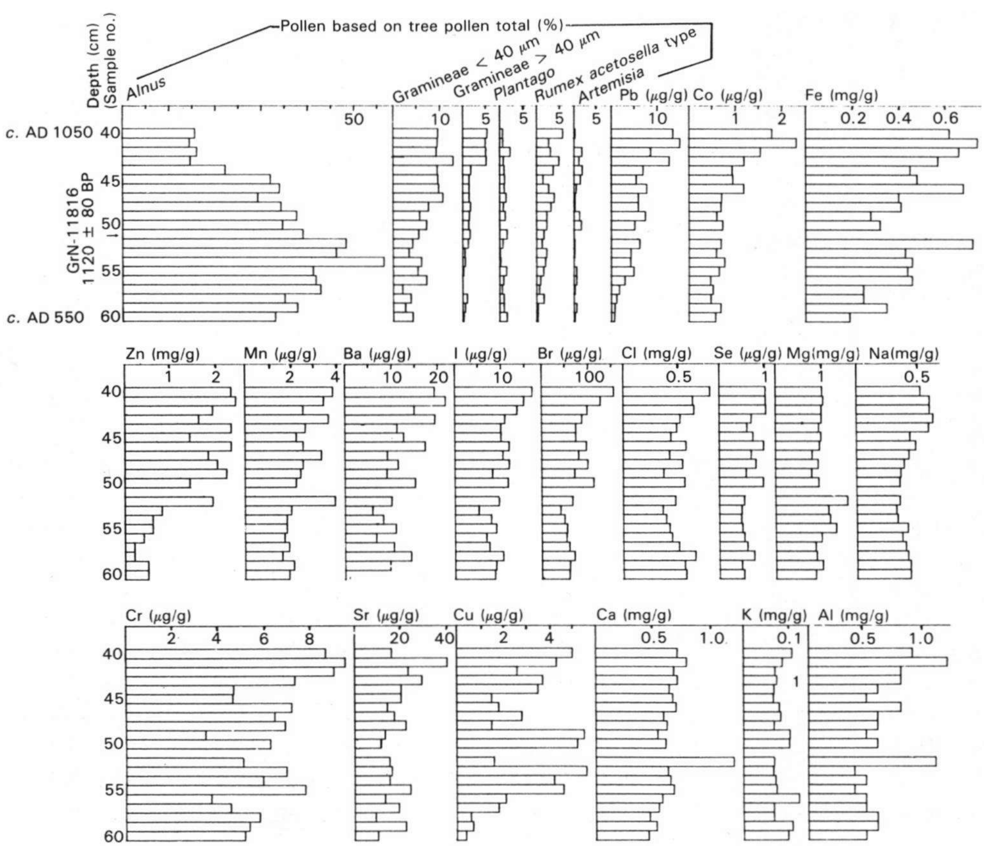

Fig. 2. Meerstalblok-K2. Selection of palynological indicators for human influence on the regional vegetation, and chemical records of the peat deposit. Sample 51 was not available for chemical analysis because it had been used for ^{14}C dating.

Figure 2 shows the chemical records and a selection of palynological records for Meerstalblok. This sample sequence (40–60 cm depth) of Meerstalblok-K2 represents the period of *c.* AD 550 to AD 1050 (Dupont 1986). Dates, given in calendar years, are computed by means of 'pollen density dating' (Middeldorp 1986). The pollen diagram of this interval is mainly characterized by the increase of apophytes and anthropochors. The curves of wild grasses and *Secale* also show a gradual continuous rise, corresponding with the population density increase during the Early Medieval Period which followed several centuries of low values characteristic for the Migration Period. The strictly local peat-forming vegetation was dominated by *Sphagnum papillosum* and *S. magellanicum*, with some *Sphagnum* sect. Acutifolia, *Sphagnum* sect. Cuspidata, *Andromeda polifolia* and *Erica tetralix.*

Comparison of chemical and palynological data shows parallel trends between the fluctuations of Pb, Co and Fe concentrations and the pollen curves of plants that indicate man's impact on the vegetation. At the present state of knowledge the geochemical behaviour of many of the recorded elements cannot be interpreted satisfactorily. Nevertheless in Fig. 2 the complete analytical results are presented.

DISCUSSION

In geochemical monitoring aerosol accumulation is interpreted to reflect aerosol fallout but, often independent from fluctuations in aerosol deposition, there are some other factors that control the concentrations in peat deposits.

1. The accumulation rate of the peat which is related with net organic production. A low accumulation rate, caused by, e.g. more intense oxidation processes (losses mainly of carbon) will result in enrichment of the inorganic fraction. Relatively large quantities of deposited aerosols affect the trophic conditions in the bog and may change the local vegetation composition and growth. An example of relatively low peat accumulation rate and changed local peat-forming vegetation is given by Van Geel & Middeldorp (1988) in their study of an Irish raised bog: the 17th century was characterized by a complete deforestation and the local bog vegetation in the treeless period was a very slowly accumulating *Narthecium* root peat. Detailed studies of raised bog deposits have shown considerable fluctuations of the peat accumulation rate. Only if we use a fine-resolution radiocarbon time control (van Geel & Mook, 1989) to calibrate 'pollen density dating' (Middeldorp 1986) can influx of aerosols (per unit of time and per unit of surface) be computed from the geochemical record.

2. The varying spectrum of peat-forming plants will be reflected in the geochemical results (species effect; Livett *et al.* 1979; Clymo 1983). First, vascular plants will cause other effects on the cycling of inorganic elements through root uptake and litter fall than mosses. Secondly, Ericaceae, *Eriophorum* and some other vascular bog plants are missing the special ability of *Sphagnum* species to accumulate heavy metals; however, Ericaceae accumulate considerable quantities of manganese in their leaves (Ernst *et al.* 1974). Thirdly, the contact surface of the bog differs considerably from one bog vegetation type to another, causing differences in the capture of aerosols in the bog ecosystem. The present study shows very low Pb concentrations (Fig. 1, 9–27 cm) in peat samples with many *Eriophorum* remains. Apart from a relationship with a relatively low human population density during this period, these low records probably reflect a 'species effect' in addition to a 'dilution effect' because of a high accumulation rate of *Eriophorum* peat (see Fig. 1).

3. According to Aaby *et al.* (1979) the heavy metal content of the wet 'hollow peat' is different from the content in 'hummock peat' formed in relatively dry conditions (see also Aulio 1982 and Pakarinen 1981). Aaby & Jacobsen (1979) showed that the storage of heavy metals in relatively wet parts of raised bogs (hollows) may be used as a recorder of changes in the deposition in the past, but according to Malmer (1988) hummock species should serve better than hollow species. Malmer argues that the more anaerobic environment in the hollows contributes to an enrichment in the lower parts of the moss layer. However, the better aeration of the hummocks, giving rise to a higher redox potential and a lower pH around the lower parts of the mosses, increases the mobility of several metals. This lower pH results from a higher cation exchange capacity of the hummock-forming species (Clymo 1963; Clymo & Hayward 1982; Andrus 1986).

There are always subsequent changes in strictly local moist conditions at a particular sample site (climate-induced or related to the internal dynamics of the bog) and such changes will have an effect on the accumulation of chemical elements (site effect) as well as on the botanical spectrum of peat-forming plants (species effect; see 2.). If site and species effects are important, a correlation is expected between one or more elements and the composition of the peat. However, the comparison of the chemical records of the present study with the former plant composition (as recorded by macrofossil analysis) does not

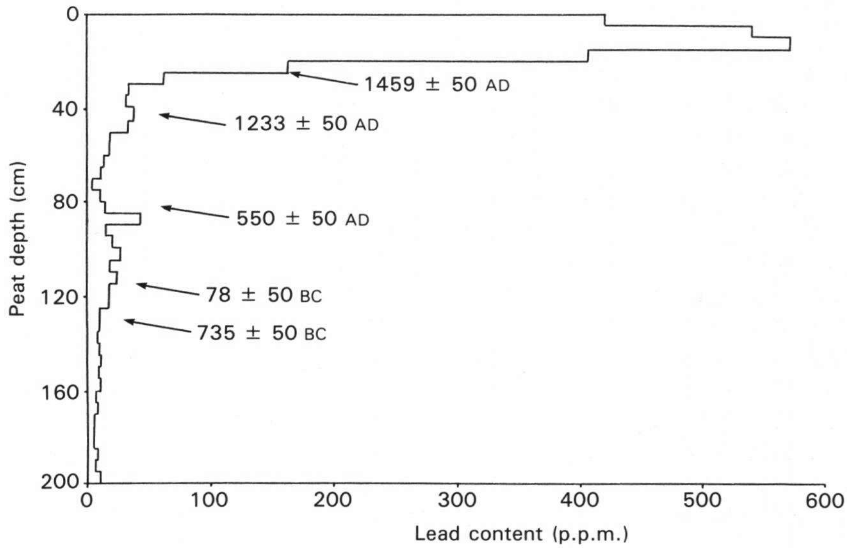

Fig. 3. Lead analysis of a peat profile from Featherbed Moss (Derbyshire, UK), adapted from Lee & Tallis (1973). Reprinted with permission from *Nature*: copyright 1973 MacMillan Magazines Ltd.

show appreciable evidence for these effects. The chemical concentration values are dominated by the original aerosol input and only to a minor extent influenced by species and site effects.

Lee & Tallis (1973) studied the lead deposition of a peat profile from Featherbed Moss (Snake Pass, Derbyshire, UK; Fig. 3). Radiocarbon determinations by Switsur & West (1973) and the pollen records of Tallis (1964) and Hicks (1971) show that clearance of the forest in the southern Pennines was small scale and temporary throughout the Neolithic and Bronze Age and became extensive during the Iron Age and Roman times. There was a decrease of human impact during the post-Roman, pre-Norman period; subsequently, cleared areas expanded again. In their Featherbed Moss records Lee & Tallis (1973) recognized lead pollution during Roman times and interpreted it as the effect of Roman lead mining activities in the area (see also Ernst *et al.* 1974, for similar results in Germany). This maximum of lead pollution during Roman times was followed by a minimum, and a Late Medieval sharp increase correlates with the rise of Derbyshire lead industry. However, the lead record of the Featherbed Moss profile shows an earlier, minor, but evident increase of the lead content of the peat deposit at *c.* 700 BC. Lee & Tallis (1973) neither mentioned nor interpreted this prehistoric increase, but in our opinion this phenomenon (which has nothing to do with lead industry) reflects the increased influx of soil dust at the start of the Iron Age in the area. According to Livett *et al.* (1979) the earliest detectable enrichments for lead (attributed to mining and extraction of lead on a small scale) date back to the early Middle Ages but the study by Lee & Tallis (1973) and the present study of Engbertsdijksveen and Meerstalblok show that lead can be used as a tracer in geochemical monitoring of prehistoric deposits as well. The records of Meerstalblok (Fig. 2) may show that Co and Fe can also be used. The records of these elements probably also reveal early pollution.

We may conclude that during prehistoric and protohistoric periods human activities already had a considerable impact on the atmospheric concentration of these elements:

anthropogenic aerosols dominated over natural sources. The influx of some elements was probably also (partly) related to metal working (metallurgic dust and fumes). Lead was not used as a metal for tools before the Roman Iron Age. During the pre-Roman Iron Age the local use of bog iron ore is evident from the archaeological studies (Brongers & Woltering 1978; Laban *et al.* 1988). It is postulated here that iron smelters injected lead aerosols and other metals together with Fe into the atmosphere. However, at present the possible effect of prehistoric smelter emissions cannot be separated from the soil dust component and other anthropogenic and natural sources.

The increasing population density during the Bronze Age and Iron Age will have resulted in an increasingly open landscape with larger areas of bare, loose and mobile soils. The natural forest vegetation was strongly reduced and the fields were less protected against wind. Large amounts of dust can be raised in the atmosphere during ploughing and harrowing of dry soils. Relatively high tree pollen influxes indicate that the reduced anthropogenic activity during the Migration Period led to reforestation (Van der Molen & Hoekstra 1988). The palynological records indicate a temporary population density decrease and this phenomenon is also recorded geochemically (Fig. 1).

The recorded Pb levels in raised bogs of holocene age indicate that during prehistoric periods (preceding the use of lead as a metal) atmospheric concentrations of lead were related to human activities (soil dust, fires). Atmospheric fluctuations of lead may also have had a direct effect on the lead content in bones of man and animals. Relatively high lead levels in human bones from beyond the Roman frontier or from pre-Roman contexts and in animal bones (Runia 1987) may be partly explained as the effects of increased uptake of airborne lead via the lungs. The sharp increase of lead concentrations in bones of Roman times would correspond to a period of intensification of agriculture when even marginal land was increasingly taken into cultivation. Widespread abandonment in NW Europe and a return to more extensive forms of agriculture in the early middle ages may account for the marked differences in bone concentrations in this period, with ensuing increases once more reflecting agricultural expansion, as well as the increasing use of lead for industrial purposes (Drasch 1982).

The goal of a recently started scientific programme of the European Science Foundation is to study the possible mutual relationships between the European paleoclimate and man since the last glaciation (ESF 1988). The magnitude of prehistoric agricultural and industrial activities are at the starting point for understanding the present-day anthropogenic changes of climate associated with world-wide industrial activity. Geochemical monitoring of raised bog deposits is one of the ways to come to a better knowledge and to answer the question of whether anthropogenic changes of climate have taken place, e.g. fine dust particles in a moist atmosphere can act as condensation nuclei for cloud and rain formation (Pye 1987). Deforestation and land management may cause changes in albedo, water cycles and atmospheric CO_2 content.

Raised bogs are an especially interesting archive because the fossil micro- and macrobotanical spectra and the composition of some climate-indicative stable isotopes can be recorded and interpreted with the same fine time-resolution as the element analysis of the same peat cores.

We wonder how detailed geochemical monitoring of raised bog deposits can be. Will it be possible to discern (by chemical characterization) the different sources of soil dust? (e.g. the probable increasing influence of Saharan dust as opposed to the influx of regional soil dust). Will it be possible to recognize levels with relatively high concentrations of volcanic or cosmic dust? (volcanic dust-veils in the lower stratosphere can lead to short-term cooling of global climate). If specific geochemical characterization of volcanic dust—maybe necessarily at isotope level—is possible than an extra possibility of correlation and indirect dating between European bog deposits (and even more distant locations) will become available (fine-resolution tephrochronology). At the same time the possible short-term climatic effect of volcanic dust emissions (cf. Baillie & Munro 1988) can be recorded by studying the (subsequently climatologically induced) response of former bog plants (e.g. changes in spectrum of species) and their isotope composition.

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