

Stratigraphy of nutrients and metals in sediment profiles of two dimictic lakes in North-Eastern Germany

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Received: 30 October 2006 / Accepted: 23 September 2007 / Published online: 12 October 2007
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Abstract A sedimentological study of two dimictic lakes in North-Eastern Germany provided a record of anthropogenic impacts and historical changes of water quality. The upper 50 cm sediment profiles were compared for major nutrients and selected major trace elements. The sediments were dated by ^{210}Pb and ^{137}Cs measurement. The upper 50 cm sediment profiles represent approximately the last 100 years of history in both lakes. Element analyses show different characteristic stratigraphic patterns in both lakes. Based on the nutrient and metal stratification, three characteristic time periods can be documented for both lakes. In addition to agricultural use of the catchment area, atmospheric pollution greatly influenced the metal concentration in the sediment layers. Variation in the external loading and redox conditions in the hypolimnion explain the variation in the composition and accumulation of metals in the sediment stratigraphy. No increases or changes in the trophic level of either lake could be documented based on the accumulation of the nutrients C, N and P. The ratio of Fe/Mn and Fe/Ca characterized the changing redox conditions. The stratigraphy of Pb and Zn agrees with the historical variation in atmospheric pollution and confirms literature values for Central and North Europe. The drop in Pb and Zn over the last 10–15 years is a regional effect in North-Eastern Germany.

Keywords Lake sediment · Palaeolimnology · Geochemistry · Nutrients · Trace metals · Redox transformation · Water Management · Biogeochemistry · Water Quality

Introduction

Lake sediments consist of mixtures of particles from external sources and from internal biological and geochemical processes (Hongve 2003). Sediment stratigraphy therefore provides information about anthropogenic impacts from the catchment area and atmospheric pollution. Paleolimnology as a science has developed rapidly by introducing new methods of accurately dating recent sediments, especially by using radiometric methods (Abril 2003).

Fossil organisms and pigments can reveal the historical development of lakes and their catchment areas (Lotter 2001; Walker et al. 1997). Transfer functions and their reconstruction based on the investigation of diatoms in the sediment can be used to characterise various water chemistry parameters, especially pH and phosphorus (Ter Braak and van Dam 1989; Bennion et al. 1995). Fossil meio-benthos, e.g. chironomids, ostracods and cladocerans, also indicate past environments in lakes, including changes in hypolimnetic oxygen conditions (Löffler 1986; Scharf 1998).

In addition to the fossil organisms, geochemical sediment analyses are also valuable for reconstructing eutrophication and atmospheric pollution (Bengston and Persson 1978; Rippey 1990). The concentrations of different metals yield information on variations in the global environment, catchment areas and processes in the lake. Aluminum and lithium are typical metals that reflect

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erosion in catchments (Engstrom and Wright 1984). Lead, zinc and sulfur, on the other hand, are better indicators of atmospheric pollution of the limnic waters. The nutrients carbon, nitrogen and phosphorus characterise the nutrient accumulation and eutrophication effect in lakes. Redox-sensitive elements such as iron and manganese have also been used to provide information on changes in hypolimnetic oxygen levels (Schaller et al. 1997; Garrison and Wakeman 2000).

Chemical and biological processes, especially at the sediment-water interface during the time of deposition, transform nutrients and release part of the sedimentary matter (Bostroem et al. 1988). Penn et al. (1995) calculated the diagenesis of P from measurements of labile P components and reported a constant ratio of labile to refractory phosphorus in freshly deposited sediment. Kenney et al. (2001) interpreted polyphosphate accumulation in sediments as an effect of high phosphorus load and phosphorus storage in algal cells.

This study describes the stratification of metals and nutrients in the upper 50 cm of sediments of two dimictic lakes in North-Eastern Germany. The goal is to obtain better information about atmospheric pollution (global effects in Europe) as well as eutrophication processes

(regional effects from the catchment area) in lakes here. In addition, the metal concentration and ratio will be used to reconstruct the variation in redox conditions.

Materials and methods

Study areas

Lake Dudinghausen and Lake Tiefer are located in Mecklenburg-Vorpommern (North-Eastern Germany), approximately 40 km southeast of Rostock (Fig. 1). Relevant morphometric and water quality parameters are presented in Table 1. Both lakes have thermic stratification and anoxic hypolimnion with a P release from the surface sediment during the summer stratification (Selig and Schlungbaum 2003). In addition to the phytoplankton community in the epilimnion, a second primary production layer with phototrophic sulfur bacteria was observed recently in the hypolimnion of Lake Dudinghausen (Selig et al. 2004).

Based on measurements in 1999 and 2000, the lakes can be classified as mesotrophic (Lake Dudinghausen) and low eutrophic (Lake Tiefer) according to OECD (1982) criteria.

Fig. 1 Geographic location of Lake Dudinghausen and Lake Tiefer in North-Eastern Germany

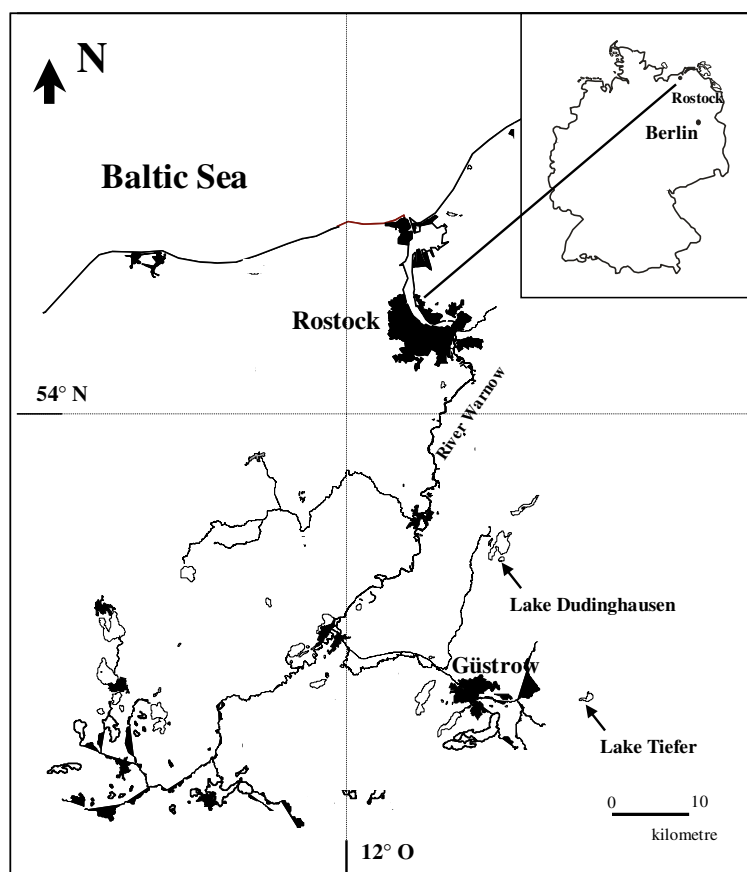


Table 1 Morphological and limnological characteristics of Lake Dudinghausen and Lake Tiefer

	Lake Dudinghausen	Lake Tiefer
Surface area in ha	18.8	15.9
Volume in m ³	1.29 × 10 ⁶	1.69 × 10 ⁶
Mean depth in m	6.9	10.7
Maximum depth in m	15.2	30.6
Catchment area in ha	25	144
Total phosphorus in µg l ⁻¹	32–80	21–65
Secchi depth in m	0.6–2.0	0.7–2.2
Chlorophyll a in µg l ⁻¹	2–15	2–13
Classification 1999/2000	mesotrophic	eutrophic

Sampling

Sediment cores were collected at the deepest parts of the lakes with a sediment corer (Mondsee corer—UWITEC) during spring 2000 (Lake Dudinghausen) and 2002 (Lake Tiefer). One core was selected for nutrient and metal analyses and a second core was used to analyse ²¹⁰Pb and ¹³⁷Cs. The sediment cores were transported to the laboratory, cut and 1-cm slices were frozen at -80°C and freeze-dried.

Sediment dating

The sediments were dated by the Leibniz Laboratory for Radiometric Research by the University Kiel. The contents of the radionuclides ²¹⁰Pb and ¹³⁷Cs were measured using a Germanium-semiconductor-detector-system for a minimum of 48 h based on the gamma radiation of 46.5 and 661 kV (Appleby et al. 1992). Sediment core chronology was calculated using the constant rate of supply (CRS) model (Appleby and Oldfield 1983). The minerogenic ²¹⁰Pb background was taken to be 10 Bq kg⁻¹.

Nutrient and metal analysis

For analysis of nutrients and metals, sediments were dried at 105°C. Total carbon (TC), total organic carbon (TOC—after acidification with 0.1 N HCl) and total nitrogen (TN) were analysed with a C/N-Analyser (HERAEUS elemental vario EL) according to Verardo et al. (1990). Inorganic carbon (TIC) was calculated as the difference between TC and TOC. Sulphur was analysed with a C/S-Analyser (“Multi-EA-2000” from Analytik Jena) after combustion at 1,300°C and IR-detection.

The trace elements P, Fe, Mn, Al, Ca, Mg, Cu, Li, Mn, Pb and Zn were digested (pressure digestion bomb -3 h

180°C) using a combination of nitric, hydrochloric, perchloric and hydrofluoric acids and the digestate was analysed by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES, Neumann et al. 1997).

Statistical analysis

The Spearman rank correlation coefficient was computed to test the association between pairs of parameters. In addition to the correlation analysis, the sediment horizons were grouped using an agglomerative hierarchical cluster analysis (non-transformed data, fusion: complete linkage, distance: Euclidian distance, software SPSS).

Results

Sediment dating

²¹⁰Pb profile shows an exponential decline from the sediment–water interface to 30 cm in Lake Dudinghausen (Fig. 2a). In the upper sediment the ²¹⁰Pb activity fluctuates widely but at a low level. In Lake Tiefer the ²¹⁰Pb activity shows no increase in the upper 5 cm (Fig. 2b). Between 5 and 20 cm, an exponential decline was also found in this lake. The results of the CRS model suggest that the sediment cores spanned the 100-year period of interest here and have not experienced significant bioturbation or other mixing—except the upper 5 cm layer in Lake Tiefer. In both lakes a time period from 1923–1926 to 2000 was calculated in the 40-cm sediment layer according to the ²¹⁰Pb activity profiles.

Based on the ¹³⁷CS activity profiles, the 1963 peak from nuclear bomb tests were found at 19 cm depth in Lake Dudinghausen and at 11 cm in Lake Tiefer (Fig. 2c, d). The nuclear accident at Chernobyl in 1986 is documented in both lakes (in 5 cm for Lake Dudinghausen and 4 cm for Lake Tiefer). These data documented a higher sedimentation rate (twofold higher) in Lake Dudinghausen between 1963 and 1986. The different sedimentation rate phases according to ¹³⁷CS activity profiles are described in Table 2. After 1986 the sedimentation rates were similar in both lakes. The higher sedimentation rate in Lake Dudinghausen was validated by ²¹⁰Pb dating. Whereas the upper 10 cm documented the last 20 years in Lake Tiefer, in Lake Dudinghausen it represented the last 28 years.

Nutrient and metal accumulation

TOC, TN and P concentrations increased in the upper sediment in both lakes (Fig. 3). Whereas the values

Fig. 2 ^{210}Pb and ^{137}Cs profiles of Lake Dudinghausen (a, b) and Lake Tiefer (c, d)

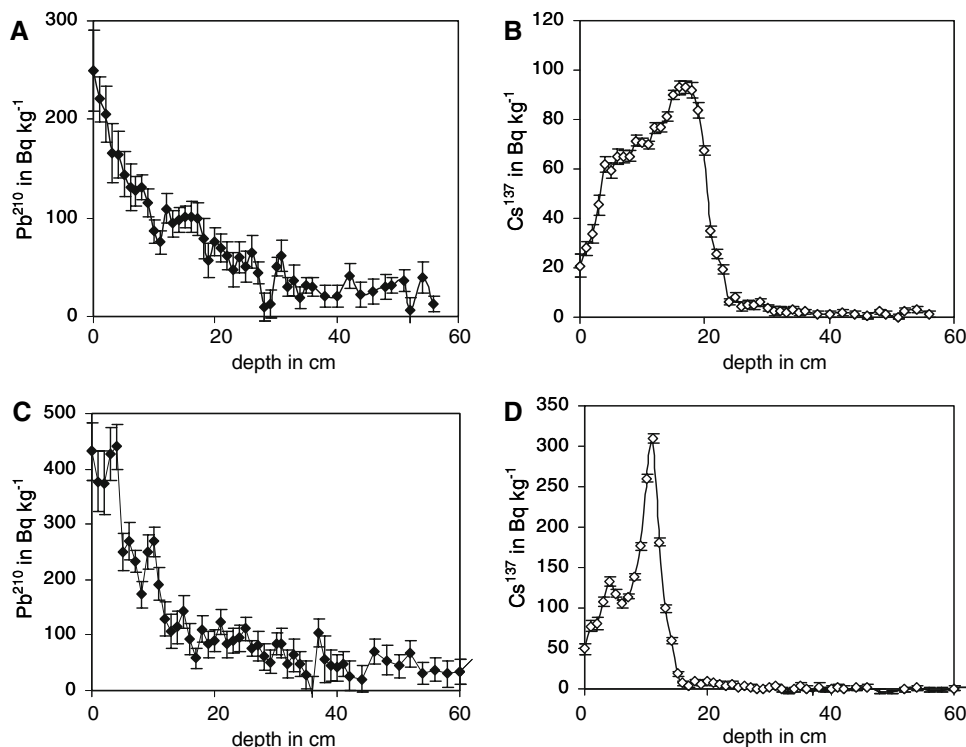


Table 2 Calculated sedimentation rates based on ^{137}CS activity profiles

Time period	Lake Tiefer (cm a^{-1})	Lake Dudinghausen (cm a^{-1})
1954–1963	0.44	0.33
1963–1986	0.30	0.61
after 1986	0.22	0.28

increased in the upper 10 cm in Lake Tiefer (representing the last 20 years), in Lake Dudinghausen they increased only in the upper 7 cm (representing the last 18 years). The P concentration was $<1 \text{ mg g}^{-1} \text{ dw}$ in both lakes in the deeper lake sediment. In the upper 9 or 7 cm, the values increased to the maximum concentration of 2.0 (Lake Tiefer) and 2.5 (Lake Dudinghausen) $\text{mg g}^{-1} \text{ dw}$ toward the surface.

The C/N ratio fluctuated minimally around a mean of about 10. This ratio was lower only in the upper 3 cm. In contrast to the C/N ratio, the C/P and N/P ratios decreased in the both sediment cores from the bottom to the sediment surface. These ratios were clearly higher in Lake Tiefer in the whole sediment core.

Both lakes showed a high correlation between the geogenic elements Al, Li and Mg (Table 3). Whereas in Lake Dudinghausen these concentrations slowly decreased toward the surface, Lake Tiefer showed a higher variation: a sudden increase occurred in the upper 10 cm horizon, for example.

In Lake Dudinghausen the P concentration was highly correlated with Mn and poorly correlated with Fe (Table 3), whereas in Lake Tiefer there was no correlation. Fe decreased and Mn increased toward the surface in Lake Dudinghausen.

The Fe/Mn and Fe/Ca ratios revealed a similar course in both lakes. Whereas the ratios increased between 50 and 40 cm, they fell continuously toward the surface in Lake Dudinghausen. In Lake Tiefer, the ratios dropped abruptly at 40 cm but varied less in the upper 40 cm. Here, parallel to the geogenic elements (Al, Li, Mg), both ratios increased in the 9–10 cm horizon.

In both lakes the S concentration showed a similar trend, but with different time scale. Two peaks occurred in the sediment profiles (37 and 13 cm in Lake Dudinghausen and 42 and 18 cm in Lake Tiefer). The concentrations dropped distinctly in the upper sediment layer.

The Cu concentrations were low: $12\text{--}18 \mu\text{g g}^{-1} \text{ dw}$ at 50 cm depth. The values increased to $30 \mu\text{g g}^{-1} \text{ dw}$ in Lake Dudinghausen and $50 \mu\text{g g}^{-1} \text{ dw}$ in Lake Tiefer. The Cu concentration decreased notably only in the upper 2 cm in both lakes.

For Zn and Pb, a continuous increase was evident from the bottom of the sediment core to the horizons 17 cm (Lake Dudinghausen) and 15 cm (Lake Tiefer). The sediment layers with the highest concentration represent the early 1960s in both lakes. The maximum concentrations were higher in Lake Tiefer ($188 \mu\text{g Zn g}^{-1} \text{ dw}$ and $96 \mu\text{g Pb g}^{-1} \text{ dw}$). In the upper sediment layer the

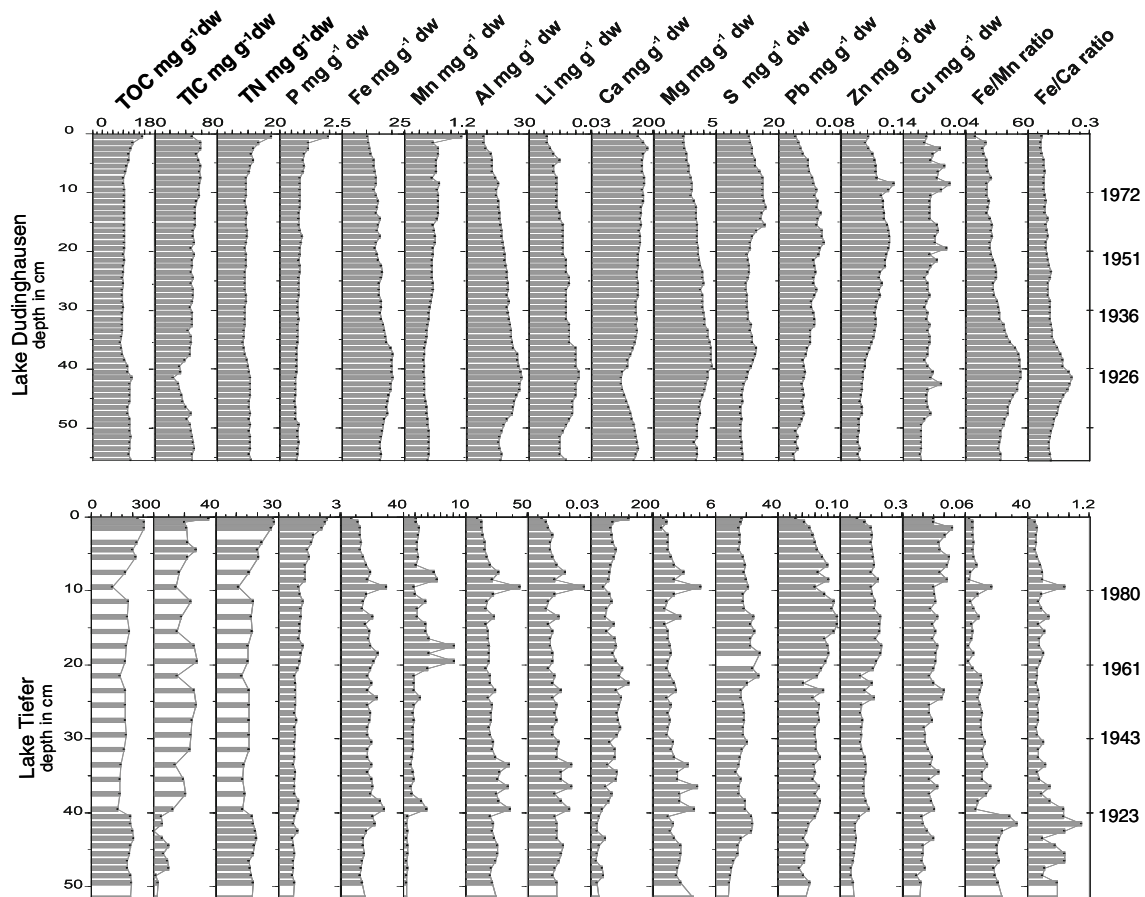


Fig. 3 Stratigraphy of nutrients and metals in sediment cores of Lake Dudinghausen and Lake Tiefer

concentrations decreased, but did not return to the background values at 50 cm.

Cluster analysis

The cluster analysis of the sediment horizons yielded four clusters for Lake Dudinghausen and three clusters for Lake Tiefer (Fig. 4). In Lake Dudinghausen the sediment horizons were subdivided into the sections 0–20 cm (after 1951), 20–36 cm (1950–1930) and two clusters before 1925 (41–46 cm and 37–40 cm/ 47–55 cm). The cluster analysis of Lake Tiefer revealed one phase before 1920 (39 cm depth), another between 1920 and 1985 (37–5 cm), and a third after 1985.

Discussion

For Lake Dudinghausen the first human settlement in the catchment area was already established 4,800 years ago (Dreßler et al. 2006), whereas for Lake Tiefer this date is 4,000 BP (Subboreal - Selig et al. 2002). Despite the long

period of human activity, a strong increase in eutrophication has only been documented in the upper 50 cm sediment layer, which was investigated in this publication. The diatom-inferred TP (Di-TP) revealed an increased P concentration in the water body of both lakes in the last 75–100 years (Dreßler et al. 2007; Selig et al. 2002). According to the sediment dating, the short sediment cores of 50 cm describe only the effects of the industrial phase over the last 100 years, the influence of the varied political situations in East Germany (reunification), and the initial effect of the decreasing nutrient loading and atmospheric pollution in the last 10 years. Although both lakes are located close to one another, the sediment cores show variations for nutrients and metals between both lakes.

Pre-Second World War period

In both lakes, we found an initial increase in Fe/Mn and Fe/Ca ratios, followed by a strong decrease. Changes of these ratios in the sediments are linked to changing redox conditions at the sediment surface (Itkonen et al. 1999; Loizeau et al. 2001; Koinig et al. 2003). Generally,

Table 3 Spearman rank correlation coefficient ($P < 0.001$) between nutrient and metals in sediment profiles of Lake Dudinghausen and Lake Tiefer

	C _{org}	C _{inorg}	N	P	Fe	Mn	Al	Li	Ca	Mg	S	Pb	Zn	Cu
Lake Dudinghausen														
C _{org}	–	0.039	0.849	0.243	0.022	0.063	0.084	0.058	0.049	0.201	0.168	0.532	0.368	0.052
C _{inorg}		–	0.001	0.182	0.698	0.375	0.726	0.711	0.900	0.527	0.270	0.089	0.238	0.075
N			–	0.557	0.111	0.228	0.196	0.155	0.001	0.300	0.058	0.357	0.185	0.019
P				–	0.475	0.742	0.494	0.478	0.225	0.433	0.042	0.005	0.026	0.026
Fe					–	0.676	0.879	0.874	0.769	0.777	0.129	0.051	0.217	0.097
Mn						–	0.614	0.634	0.468	0.556	0.205	0.063	0.232	0.093
Al							–	0.952	0.730	0.877	0.134	0.001	0.070	0.037
Li								–	0.741	0.823	0.165	0.004	0.096	0.052
Ca									–	0.484	0.317	0.164	0.363	0.094
Mg										–	0.068	0.005	0.028	0.065
S											–	0.337	0.436	0.277
Pb												–	0.859	0.237
Zn													–	0.360
Cu														–
Lake Tiefer														
C _{org}	–	0.004	0.908	0.426	0.584	0.015	0.583	0.500	0.001	0.354	0.016	0.097	0.008	0.049
C _{inorg}		–	0.032	0.283	0.011	0.280	0.130	0.111	0.813	0.197	0.001	0.165	0.330	0.387
N			–	0.650	0.544	0.007	0.525	0.450	0.024	0.323	0.006	0.113	0.001	0.087
P				–	0.193	0.092	0.312	0.249	0.220	0.204	0.003	0.002	0.148	0.379
Fe					–	0.063	0.578	0.529	0.038	0.310	0.096	0.200	0.110	6.1–E7
Mn						–	0.016	0.022	0.180	0.032	0.003	0.330	0.576	0.257
Al							–	0.939	0.181	0.812	0.002	0.003	0.001	0.042
Li								–	0.166	0.736	0.007	0.003	0.001	0.018
Ca									–	0.256	0.007	0.073	0.168	0.259
Mg										–	0.017	0.007	0.016	0.071
S											–	0.118	0.075	0.037
Pb												–	0.692	0.233
Zn													–	0.560
Cu														–

increased bioproduction and therefore increased anoxia in deeper waters will lead to a stronger solution of Mn and Ca compared to Fe (the reduced form of Fe is less stable in the water column than that of Mn and Ca) from the sediment. This is reflected in lower Mn and Ca concentrations in the sediment and correspondingly high Fe/Mn and Fe/Ca ratios. In Lake Dudinghausen, the increasingly anoxic conditions on the sediment-water interface were also supported by the ratio of chlorophyll derivatives and carotenoids (Dreßler et al. 2007).

In the second part of this time period, the Fe/Mn and Fe/Ca ratios decreased. Similarly, decreasing Al and Fe values suggest reduced exogenous mineral inputs due to increased soil stabilisation and decreased erosion for both lakes. This probably reflects decreasing nutrient loads and reduced agricultural activity in the catchment areas. One explanation for this development is the influence of the Second

World War on agriculture in Germany. Dreßler et al. (2007) confirm the decline in the trophic level based on the decreasing of Di-TP.

P accumulation in the sediment did not reflect the changing redox conditions or the higher trophic level in both lakes; it is known that the geochemical phosphorus record may not relate directly to the phosphorus loading history (Engstrom and Wright 1984). The difference between P loading and sediment-accumulated P concentrations is primarily due to post-depositional mobility, which itself involves internal release from the upper sediment. Selig et al. (2005) investigated sediment cores of both lakes representing deposition over the last 4,000 and 10,000 years. The highest phosphorus concentrations were recorded in deeper, anoxic sediments in both lake sediments and not in the upper 50 cm sediment layer. In Lake Tiefer, high P in sediment was associated with high

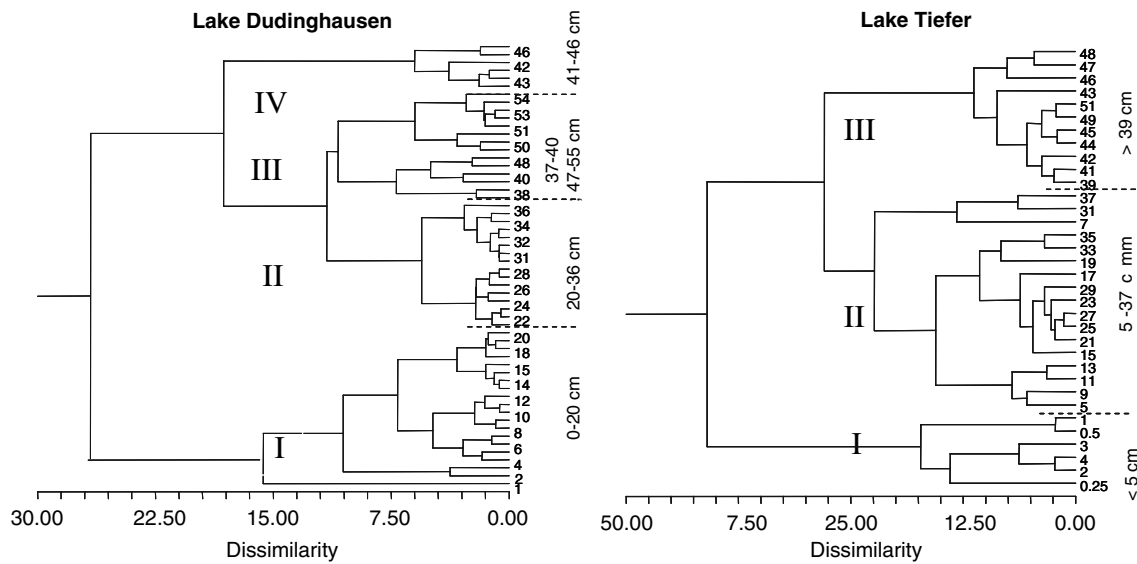


Fig. 4 Hierarchical cluster analysis (non-transformed data, fusion: complete linkage, distance: euclidian distance) of sediment sample of the both lakes

concentrations of settlement-related pollen (e.g. *Plantago* spp., *Rumex* spp., *Secale* spp.) and high amounts of aluminium, which indicate erosional inputs (Selig et al. 2002). In contrast, the high P in Lake Dudinghausen sediment was described as precipitation with iron (vivianite)—an effect of high inflow of dissolved iron with groundwater (Selig et al. 2005). In the 50–10 cm sediment layers the P concentration was stable around 1 mg g⁻¹ dw.

In contrast to the decreasing Al und Fe concentrations, Pb and Zn increased continuously during this time period. The concentrations, however, clearly exceed the background concentration. As opposed to these two metals, Cu did not increase in the sediment profiles of either lake. This effect cannot be explained by altered catchment use. The heavy metals Pb and Zn can be reliably used to reconstruct regional trends of atmospheric deposition. Both lakes have a small catchment area and lack industrial production sites. Anthropogenic inputs of heavy metals from the catchment areas are therefore thought to be low. Both metals are characteristic of atmospheric pollution in North Europe.

Post-Second World War period

After the Second World War the agrarian structure in East Germany was completely reorganised. The small-village landscape was transformed into an industrialised one. The reorganisation of agriculture has clearly affected the eutrophication of lakes in Europe (Olsson et al. 1997): a much higher nutrient load and higher eutrophic state are the result. This can be documented by increasing Di-TP values in Lake Dudinghausen (Dreßler et al. 2007). In contrast, the

nutrient accumulation in the lake sediments did not increase during this period. C, N and P show no variation in these sediment layers.

Surprisingly, the Fe/Mn and Fe/Ca ratios did not increase after the war. Perhaps the advanced trophic state led to more pronounced oxygen depletion above the sediment (temporally and spatially) at this time. Thus, both Mn and Fe were equally released from the sediment into the water, leading to stable Fe/Mn and Fe/Ca ratios in the sediments.

In accordance with the pre-war period, the Pb and Zn concentrations increased permanently. The highest concentrations occurred between 1970 and 1980. These heavy metals can be reliably used to reconstruct regional trends of atmospheric deposition (both lakes have a small catchment area and lack industrial production sites). Their increase in the sediment cores mainly reflects atmospheric deposition linked to fossil fuel combustion. A comparison with other North and Central European studies shows similar patterns for both heavy metals (Goslar et al. 1999; Olsson et al. 1997). In contrast to the Pb and Zn concentration, Cu did not increase during this period.

Time period after 1985: period of reunification

The upper 10 cm of surface sediment represents the deposition of approximately the last 20 years in both lakes. The highest TOC, TN and P concentrations in both lakes occurred in the surface sediment layer. All three parameters steadily increased in the upper 10 cm, and the C:P ratio increased. Psenner et al. (1984) proposed to subtract the P

concentration from the deeper, permanently anoxic sediment layers (as a stable background concentration) to describe the anthropogenic P accumulation in the sediment. Based on this definition, $1 \text{ mg P g}^{-1} \text{ dw}$ is the background concentration of both lakes. Accordingly, more than 50% of the P in the upper sediment layers is the result of anthropogenic use of the catchment area. Kemp et al. (1976) described lake productivity based on the parameter TOC and P in sediments. The diatom-inferred TP function revealed an increased P concentration in the water body of both lakes with the highest value being $80 \mu\text{g L}^{-1}$ (Lake Dudinghausen—Dreßler et al. 2007) and $140 \mu\text{g L}^{-1}$ (Lake Tiefer—Selig et al. 2002) from 1980–1985.

Selig et al. (2005) described phosphorus binding using fractionation techniques in the upper 10 cm sediment layer. The redox-sensitive P was higher in the upper 1 cm in both lakes. In Lake Dudinghausen, 15% of the extractable P was redox-sensitive bound P, whereas only 4% of the extractable P was determined in the redox-sensitive fraction in Lake Tiefer. Despite the recent anoxic conditions during summer stratification, the redox-sensitive elements (P, Fe, Mn) differed notably in the two lakes. Among these elements the correlation was low only between P and Mn in Lake Dudinghausen, whereas the other elements showed no correlations. A high correlation between P and Fe is typical only for oxic sediment layers (Einsele 1936). Selig and Schlungbaum (2003) investigated the temporary variation of P binding in the upper 2 cm sediment under changing to anoxic conditions during summer stratification in both lakes. The calculated benthic P release was higher in Lake Dudinghausen, and the P binding changed in both lakes to a sediment depth of 2 cm. The present results reflected an oxic situation (sampling in spring before summer stratification) on the sediment-water interface and therefore a maximum P binding in the upper sediment.

Pb and Zn concentrations have decreased in the last 20 years. The post-1990 decline of Pb is a response to Pb-pollution control measures on leaded gasoline that began in East Germany with the reunification of Germany. Such an effect has also been described in other countries after introduction of unleaded gasoline (Dixit et al. 2000). The higher concentration of Pb and Zn in Lake Tiefer versus Lake Dudinghausen can be explained its proximity to the city Güstrow and to the highway. Gallagher et al. (2004) documented a clear influence of regional sources on the Pb concentrations in six lakes in British Columbia. We observed the same effect here.

Conclusions

This study provides information about supraregional and regional impacts on the history of two lakes in North-

Eastern Germany. For both lakes, three characteristic time periods can be documented. In addition to agricultural activities in the catchment area, atmospheric pollution greatly influenced the metal concentration in the sediment.

In contrast to the investigation of the diatom-inferred TP, TOC, TN and P concentrations in the sediment layers could not describe the variation of trophic level in either lake. In both lakes, nutrients accumulated in the upper 10 cm—representing the last 20 years—whereas the Di-TP decreased.

Increased eutrophication—documented based on the diatom TP transfer function—explains the altered redox conditions at the surface sediment. The Fe/Mn and Fe/Ca ratios documented a variation in the redox conditions at the water-sediment interface; they suggest seasonal anoxia at greater depths in the water above the sediment since approximately 1920–1930 in both lakes.

The increased atmospheric pollution with heavy metals here is compared with the situation in Central Europe. The strong decrease of Pb and Zn in the last 10 years reflects a regional effect and the new political situation in North-Eastern Germany.

Acknowledgments We gratefully thank Dr. Helmut Erlenkeuser, Leibniz Laboratory for Radiometric Research of the University Kiel, for analysis of ^{210}Pb and ^{137}CS activity profiles. The Ministry of Environmental Mecklenburg-Vorpommern, Germany, supported this study.

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