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Heavy metal content in sediments of lakes in southern Bavaria as a sign of long-term environmental impact

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ABSTRACT The great age of lake sediment permits a reasonable estimation of the extent, distribution and origin of heavy metal contamination caused by man. Cores have been analysed from 25 lakes in southern Bavaria and reflect the history of sedimentation over periods of up to 15 000 years. The first signs of change in profiles of heavy metal concentrations mostly occur at a depth of 0.5 to 2.0 m below the sediment surface, are generally dated as thirteenth century and are associated with large-scale deforestation. Increases in zinc associated with the mining and use of coal start after the eighteenth century, whereas definite increases in lead and cadmium are apparent after the beginning of the twentieth century and two to three decades later, respectively. Although heavy metal concentrations in most lake sediments have not reached toxic levels, the impact of heavy metal pollution has been very strong over the last 1500 years and has even caused contamination in lakes remote from centres of population and industry. The increase in heavy metal concentrations has been most alarming for artificial lakes, such as the "Speichersee", which is affected by sewage effluent from Munich. The information provided by the cores on bioproduction and palaeoclimatic and palaeolimnic conditions is also discussed.

Teneur en métaux lourds des sédiments des lacs du sud de la Bavière: indicateur de l'impact à long terme de l'environnement
RESUME La grand âge des sédiments dans les lacs permet une estimation convenable de l'étendue, de la répartition et de l'origine de la contamination en métaux lourds provoquée par l'homme. Des carottes on été analysées pour 25 lacs dans le sud de la Bavière et l'ensemble des analyses faites sur ces échantillons reflête l'histoire de la sédimentation sur des périodes qui vont jusqu'à 15 000 ans. Les premiers signes de modification dans les profils de concentration en métaux lourds surviennent surtout à une profondeur de 0.5 à 2.0 m sous la surface des sédiments, ils sont en général datés du treizième siècle et sont associés à l'abattage des forêts sur une large échelle. L'augmentation de la concentration en zinc associé avec l'exploitation des mines et l'utilisation du charbon commence après le dix huitième siècle, alors que

les accroîssements des teneurs en plomb et en cadmium apparaissent respectivement après le début du vingtième siècle et deux ou trois décennies plus tard. Quoique les fortes concentrations en métaux lourds dans les sédiments de la plupart des lacs n'ont pas atteint un niveau toxique, l'impact de la pollution par ces métaux lourds a été très marqué au cours des 1500 dernières années et a même donné lieu à la contamination dans des lacs loin de tous centres habités ou industriels. L'augmentation de la concentration en métaux lourds a été le plus alarmant pour des lacs artificiels tels que le "Speichersee" qui est affecté par les effluents de la ville de Munich. On traite aussi des informations fournies par les carottes sur la bioproduction, les conditions paléoclimatiques et paléolimnologiques.

INTRODUCTION

The limnic sedimentation basins of lakes represent, on a geological timescale, only a short period of sedimentation, but the period of more or less undisturbed sedimentation of lake sediment is sufficient to reach interesting conclusions concerning palaeoecological, palaeolimnic and environmentally relevant facts on the basis of a series of chemical and physical parameters of the sediments.

METHODS

The possibility of taking sediment cores from lakes which reach as far as possible into the deposits and cover the longest period of the sedimentation history of the lake is of importance for the investigation of palaeoecological problems. Using apparatus developed by Züllig (but considerably modified) and provided by the DFG, it has been possible for the Institute of Geography of the University of Munich to remove sediment cores of up to 6 m length from 25 southern Bavarian and Alpine lakes. From these lake cores, in which little allochthonous sediment has been deposited, it is possible to obtain information concerning the Lateglacial period and the postglacial history of sedimentation.

The age of the individual strata can be determined with sufficient accuracy using various methods. If the cores show a regular alternation of annual strata, it is possible to give a rough estimation of the age on the basis of these "varves". If sufficient organic carbon is present in the sediment, a radiocarbon dating can be undertaken to give a comparatively accurate estimate of its age. Unfortunately, the so-called "hard water effect" occurs due to the high amounts of dissolved carbonate contained in the water of the prealpine regions. Algae, which are deposited later, also incorporate into their organic material during photosynthesis the geologically older carbon from soluble bicarbonate which contains low amounts of ^{14}C, so that the radiocarbon dating method results in an increase of up to 20% in the age. Another method of dating is based on the use of pollen analysis which has been used with great success in the dating of turf. In the case of the most recent lake deposits (after 1945) it is possible to use radionucleides (e.g. ^{137}Cs with its maximum in 1963). The most successful methods make use of events for which exact data are available and which have generated clear changes (e.g. in particle size or in the chemical-biological composition) in the sediment (e.g. the large scale changes caused by large man-made changes in a drainage area, such as the canalization of the Isar into Walchensee in 1923).

HEAVY METAL IN LAKE SEDIMENTS AS AN INDICATOR OF MODERN CIVILIZATION

The great age of lake sediment in comparison with the history of man makes it possible to give a plausible estimation of the extent, the distribution and the origin of the heavy metal contamination caused by man directly by their use or by their release in the drainage area of the respective lake and/or in a wider area in the case of transport in the atmosphere. A knowledge of pre-civilization heavy metal accumulation is necessary in order to obtain the extent of heavy metal emission in the civilization period. The cadmium, lead, copper, zinc and chromium contents of the sediment core from Lake Ammer (Ammersee, Upper Bavaria, southeast of Augsburg) is shown as an example (Fig.1). The length of the core is 382 cm and

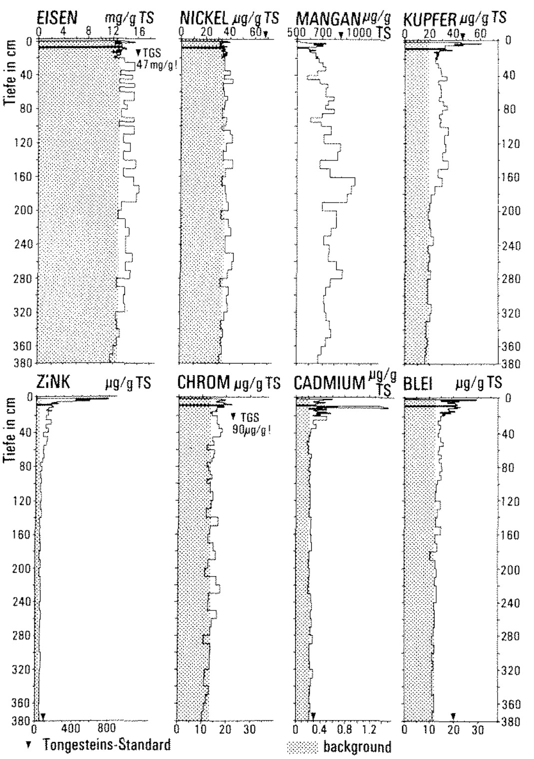


FIG.1 Content of iron, nickel, manganese, copper, zinc, chromium, cadmium and lead in a sediment core from Lake Ammer (Ammersee, Upper Bavaria).

has been shown with the help of varve dating to cover a period of 1500 years. The deeper layers have a practically undisturbed profile of very low concentrations which represent the natural background levels of the Lake Ammer drainage area (988 km^2). The first signs of change in the concentration profile start at a depth of about 190 cm (dated by varves as thirteenth century) and can be related to the large-scale deforestation in the Lake Ammer drainage area (with resulting increase of denudation) undertaken by the monasteries in Wessobrunn and Polling. At the same time there was an increase in the local pottery and woodwork industries, which used heavy metal colouring agents, in Murnau, Weilheim and the Ettal monastery (founded in 1330).

After the eighteenth century it is possible to see the first signs of an increase in zinc, which can be connected with the start of coal mining in Peissenberg and Peiting (Upper Bavaria). From 1818 to 1970 the population figures for the Lake Ammer drainage basin rose from 25 000 to 105 000. At the same time the number of heavy metal manufacturing businesses increased including some galvanizing and light metal firms (for example aircraft manufacture in the Second World War and the associated use of paints containing cadmium for camouflage). This effect was reinforced by the increased emission of zinc, lead and cadmium from the coal mines and/or from the domestic use of coal (airborne ash) and recently from the high consumption of oil products. A strong correlation can be seen between the heavy metal content of sediments, for example in the Baltic Sea, and the industrial development in central Europe. In spite of the fact that the heavy metal concentrations in Lake Ammer sediment have not yet reached the toxic level and the concentrations are in general below that of the "clay standard" (Tongesteinsstandard) with its natural content of heavy metals (the standard for carbonates in the Northern Limestone Alps is generally below the international Tongesteinsstandard) an increase of up to 20 times (in the case of zinc) over background levels in a period of 1500 years is apparent and indicates a strong impact of heavy metal pollution in the drainage area. The geo-accumulation-index of German Müller (1979) has been calculated for various southern Bavarian lakes (Table 1). Even in the lakes far from industrial areas and population centres (e.g. Grosser Arbersee, Riegsee, Steinsee, Wesslinger See, Osterseen) without the presence of immediate heavy metal emission, an increase in lead in the upper layer of sediment is considerable and points to a constant transport of heavy metal into these lakes from the atmosphere. The situation for cadmium is similar although not quite so serious. The increase in heavy metals in many of the artificial or strongly man-influenced lakes is alarming. The 50-year-old reservoir ("Speichersee") in the northeast of Munich, through which 80% of the Munich sewage flow, has the highest concentration of heavy metals in those areas where the finest suspended particles are deposited (Fig.2). These high concentrations are an indication of the heavy metal pollution from the Munich environment (see "Munich Sewage Scandal" of 1978, Koch & Varenholt, 1983). Although these highly toxic heavy metals are temporarily fixed in the sediment and appear to be harmless, the organisms living in the ground can incorporate them in the food web, they can be taken up by the water during floods or can be remobilized from the sediment under anaerobic conditions (especially Fe, Mn).

TABLE 1 The maximum and minimum concentration (ppm) and the I_{geo}-value in some lakes of south Bavaria

See	Cu			Pb			Cr			Cd			Zn		
	Max	Min	Igeo	Max	Min	Igeo	Max	Min	Igeo	Max	Min	Igeo	Max	Min	Igeo
Ammersee	60	20	1,0	30	12	0,7	22	14	0,06	1,5	0,21	2,2	800	50	3,4
Wörthsee	65	40	0,1	125	18	2,2	22	7	0,5	1,2	0,95	1,4	340	50	2,2
Pilsensee				46	5	2,5	19	3	2,1	0,1	0,01	1,5	55	13	1,5
Walchensee	24	6,9	1,2	11,5	1	3,8	43	6	2,2	0,6	0,34	0,1	105	18	1,9
Kochelsee				310	50	2,0							480	50	2,6
Gr. Ostersee	33	30	-0,4	60	8	2,3				0,5	0,10	1,6	30	20	0
Fohnsee	36	27	-0,2	49	14	1,2				0,9	0,20	1,5	125	30	1,5
Waschsee	81	42	0,4	68	12	1,9				1,4	0,30	1,6	520	100	1,8
Herrensee	28	32	-0,7	80	3	4,1				0,6	0,30	0,3	60	100	-1,3
Sengsee	50	30	0,2	44	11	1,4				1,2	0,30	1,4	110	30	1,3
Gr. Arbersee	27	32	-0,8	52	8	2,1	0,3	0,4	-0,9	0,4	0,28	-0	82	70	-0,2
Weßlinger See	70	60	-0,4	244	23	2,8	13	20	-1,2	1,7	0,50	1,1	308	27	2,9
Steinsee															
Alpsee															
Alatsee				in Bearbeitung											
Riegsee															
Höllensteinsee	80	"	0,7	155	"	3,7	80	"	3,3	5,9	"	3,8	420	"	2,0
Speichersee	420	x	3,8	2100	x	6,8	830	x	5,3	100	x	8,2	2500	x	5,0
Ausgl.-Weiher	123	x	2,1	609	x	5,0				31	x	6,4	259	x	2,1
Isar/Eching	203	x	2,8	685	x	5,2				30	x	6,4	243	x	2,0
N.-aichbach	142	x	2,3	1950	x	6,7				23	x	6,0	765	x	3,6

" Background aus Gr. Arbersee, da der Schwarze Regen im Gr. Arbersee entspringt
x Background Ammersee, da Background-Werte aus dem Isarbereich bislang fehlen

Igeo = < 0 praktisch unbelastet
0-1 unbelastet - mäßig belastet
1-2 mäßig belastet
2-3 mäßig - stark belastet
3-4 stark belastet
4-5 stark - übermäßig belastet
> 5 übermäßig belastet

An interesting example concerns Lake Walchen (Walchensee), which is the largest of the German Alpine lakes and has been used since 1924 for the production of hydroelectricity. The fact that the natural drainage area of 74 km^2 was too small to provide the necessary water for the effective functioning of the hydroelectric scheme (Walchensee-Kraftwerk) led to the canalization of the Upper Isar in 1924 and the Rissbach in 1949 which respectively provided 13 and 8 $m^3 s^{-1}$ runoff into the Walchensee and increased its drainage area to 770 km^2. If the change in level of the concentration of selected heavy metals is plotted against increasing depth, it becomes clear, that canalization corresponds with a sudden decrease in the heavy metal concentration (Fig.3). However, if the increase in the rate of sedimentation of about 10 times, which was caused by the canalization of the Isar, is taken into account a completely different picture of the trends in zinc deposition emerges. Before the canalization, approximately 20 mg $m^{-2} year^{-1}$ of zinc were deposited (Fig.4), but in 1924 an increase of up to 25 times this amount was registered. In the following years and up until 1950 values for the sedimentation ranged between 260 and 300 mg $m^{-2} year^{-1}$,

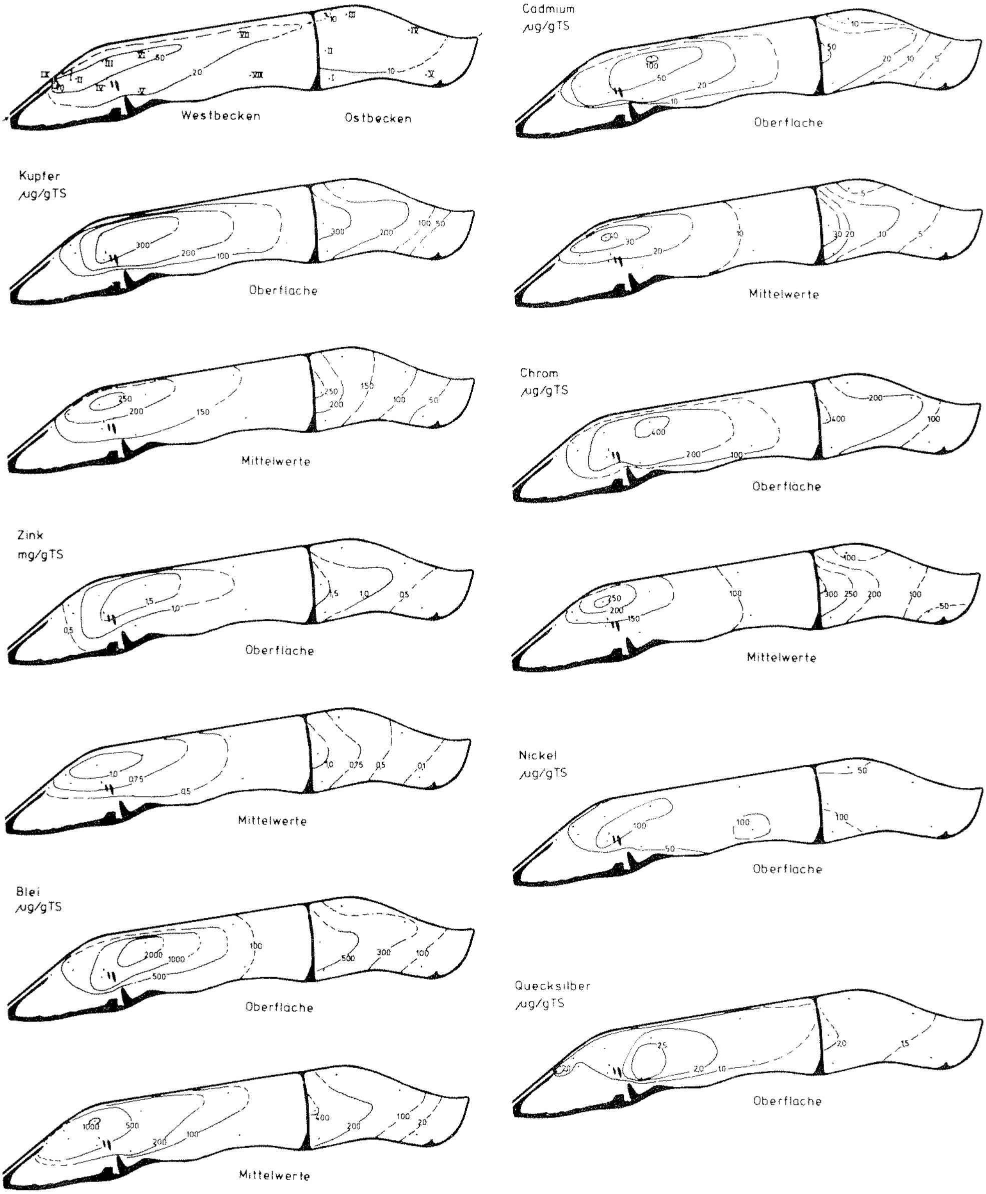


FIG.2 Distribution pattern of heavy metals in the sediment of the "Speichersee" northeast of Munich, surface sample (above), mean value of whole core (below).

but rates of deposition have more than doubled in the period since 1950 and the present day. The other heavy metals that have been investigated show a similar picture. The isolated maximum in 1924 also occurs for nickel, cadmium, chromium and calcium and appears to indicate a washing out of the newly constructed canal.

Long-term transport of heavy metals emitted by anthropogenic processes in the form of dust in the atmosphere is not only found

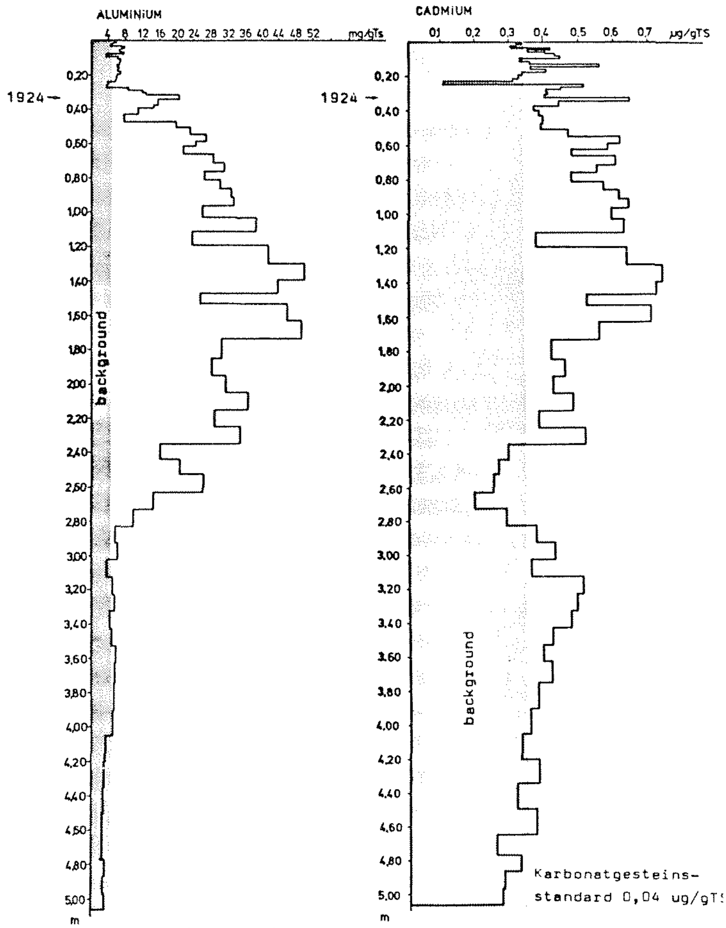


FIG.3 Vertical distribution of aluminium and cadmium in Lake Walchen (Walchensee, Upper Bavaria).

in lakes near to industrial centres. The geochemical investigation of a sediment core from the Arctic Access Lake (Müller & Barsch, 1980) also shows an increase in heavy metal content, in particular of lead and zinc after 1900. The lead concentration increased from 2.9 to 10.3 ppm, that of zinc from 40 to 80 ppm and that of cadmium from 0.11 to 0.26 ppm. According to Brunner & Baccini (1981) a large part of the heavy metal transported in the atmosphere from the west is washed out over the Alps.

For the investigation area (Southern Bavaria and the Alps) information on the following is desirable:

(a) the separation in sediments of heavy metals derived from local and more distant sources,

(b) the analysis of the origin (position) of the distant emissions,

(c) quantification of the specific characteristics of lakes regarding the deposition of heavy metals.

Thomas (1981) has taken similar problems into account in the investigation of the increase of heavy metals and various organic pesticides, etc. in his work on recent mosses. It is not yet possible to say, to what extent progress can be made by the investigation of lake sediments in view of the very complicated processes affecting sedimentation in lakes.

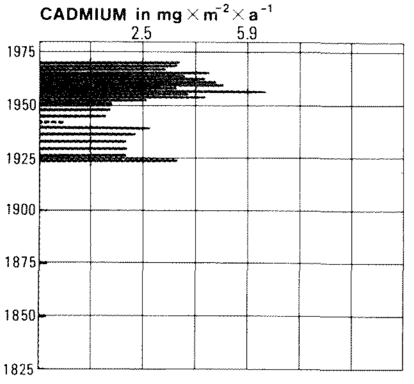


FIG.4 Heavy metal concentrations in Lake Walchen, recalculated as sedimentation rate per square metre per year and using cadmium as a selected example.

BIOPRODUCTION

With the help of an absolute dating of the age of sediment cores it is also possible to derive conclusions concerning the development of the bioproduction and/or eutrophication. The bioproduction in Great Arber Lake (Grosser Arbersee) evidences an increase at 10 000 years BP on the basis of the greater amount of organic matter in the sediment. The maximum organic deposition took place between 4500 and 5000 years BP, i.e. at the change from the warmer Atlantic to the cooler Subboreal period. It is also possible that an increased transport of suspended organic matter (humic substances) took place at this time from the vegetation cover of the drainage area, which in general showed higher metabolism than today, particularly in these higher regions. Also the change in the facies between the Late- and Postglacial around 10 000 years BP can be determined at a depth between 3 and 5 m for all those lakes in which the autochthonous deposition from the water body is greater than the allochthonous inflow of suspended material (i.e. Lake Walchen, Steinsee, Riegsee, Lake Wessling). In addition, increased organic deposition which occurred at the climatic optimum and a more recent general increase in the rate of deposition, especially for organic material, suggest the occurrence of eutrophication.

Chlorophyll a, the pigment contained in green plants, can be used with a certain element of error as an indirect measure both of the production of phytoplankton and also for the production of the biomass (Vollenweider, 1979). Phaeopigments are decomposition products of chlorophyll. Since chlorophyll can also be chemically decomposed in a dry condition, only the sum of the amounts of

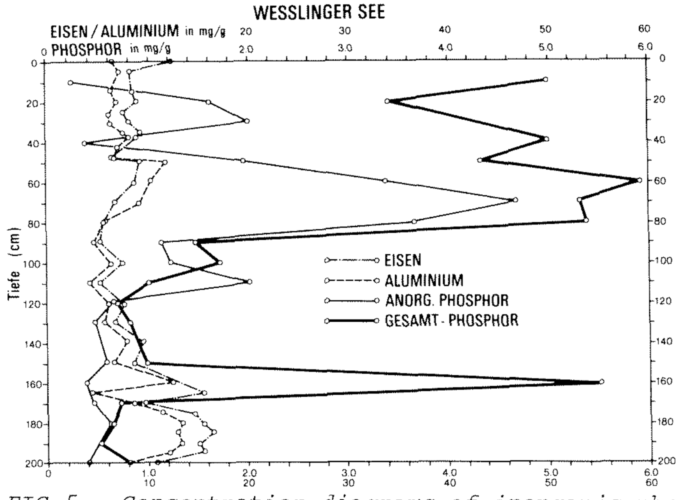
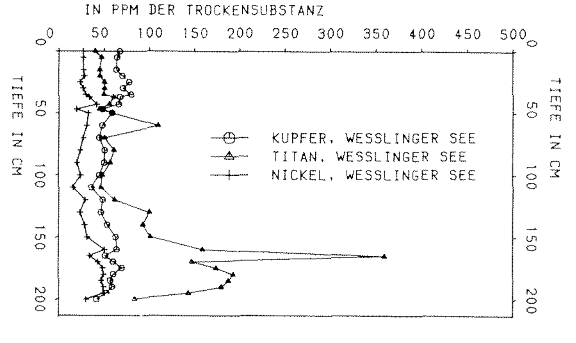


FIG.5 Concentration diagrams of inorganic phosphorus, total phosphorus, iron and aluminium in Lake Wessling (Wesslingersee, Upper Bavaria).

(a)



(b)

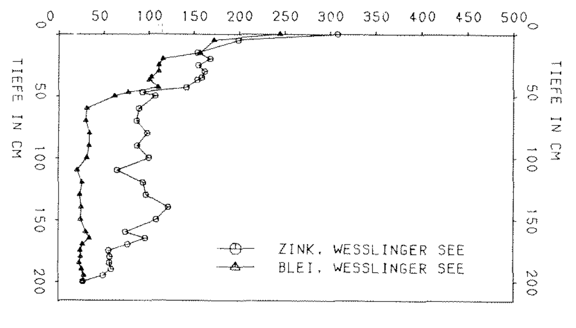


FIG.6 The concentration of copper, titanium, nickel, zinc and lead in Lake Wessling.

chlorophyll a and phaeopigments are usually shown in a distribution diagram. In such distribution diagrams (e.g. for Lake Wessling) it is possible to see a definite increase in the upper layers. From 1969 the amount decreases, which is possibly the result of the newly built drainage system. A further clear indication of eutrophic processes can be deduced from the increase in the amount of organic phosphorus (Fig.5). The increase coincides with a sudden rise in the population of Wessling after 1945. A similar increase in the metals copper, zinc, lead and cadmium can also be considered to be caused by the influence of modern civilization (Fig.6). The cause of the decrease in the content of phosphorus from the 40 cm layer to the surface of the sediment is presumably correlated with the remobilizing of substances from the sediment under anaerobic conditions, which are caused by the high bioproduction in Lake Wessling. Decomposition processes consume the oxygen in the lower hypolimnion in a short time, and the contact layer between sediment and water becomes free of oxygen and contains H_2S shortly after the beginning of the summer stagnation. The heavy metals iron and manganese are in the same way redissolved in reducing conditions. The identical impoverished layer that appears in all three distribut-

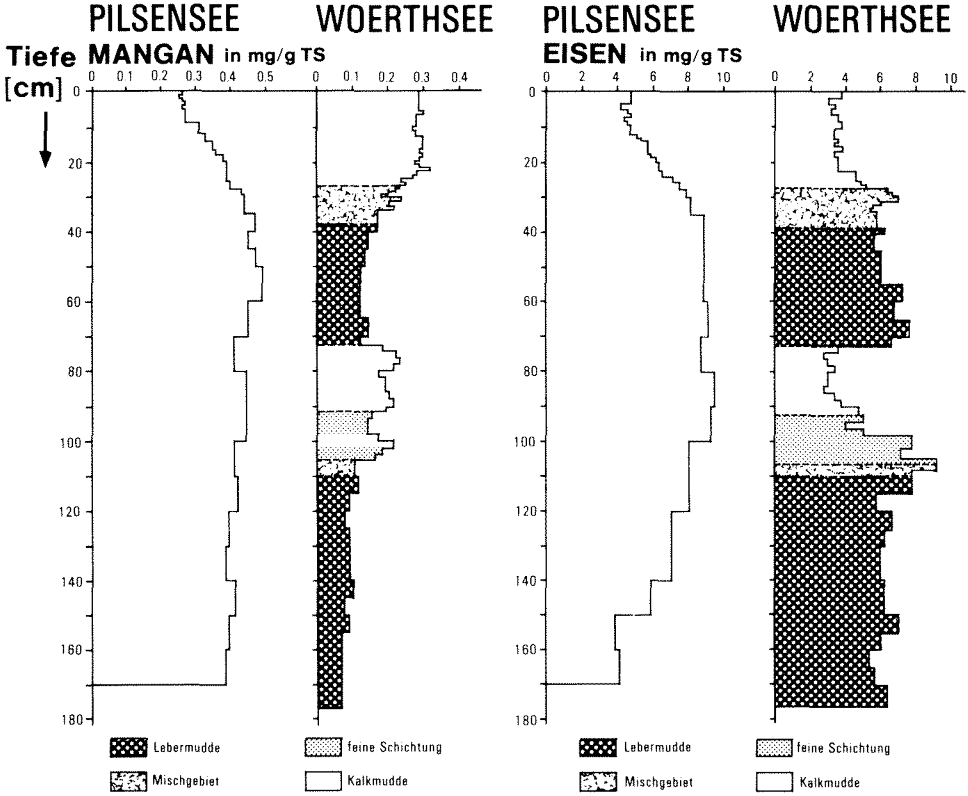


FIG.7 Concentration diagram of manganese and iron in Pilsensee and Wörthsee (Upper Bavaria).

ion diagrams (phosphorus, iron and manganese) is almost certainly caused by a similar dissolving of these three elements under anaerobic conditions.

Solution of the iron and manganese in the sediment, which is dependent on the redox potential, shows a correlation (Fig.7) with the sediment cores from Lake Pilsen (polytrophic, but during the period of summer stagnation oxygen is absent below *c.* 5 m depth) and from Lake Wörth (eutrophic, but with some oxygen in the hypolimnion during the stagnation). While in Lake Pilsen a reduction of iron and manganese in the upper sediment layers can be seen, this is not present in Lake Wörth. If the beginning of this decrease in Lake Pilsen could be dated, it would be possible to come to conclusions about when this lake began to be free of oxygen in summer and thus showed polytrophic conditions.

PALAEOCLIMATIC AND PALAEOLIMNIC CONCLUSIONS

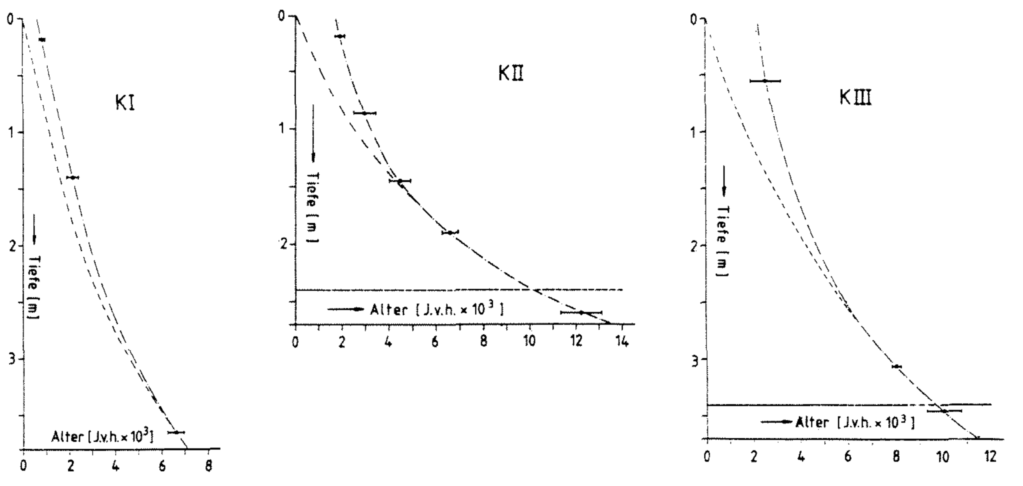
With the help of ^{2}H, ^{13}C, and ^{14}C-isotope analysis it is possible to carry out research on a series of other palaeoecological questions, as can be shown in the case of the three sediment cores from the Great Lake Arber (Grosser Arbersee, Bavarian Forest). The author is very grateful for the reliable radiocarbon dating of the age of the sediment core that was provided by the Institute of Radiohydrometry (M.Wolf & W.Rauert) of the "Gesellschaft für Strahlen-und Umweltforschung" in Munich/Neuherberg. These results are given in Table 2 and shown in Fig.8. The ^{14}C-model ages, that were corrected with the help of ^{13}C, in comparison with Suess (1970), Stuiver (1970) and Barbetti are 800 years too young. There is good correlation between the ^{14}C-ages of the older sections of the cores and the ages obtained using pollen analysis, but in the younger sections the ^{14}C-ages are systematically greater. This could be a result, for example, of the mixing of relatively soft (water content up to 94%) upper layers of sediment with older layers, or due to the addition of older humic substances from the drainage area to the sediment. The effect of hard water can, however, be discounted due to the crystalline drainage area. For the same reason, the possibility of isotope exchange processes with the dissolved CO_2 and/or through methane production seems to be unlikely. Another indication that ^{14}C-ages around 10 000 years BP are correct is the correspondence of the age curves of K II and K III with the change in facies from the Lateglacial to the Postglacial, that was dated at 10 300 ± 300 years BP in Scandinavia and obviously falls within the period 9600-10 000 years BP in the Great Lake Arber and also in other lakes of southern Bavaria. The change in facies in the Great Lake Arber is indicated by a relatively sudden change from the grey to light brown and more sandy and clayey muds of the Lateglacial to the black brown, strongly organic muds (Mudden) of the Postglacial (Fig.9). In Fig.10 the ^{13}C-measurement results of K I, K II and K III and also ^{2}H-measurement results of K II are plotted against the ^{14}C-ages of the sediment samples. A comparison of the $\delta^{13}C$-values of the three cores shows,

TABLE 2 Radiocarbon dating of a sediment core from Lake Arber (Grosser Arbersee)

Tiefe (cm)	Proben-Nr.	$\delta^{13}C$ (‰)	$\delta^{2}H$ (‰)	korr. ^{14}C-Alter (J.v.h.x10^3)
0 bis 10	29	-27,71	-119,4	1,8
50 bis 60	24	-28,05	-119,0	2,4
100 bis 110	19	-28,05	-115,9	3,3
160 bis 170	13	-27,68	-112,9	5,3
210 bis 220	8	-28,30	-118,3	8,2
240 bis 247,5	5	-28,67	-120,3	10,5
247,5 bis 255	4	-27,44	-107,2	11,3
255 bis 260	3	-27,76	-103,3	12,0
260 bis 265	2	-26,43	-101,6	12,5
265 bis 270	1	-27,10	-104,9	13,2

$\delta^{13}C$- und $\delta^{2}H$-Werte von Sedimentproben (Einzelproben) des Kerns K II mit jeweiliger Tiefe im Sedimentkern (von oben) und dem zugehörigen korrigierten ^{14}C-Alter

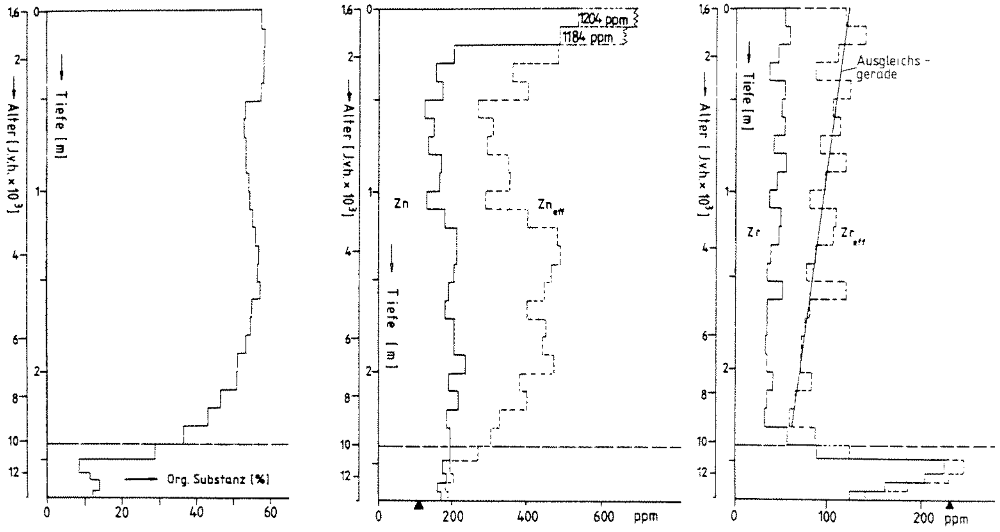
for each of the cores, more or less constant ^{13}C-values in the section between 2000 and 5000 years BP. With increasing distance of the position of the core from the inflow, a change in the $\delta^{13}C$-values to a lower level was apparent. This effect could be explained by a



Korrigierte ^{14}C-Alter der drei Sedimentkerne K I, K II und K III in Abhängigkeit von der Tiefe unter der Sedimentoberfläche

- ├──●──┤ korrigierte ^{14}C-Alter mit zweifacher Standardabweichung
- —·—·— aufgrund der ^{14}C-Altersdaten möglicher Verlauf der Alterskurve
- — — — möglicher Verlauf der Alterskurve bei Annahme eines "Huminstoffeffektes" und/oder ähnlicher Effekte
- ———— Faziesgrenze Spätglazial/Postglazial in den datierten Sedimentkernen

FIG.8 Corrected ^{13}C-ages of three sediment cores from Lake Arber (Grosser Arbersee).



Gehalt (in Gew.-%) an organischer Substanz, Zn und Zr im Sediment des Kerns K II, aufgetragen gegen die Tiefe unter der Sedimentoberfläche bzw. gegen korrigierte ^{14}C-Alter. Durchgezogene Linie: Gehalt, bezogen auf Trockensubstanz; gestrichelte Linie: Gehalt, bezogen auf Glückrückstand. ▲ "Moränenstandard" (mittlerer Zn- bzw. Zr-Gehalt im Moränenmaterial).

FIG.9 Organic matter content (%) in core II from Lake Arber (Grosser Arbersee) and the concentration diagrams of zircon and zinc.

lower content of organic substance originating from land plants (for the area examined the usual vegetation showed values of $\delta^{13}C$ of *c.* -27‰, e.g. Osmund & Ziegler, 1975) and a higher content of limnic plankton ($\delta^{13}C$ *c.* -30‰, Pearson & Coplen, 1978) at greater distances from the inflow. A comparison of the ^{2}H- and ^{13}C-age profiles of K II shows an almost similar profile for the content of both isotopes above the age of 20 000 years BP. Within the Postglacial both isotope profiles demonstrate a single maximum (shown however in each case by only one sample) at about 5000 years BP. This is possibly connected with the climatic optimum. For the ^{14}C-ages above 11 000 years BP, the $\delta^{2}H$- and $\delta^{13}C$- values rise appreciably. The combustable material of the samples shows a carbon content of approximately 33%, while the corresponding content of the postglacial samples was found, as expected, to be 50%. The lower carbon content is possibly due to fermentation processes. For the chemical reactions in organic materials (turf) Schiegl (1970) determined an empirical relationship between the $\delta^{2}H$ values and the carbon content between 50% and 65% carbon content. If one presumes that this relationship can also be applied to the samples of the Great Lake Arber with their lower content, then a reduction of about -115‰ must be taken into account for $\delta^{2}H$- values for samples of muds with a ^{14}C-age of over 11 000 years BP. Using the $\delta^{2}H$-temperature correlation from Schiegl (1970), this would result in an increase in temperature from the Lateglacial to the Postglacial of about 10°C, which is in fact a plausible value (e.g. Rudloff, 1980). Further research is necessary for the confirmation of these findings and, in particular, for the interpretation of the $\delta^{13}C$

values in the Lateglacial.

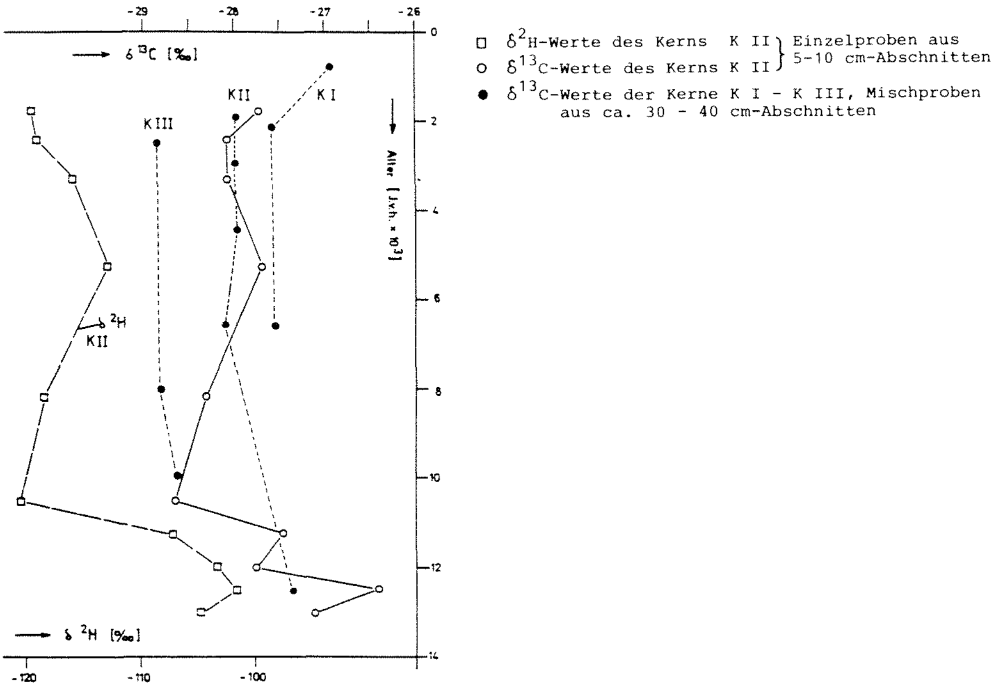


FIG.10 δ^2H- and $\delta^{13}C$- values of sediment samples from Lake Arber (Grosser Arbersee, Bavarian Forest).

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