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PARTIAL RECOVERY OF MOORLAND POOLS FROM ACIDIFICATION: INDICATIONS BY CHEMISTRY AND DIATOMS

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KEYWORDS: reversibility, acidification, diatoms, chemistry, sulphur deposition, moorland pools, The Netherlands.

ABSTRACT

In 1994 the atmospheric deposition on three moorland pools in The Netherlands was only one third of its value in 1980. The effects of this reduction on these moorland pool ecosystems were examined by regular sampling of surface water chemistry and diatoms between 1979 and 1994. Moreover, diatom samples taken at irregular intervals between 1916 and 1978 were studied. In the pools Gerritsfles and Achterste Goorven, the median sulphate concentrations in 1994 were only one third of recorded values in 1980. Peak concentrations of sulphate were found after extremely dry summers. In the pool Kliplo, with an initially low concentration of sulphate, there was 16% increase of the concentration over the same period. Ammonium increased significantly in Kliplo and showed large variations in the other pools. Redundancy analysis showed that sulphate was the most important variable for the distribution of diatoms. As a consequence, the abundance of the acidification indicator *Eunotia exigua* in 1994 was only 25% of its value in 1980 in Achterste Goorven and 14% in Gerritsfles. Peaks of *E. exigua* were found after drought periods. In Kliplo no clear change was observed. In 1994 the diatom assemblages of Achterste Goorven were much more similar to those of 1916-1925 than they were in 1980. In Gerritsfles a new situation, without historical analogue, developed. Overall, the large reduction of SO_x-deposition had very positive consequences for the diatom assemblages.

INTRODUCTION

The deleterious effects of acid atmosperic deposition on soft-water ecosystems are well known (*e.g.* LAST and WATLING, 1991). In The Netherlands and adjacent countries, moorland pool ecosystems are very vulnerable to all kinds of human activities, have a high proportion of threatened plants and animals and are important objects for nature conservation (VERKAAR *et al.*, 1992; BAL *et al.*, 1995; LNV, 1995). The biota of these shallow moorland pools, particularly rare species of aquatic macrophytes (isoetids) and diatoms, are threatened by acidification, as reviewed by VAN DAM and BUSKENS (1993).

For the protection of these and other ecosystems, measures have been taken to reduce immissions of nitrogen and, particularly, sulphur compounds in The Netherlands (NIJPELS, 1987). This has resulted in an average reduction of the total deposition of sulphur compounds from 1.6 kmol ha^{-1} y^{-1} in 1980 to 0.7 kmol ha^{-1} y^{-1} in 1989 (ERISMAN, 1992).

It is the purpose of the present paper to describe the effects of these measures on the chemistry and biota, particularly diatoms, of three selected moorland pools which were regularly sampled before and after reduction of the deposition. Diatoms were chosen as biological indicators as they have very strong relationships with acidification and are often used in acidification studies as biological indicators (VAN DAM, 1988; CHARLES *et al.*, 1989; BATTARBEE, 1994).

The research was part of an interdisciplinary study, including deposition and analysis with hydrological and ecological models (see VAN DAM *et al.*, 1996).

STUDY SITES

Five stations were studied in three pools. Kliplo (one station) was relatively unaffected by acid precipitation when our observations started, Achterste Goorven (three stations) was very seriously affected, and Gerritsfles (one station) had an intermediate position. Together, these pools are rather representative for acidifying moorland pools in The Netherlands. All of these pools are situated in nature reserves and are relatively undisturbed by human activities. The landscape development, paleolimnology, hydrology, chemistry, biology and acidification history of these pools are well known (VAN DAM, 1988; VAN DAM and BUSKENS, 1993) and are only briefly described here.

Achterste Goorven (area 2.4 ha, mean depth 0.6 m) is situated in the southern Netherlands. This long, narrow pool is surrounded by aeolian drift sands, mainly planted with Scottisch pines. In extremely dry years, like 1976, over 70% of the pool may dry out. The amplitude of the water level over the period of observation was 0.8 m. The pool is partially fed by groundwater and drains at the western side to Voorste Goorven with a small outlet. In the 19th century, Achterste Goorven was probably enriched with agricultural drainage water via Voorste Goorven. *Nymphaea alba* is the most conspicuous aquatic macrophyte. *Juncus bulbosus* is rather abundant: its development was particularly vigorous after the drought of 1976. In the early twentieth century this species was much less abundant, while soft-water macrophytes were much more abundant. Achterste Goorven is oligotrophic (median chlorophyll *a* 10.7 µg l^{-1}). The three sampling stations AGA, AGB and AGE are in a gradient from west to east and denoted as Goorven 1, 2 and 3 respectively in Fig. 1 of VAN DAM (1988).

Gerritsfles (area 6.0 ha, mean depth 0.7 m) is situated in the centre of The Netherlands. The pool borders aeolian drift sands and podzols in a relatively open landscape of purple moor-grasslands, partially overgrown with Scotisch pines, birches and shrubs. The pool has a perched water table and a very small catchment area. The amplitude of the water level is 0.5 m. The pool is permanently filled with water, although half of its bottom desiccates in extremely dry years. In the second half of the nineteenth century, the pool was used for sheep dipping and in the first half of the present century for bathing. The bottom of the open water is nearly totally covered by the moss *Sphagnum inundatum*. The abundance of *Juncus bulbosus* has increased considerably since the beginning of this century, when soft-water macrophytes were still present. Gerritsfles is very oligotrophic (median chlorophyll *a* 1.5 µg l^{-1}).

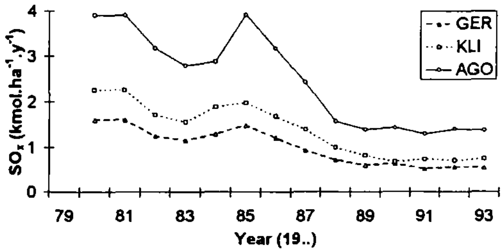

Fig. 1. Longterm variation in total deposition of SO_x at the sites Gerritsfles (GER), Kliplo (KLI), and Achterste Goorven (AGO) (after J.W. ERISMAN in VAN DAM *et al.*, 1996).

Kliplo (area 0.6 ha, mean depth 0.8 m) is located in the north of the country. The pool is mainly surrounded by aeolian drift sands, partially covered by heathland with junipers and partially with carr and open pine forest. The pool has a perched water table and a small catchment area. The amplitude of the water level is 0.55 m. The pool is permanently filled with water; even in extremely dry years less than 20% of the bottom is exposed to the atmosphere. In the nineteenth century the pool was enriched with nutrients, probably by the dipping of sheep or foddering of ducks. Later on, the pool was used for bathing. *Potamogeton natans* was very abundant until 1986, thereafter its abundance declined greatly and the open water is now almost devoid of macrophytes. Kliplo is more mesotrophic than the other two pools (median chlorophyll *a* 27.8 µg l^{-1}). Blooms of green algae may occur: a biomass as high as 364 mg m^{-3} chlorophyll *a* was measured in the summer of 1992.

Discrete measurements of atmospheric deposition at the sites are not available. Estimations were made, using measurements of wet deposition taken at stations close to the pools, wet and dry deposition taken at stations of the National Air Quality Network and from deposition models. Calculations with the ecosystem model demonstrated that there is probably considerable dry deposition of SO_x on moorland pools, but the dry deposition for NO_y and NH_x is probably negligible. The wet deposition of nitrate and ammonium between 1980 and

1993 at each of the three sites was about 0.3 and 0.7 kmol ha^{-1} y^{-1} respectively and showed no clear trend (VAN DAM *et al.*, 1996).

Deposition values for SO_x are highest for Achterste Goorven (forested landscape) and lowest for Gerritsfles (open landscape). In 1993 the deposition values were only 25-30% of their values in 1980, with the greatest decline between 1985 and 1990 (Fig. 1).

MATERIALS AND METHODS

Physics and chemistry

Samples at Achterste Goorven station E (AGE) and Gerritsfles (GER) were taken once every three months from August 1979 to December 1994. Sampling at Achterste Goorven stations A and B (AGA and AGB, respectively) was less intensive between 1980 and 1987. Kliplo (KLI) was sampled once every three months from May 1982 to December 1994.

The water level above mean sea level (MSL) was registered at each visit using a calibrated gauge and converted to the level above mean pool level. The pH field was measured nearshore. Glass electrodes for low conductivity waters (Russell or Ingold) were used from 1990 onwards.

Containers for laboratory samples were filled nearshore. The samples were kept at about their original temperature and transported to the laboratory of the Waterleidingbedrijf Midden-Nederland (WMN), where they arrived within 48 hours of sampling. Analysis of samples began immediately after arrival in the laboratory. The equipment used for the field and laboratory activities – most of the methods according to the Netherlands Normalization Institute (NEN) – have been described by VAN DAM *et al.* (1981), VAN DOBBEN *et al.* (1992), VAN DAM and ARTS (1993) and VAN DAM *et al.* (1996). Great care has been taken to ensure comparability of the measurements over the whole period, *e.g.* by analysing samples both with old and new methods over a sufficient period of time.

Alkalinity (alkc) was calculated as the difference between strong base cations and strong acid anions (SHAFFER *et al.*, 1988). Dissolved organic carbon (DOC) was measured from 1987 onwards. In almost all earlier samples, both sodium permanganate consumption and platina colour were measured. In these samples, DOC was calculated from the relationship: DOC (mmol m^{-3}) = 33.8 $KMnO_4$-consumption (mg O_2 l^{-1}) + 8.2 colour (mg Pt l^{-1} at 320 nm) + 204.4, which was assessed by regression of 127 observations from Achterste Goorven, Gerritsfles and Kliplo from 1987 to 1991 and explains 90.0% of the variation. Measurements of total inorganic carbon (TIC) were performed from 1991 onwards. Values for earlier samples were calculated by summation of CO_2 and HCO_3^-.

The laboratory results of the water analyses were carefully screened for outliers. Unexplainably extreme high or low values were removed from the data set. The concentration of organic anions was calculated from DOC according to OLIVER *et al.* (1983), with a charge density of 60 meq mol^{-1} DOC (HENRIKSEN and SEIP, 1980; BRAKKE *et al.*, 1987), and included in the sum of cations. The cation excess percentage (csur) was calculated according to csur = 100 x (Σcations - Σ anions) / (Σ cations + Σ anions). According to STUYFZAND (1989), ionic balances with $|csur| \leq 5\%$ are acceptable. However, incidentally higher values were accepted, when the valences of the ions were difficult to assess due to the abundance of organic material or iron.

Spearman rank correlation coefficients with time were calculated over the period of observation to asses long-term trends (SIEGEL, 1956). The trends were also assessed from visual inspection of the plots on each of the variables over time, because many of the variables neither linearly decreased nor increased over the period of observation.

Seasonal changes were assessed by visual inspection of graphs of physical and chemical variables plotted against time. As many of the changes were non-linear, no formal tests, *e.g.* the seasonal Kendall tau test (NEWELL, 1993), could be applied.

Diatoms

Historical samples were obtained from collections in several institutes. Recent samples were taken at irregular intervals in 1977 and 1978, and every six months from November 1979 to November 1994 in Achterste Goorven and Gerritsfles, and from May 1982 through November 1994 in Kliplo. Samples were collected by means of a plankton net (mesh width 30-40 µm), which was towed through the open water, through fields of macrophytes and carefully through the top of the mud layer on the bottom of the pools.

Methods for preparation of slides and microscopy are described by VAN DAM (1988) and VAN DAM and MERTENS (1990). In each slide, 400 valves were counted in randomly chosen fields and the percent abundance of each taxon was calculated.

From the beginning of the investigations in 1977, identifications were performed using a number of standard reference works, including HUSTEDT

(1927-1966, 1930), as listed by VAN DAM (1984). Criteria for the distinction of important taxa in the first years are given by VAN DAM *et al.* (1981). The keys by KRAMMER and LANGE-BERTALOT (1986-1991) were used immediately after their publication. However, for the identification of some *Eunotia*-, *Pinnularia*- and *Navicula*-taxa, the descriptions by ALLES *et al.* (1991), KRAMMER (1992) and KOBAYASI and NAGUMO (1988) were used, respectively.

Diatom taxonomy and nomenclature have changed continuously over the period of investigation. Therefore, the taxonomy and nomenclature in the checklist by VAN DAM *et al.* (1994) are used as a base-line in the present study, with some exceptions: *Eunotia bilunaris* var. *mucophila* Lange-Bertalot *et al.* is included in *E. bilunaris* (Ehrenberg) Mills, *E. steineckii* Petersen is included in *E. exigua* (De Brébisson) Rabenhorst, *Frustulia rhomboides* var. *crassinervia* (De Brébisson) Ross is included in *F. rhomboides* var. *saxonica* (Rabenhorst) De Toni, *Navicula heimansioides* Lange-Bertalot is included in *N. leptostriata* E.G. Jørgensen. *N. micropunctata* (Germain) Kobayasi & Nagumo is combined with *N. parasubtilissima* Kobayasi & Nagumo in the *N. micropunctata*-group.

Ecological spectra were calculated using the following groups (VAN DAM and MERTENS, 1989; DENYS and VAN STRAATEN, 1992; VAN DAM and ARTS, 1993):

- *EUN* *Eunotia exigua:* very resistant to acidification and the associated high concentration of heavy metals and aluminium;
- *ACI* trivial taxa from acid waters: wide-spread at (extremely) acid sites;
- *SOF* soft-water taxa: rarer taxa from acid sites;
- *ACH* *Achnanthes minutissima:* a eurytopic species from a wide range of freshwater habitats;
- *EUT* taxa from eutrophic waters, saprophilous taxa and other disturbance indicators.

The diatom-inferred pH (pHwa) was calculated by weighted averaging using equation 5 and deshrinking according to Table 5 in TER BRAAK and VAN DAM (1989).

The length of the gradient in the diatom assemblages was calculated with detrended correspondence analysis or DCA (TER BRAAK, 1988). As the gradient was relatively short (2.67), the importance of the measured environmental variables for the distribution of diatoms was investigated with a linear ordination technique for direct gradient analysis: redundancy analysis or RDA (JONGMAN *et al.*, 1987, TER BRAAK, 1994), using the computer programme Canoco, version 3.11 (TER BRAAK, 1988; 1990). In RDA the axes are constrained as linear combinations of the measured environmental variables. The scaling was for a correlation biplot, in which the correlation between the environmental variables and the axes can be read directly and both species and samples were standardized. All environmental variables except pH, water level and temperature, were log-transformed prior to the analysis in order to give approximately normal distributions. The 122 samples with full environmental data (Achterste Goorven A, B: Nov. 1979, May 1987 - Nov. 1994, Achterste Goorven E and Gerritsfles Nov. 1979 - Nov. 1994, Kliplo May 1982 - Nov. 1994) were used as active samples, the other 82 (mainly historical) samples as passive samples. The scores of these samples were calculated from the species composition after ordination of the active samples.

Only 26 taxa with a mean percentage abundance of at least 0.2% in the active samples were used for the ordination, as rare species often fall haphazardly in the ordination diagrams. Environmental variables were added by forward selection, after testing their significance ($P \leq 0.05$) with a Monte-Carlo permutation test. Another condition for selecting environmental variables was that they should not increase inflation factors (a measure for the mutual correlations between the environmental variables) to values above 5.

RESULTS AND DISCUSSION

Water level and chemistry

Due to natural fluctuations in precipitation and evaporation, particularly low water levels were observed in the summers of 1982 and 1986 in Gerritsfles, in 1982, 1986 and 1992 in Kliplo and in 1982, 1986, 1990 and 1991 in Achterste Goorven (Fig. 2). Particularly high water levels were found in the winters of 1988 and 1994 in the three pools.

The mean cation excess and its standard deviation are only 0.24 and 3.99% respectively, indicating that the data are reliable.

The medians and trends of selected variables are presented for Achterste Goorven E, Gerritsfles and Kliplo in Table 1. For Achterste Goorven A, B and E (AGA, AGB and AGE respectively) the medians for the period 1987-1994 are very similar. A marked gradient is present for the alkalinity, with the highest values (25 meq l^{-1}) at station A and the lowest values (–9 meq l^{-1}) at station E. The differences are most obvious for alkalinity, ammonium and sulphate in the period 1990-1992, after low water levels in the summers of 1989 and 1990 (Figs. 2 and 3) when, appa-

Table 1. Median values and trends in selected chemical and physical data. Abbreviations: corr. = Spearman rank correlation coefficient (x 100) with time, calculated from quarterly observations 1980-1994 in Achterste Goorven E and Gerritsfles (n = 60) and 1981-1994 in Kliplo (n = 55); Alkc = calculated alkalinity; DOC = dissolved inorganic carbon; TIC = total inorganic carbon. Parentheses indicate that trend is not very clear. C = constant, D = decrease, I = increase. *** = $p \leq 0.001$, ** = $p \leq 0.01$, * = $p \leq 0.05$.

		Achterste Goorven E			Gerrtitsfles			Kliplo		
variable	unit	median	corr.	trend	median	corr.	trend	median	corr.	trend
Al	mmol m^{-3}	21	-2	peaks 79, 91	5	-33**	peak 80	4	5	C
Alkc	mmol m^{-3}	-54	57***	I, dip 91-92	-37	45***	I	39	32*	C
Ca	mmol m^{-3}	64	-7	D, peak 90-92	30	14	D until 82	25	-1	C
Cl	mmol m^{-3}	394	-33**	D	240	-20	-	323	35**	peak 90-93
DOC	mmol m^{-3}	783	41**	I, dip 91-92	346	-41**	C	1082	51***	peak 91
K	mmol m^{-3}	43	-7	D, I since 92	26	-28*	D	41	72***	I
Level	m + MSL	8.35	26*	I, dip 89-91	39.92	0	C	13.01	3	C
Na	mmol m^{-3}	304	-15	D until 88, peak 90-91	194	-1	D, peak 90-91	261	35**	peak 90-93
NH_4	mmol m^{-3}	161	19	peaks 83-84, 90-91	75	-42***	peak 83-84	70	63***	I
NO_3	mmol m^{-3}	4	5	-	6	0	C	5	3	C
pH-field	-	4.3	43***	I	4.2	4	C	5.1	-19	C
pH-lab.	-	4.5	42***	I, dip 91-93	4.5	-12	I until 82	5.3	-33**	D
SO_4	mmol m^{-3}	245	-29	D, peak 90-92	104	-58***	D	73	41**	C
SO_4/Cl	-	1.19	-22	D, peak 90-92	0.92	-65***	D	0.44	29*	(I)
TIC	mmol m^{-3}	296	-59***	D, I since 92	137	-53***	D	183	-55***	D, peak 86

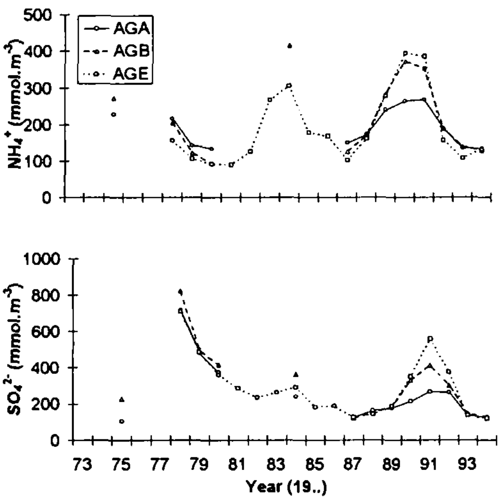

Fig 2: Changes in anual median values of NH_4^+ and SO_4^{2-} in surface water of Achterste Goorven stations A (AGA), B (AGB) and E (AGE).

rently, the largest proportion of the bottom did run dry at Achterste Goorven station E and the reduced sulphur compounds and organic nitrogen in the bottom material became mineralized and contributed to the acidification of the pool (VAN DAM, 1988).

For some variables plots of the time series are presented as median values per year in Fig. 3. The changes until 1984 have been presented and discussed earlier (VAN DAM, 1988). It was concluded that the severe drought of 1976 had a major impact on the water chemistry of Gerritsfles and particularly Achterste Goorven, where large fractions of the bottom were exposed to the atmosphere. The reduced sulphur compounds mineralized and high concentrations of sulphate, aluminium and calcium were observed, along with low values for pH and alkalinity. After the rather dry summer of 1986, low water levels occurred in the three pools Kliplo, Gerritsfles and Achterste Goorven, but the water level increased quickly in the extremely wet year of 1987. However, the drought in the summers of 1989 and 1990 was more severe, particularly in the area of Achterste Goorven. As a result the post-drought processes of 1976 repeated, including precipitation of dissolved organic carbon by complexation with dissolved aluminium (Fig. 3).

In Achterste Goorven and Gerritsfles the median sulphate concentrations declined in 1994 to 31% and 34%, respectively, of their 1980 values. In Kliplo, the median sulphate concentration in 1994 was 16% higher than in 1981. Besides the changes in sulphate concentrations, the changes in the quotient of sulphate and chloride are also presented in Fig. 3, to compensate for changes induced by evaporation. In Gerritsfles the trend of SO_4^{2-}/Cl^- is nearly linear and significant (Table 1). Although

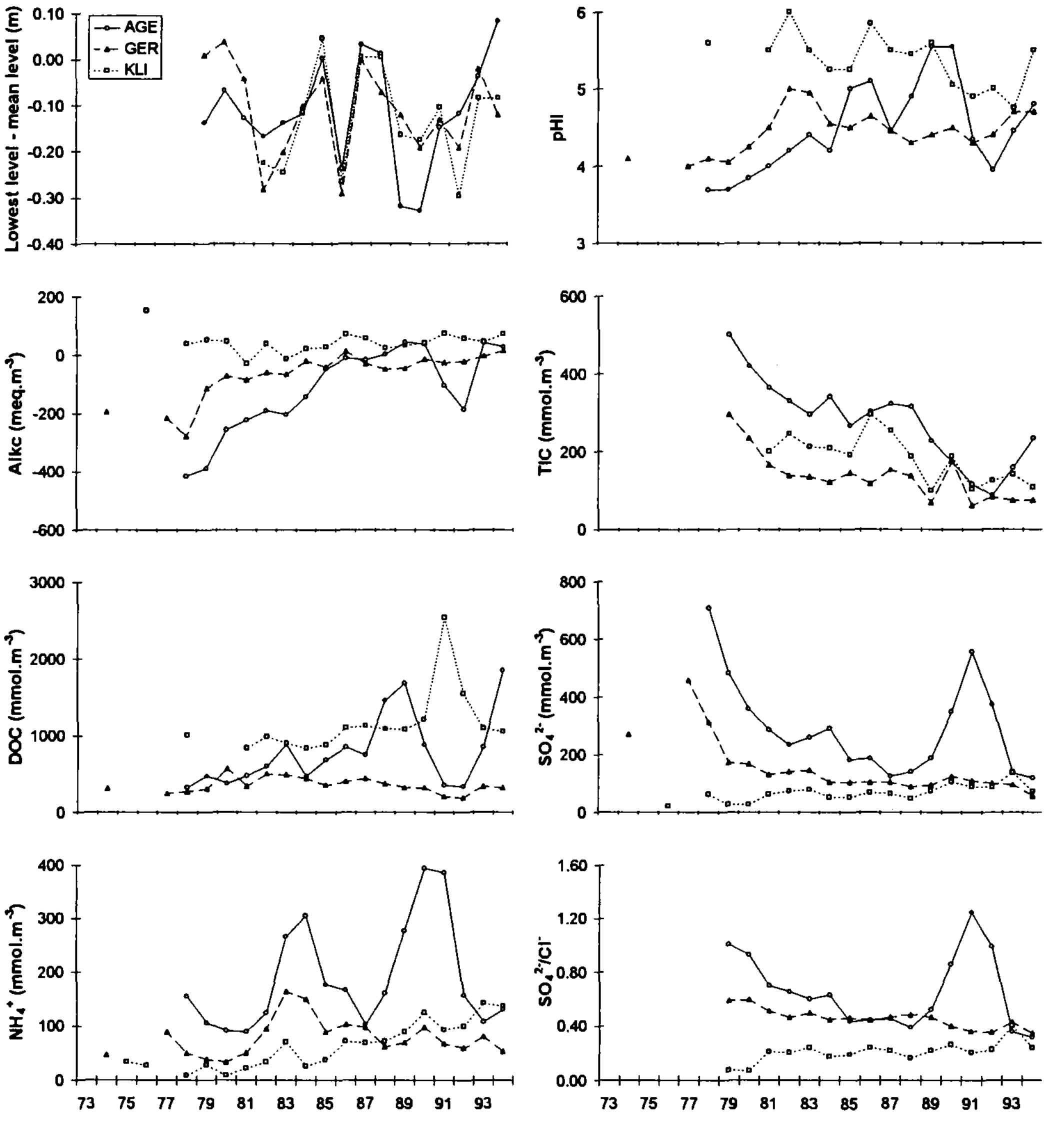

Fig 3: Changes in annual median values of selected physical and chemical variables in surface water of Achterste Goorven station E (AGE), Gerritsfles (GER) and Kliplo (KLI). pHl = pH-laboratory, Alkc = calculated alkalinity, TIC = total inorganic carbon, DOC = dissolved organic carbon.

this decline was much slower than the decline of SO_X in atmospheric deposition, the ecological model AquAcid showed that this has been due to the decline of atmospheric deposition of SO_X (VAN DAM *et al.*, 1996). Unfortunately, the model could not be calibrated for Achterste Goorven. Neither was it possible to calculate which fraction of the decline of sulphate concentration was due to the decline in atmospheric deposition of SO_X. In Kliplo, sulphate concentrations were constantly low, with perhaps a slight tendency to increase due to intensive bacterial reduction of sulphate (MARNETTE *et al.*, 1993; VAN DAM *et al.*, 1996). Large peaks of sulphate did not occur, because even at extremely low water levels the largest part of the bottom is still submerged (VAN DAM, 1988).

Ammonium at Achterste Goorven station E had peaks in 1981-1984 and 1989-1991. The latter peak coincided with the drought peak of sulphate and may have been caused by mineralization of nitrogen compounds from the sediment. However, the origin of the first peak is not clear. In Gerritsfles there was a small peak of ammonium in 1981-1984. Fluctuations of ammonium may also be caused by changes in the abundance of the macrophyte *Juncus bulbosus*, which assimilates ammonium (SCHUURKES *et al.*, 1986). In Kliplo there was a significant increase of ammonium from concentrations around 20 mmol m^{-3} in 1981 to 142 mmol m^{-3} in 1993-1994.

In the three pools concerned there has been an obvious decrease of total inorganic carbon (TIC), which is nearly the same as carbon dioxide in these acid waters (Fig. 3). The long-term decrease of carbon dioxide in moorland pools is due to the decrease of the carbon stock in the sediments by acidification. Diffusion of carbon dioxide from the air is too slow for the maintenance of concentrations which are sufficiently high for the growth of most aquatic macrophytes (ROELOFS, 1983). The increased mineralization during and after drought periods may contribute to increased levels of carbon dioxide in the water layer by diffusion from the sediment layer, which is apparently the cause of the increase of *Juncus bulbosus* in Achterste Goorven after the drought periods of 1976, 1986 and 1989-1991.

Several variables, including sulphate, chloride, pH, alkalinity, ammonium and nitrate, showed significant seasonal variation, which are discussed in VAN DAM *et al.* (1996).

Diatom taxa, ecological groups and inferred pH

In the 204 investigated samples 148 taxa were found. The majority of the taxa occurred only rarely. The 34 taxa summarized in Table 2 represent 98% of the 81 600 individuals counted. The most common taxa are a typical combination for (very) acid surface waters, as may be expected. The average number of taxa in the samples (16) was low, due to the (very) acid environment, to which few taxa are adapted.

Due to the relative increase of *Eunotia exigua* over time, the percentage abundance of ecological groups has changed drastically since the beginning of the century; the greatest decrease occurred within the soft-water taxa. The changes have been relatively minor in Kliplo. There is an obvious decline of the diatom-inferred pH in all pools, except Kliplo between 1920 and 1980 (Fig. 4).

Table 2. Average percentage abundance (P.A.) of diatom taxa in all the 204 investigated samples. Only taxa with an average percent abundance of at least 0.05% in these samples are listed. Taxa are ordered according to their average percent abundance in ecological groups. Asterisks indicate taxa included in redundancy analysis.

Ecological group / taxon	P.A.
* ***Eunotia exigua***	37.4
Acid-water taxa	
* *Frustulia rhomboides* var. *saxonica* s.l.	11.2
* *Eunotia rhomboidea*	9.3
* *Tabellaria quadriseptata*	7.4
* *Eunotia bilunaris*	5.1
* *Pinnularia gibba*	0.2
* *Eunotia paludosa*	0.2
Pinnularia microstauron	0.2
Eunotia implicata	0.1
Frustulia rhomboides	0.1
Soft-water taxa	
* *Eunotia incisa*	6.1
* *Navicula micropunctata* - group	4.0
* *Navicula leptostriata* s.l.	3.0
* *Tabellaria flocculosa*	2.7
* *Anomoeoneis vitrea* fo. *lanceolata*	2.5
* *Fragilaria exigua*	2.1
* *Eunotia naegelii*	0.9
* *Nitzschia perminuta*	0.8
* *Anomoeoneis brachysira*	0.7
* *Cymbella cesatii*	0.7
* *Pinnularia interrupta*	0.7
* *Navicula subtilissima*	0.7
* *Navicula mediocris*	0.3
* *Cymbella gracilis*	0.3
Eunotia intermedia	0.2
* *Eunotia arculus*	0.2
* *Anomoeoneis serians*	0.2
* *Anomoeoneis vitrea*	0.2
Peronia fibula	0.1
Eunotia nymanniana	0.1
Tabellaria binalis var. *elliptica*	0.1
Eunotia exigua var. *tridentula*	0.1
* ***Achnanthes minutissima***	0.7
Eutraphentic taxa	
* *Nitzschia gracilis*	0.5

Since 1978 the changes in the percentage abundance of *Eunotia exigua* in the pools Gerritsfles and Achterste Goorven E are most conspicuous. The decrease of sulphate concentrations has been more ore less parallelled by the decline of this acid-resistant diatom, although there was a lag of several years (Figs. 3 and 4). The decrease of the percentage abundance of this taxon allowed acid-water taxa and soft water taxa to increase. The diatom inferred pH had increased over time in these two pools.

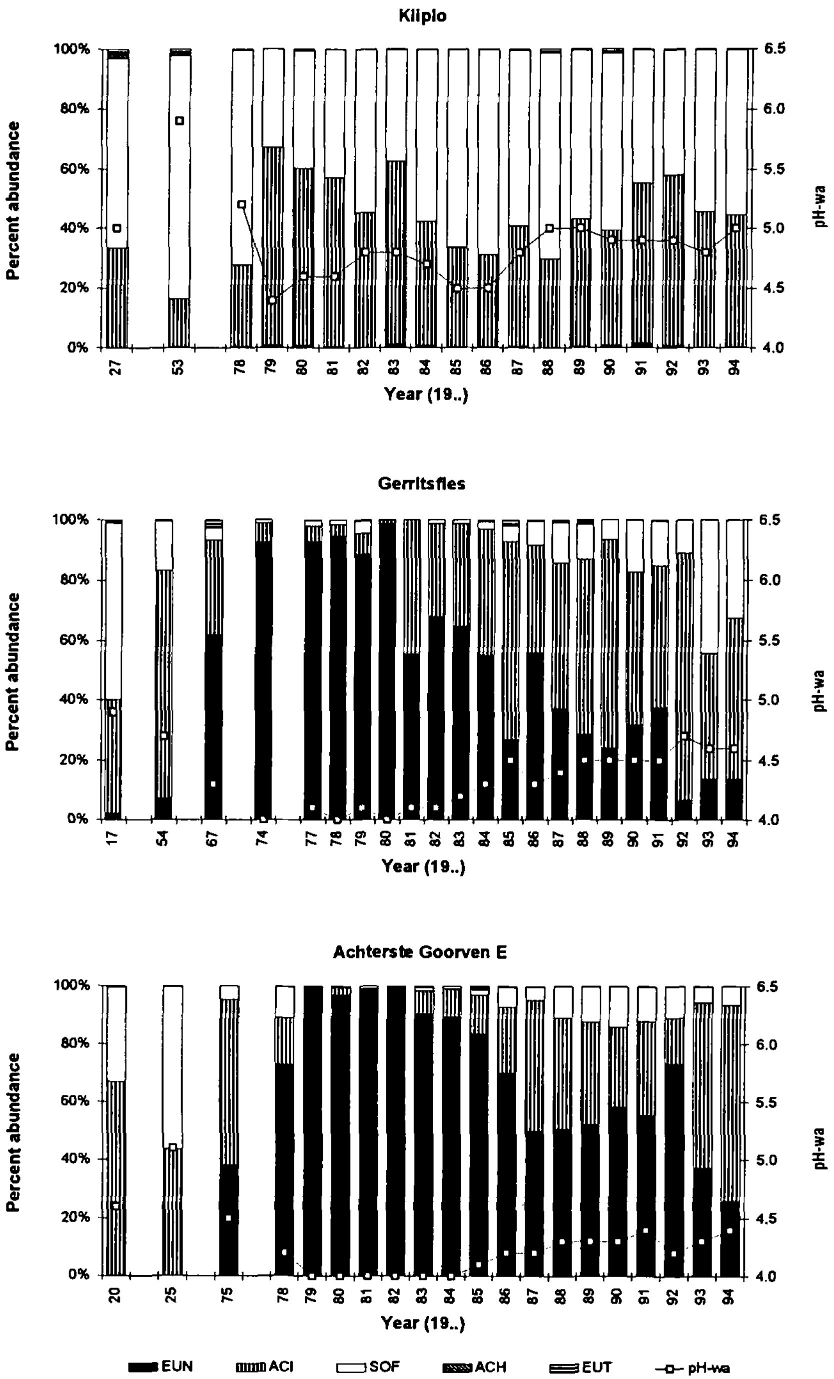

Fig. 4: Long-term changes in percentage composition of ecological groups of diatoms and diatom-inferred pH (pH-wa) in Kliplo, Gerritsfles and Achterste Goorven E. EUT = eutraphentic taxa, ACH = *Achnanthes minutissima*, SOF = soft-water taxa, ACI = trivial taxa from acid waters, EUN = *Eunotia exigua*. Early samples were pooled: E: 1919 - 1920 ⟶ 1920 (n = 4), 1921 - 1928 ⟶ 1925 (n = 6); G: 1916 - 1918 1917 (n = 3), 1950 - 1960 ⟶ 1955 (n = 4), 1964 - 1973 ⟶ 1969 (n = 4) ; K: 1924 - 1929 ⟶ 1927 (n = 2), 1948 - 1958 ⟶ 1953 (n = 2), 1962 - 1972 ⟶ 1967 (n = 3).

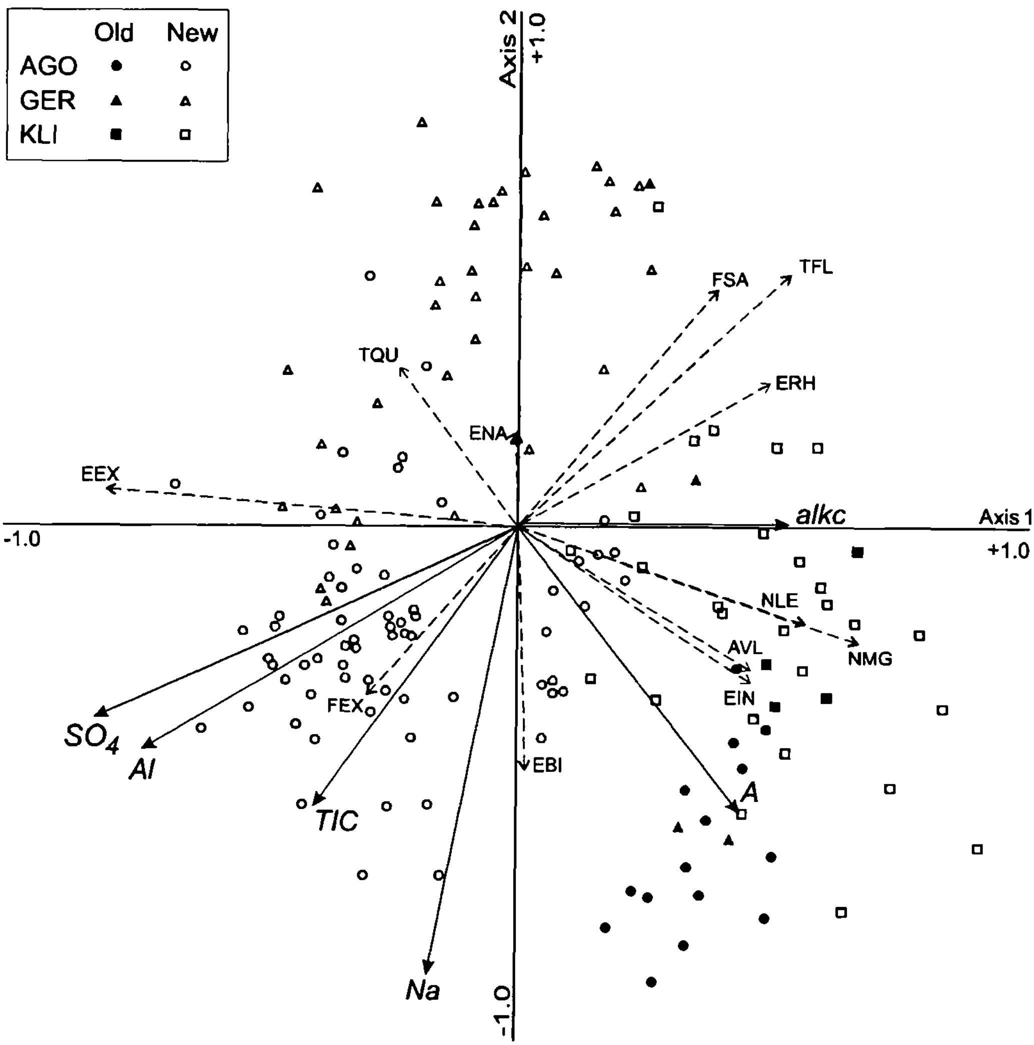

Fig 5: Correlation biplot based on a redundancy analysis displaying 38% of the variance in the log-percentage abundances and 86% of the variance in the fitted abundances. Significant ($p < 0.05$) quantitative environmental variables are indicated by solid arrows, species with a mean relative abundance >1% in the active samples are indicated by dotted arrows (AVL = *Anomoeoneis vitrea* fo. *lanceolata*, EBI = *Eunotia bilunaris*, EEX = *E. exigua*, EIN = *E. incisa*, ENA = *E. naegelii*, ERA = *E. rhomboidea*, FEX = *Fragilaria exigua*, FSA = *Frustulia rhomboides* var. *saxonica*, NLE = *Navicula leptostriata*, NMG = *N. micropunctata*-group, TFL = *Tabellaria flocculosa*, TQU = *T. quadriseptata*). Eigenvalues of the first three axes are 0.30, 0.08 and 0.05; the sum of all canonical eigenvalues is 0.44. The scale marks along the axes apply to the species and quantitative environmental variables; the sample scores were multiplied by 0.33 to fit in the coordinate system. Samples are indicated by symbols as explained in the upper left, where AGO = Achterste Goorven A, B and E, GER = Gerritsfles, KLI = Kliplo, Old = 1916 - 1958 and New = 1960 - 1994. Sample scores are linear combinations of environmental variables for 122 active samples and mean species scores for 82 passive samples (see text).

The changes in Achterste Goorven and Gerritsfles have been caused by the recovery from the severe drought of 1976 and the decrease of atmospheric deposition. In Achterste Goorven there was a second increase of *Eunotia exigua* from 1989 to 1992 following the dry summers of 1989 and 1990. The apparently delayed reaction to this drought and concomitant increase of sulphate might be due to the method of sampling. The plankton net may also remove the diatom valves in the uppermost sediment layer, which was deposited some years before the moment of sampling. In Gerritsfles a second peak of *E. exigua* was absent, as was the second peak of sulphate. Gerritsfles recovered faster than Achterste Goorven E. In the most recent years, the percentage distribution of ecological groups was similar to that in the samples taken at the beginning of this century, although the proportion of *E. exigua* is higher in the recent samples than in the old samples. Moreover, the diatom-inferred pH is lower for the recent than the old samples. At the stations in Achterste Goorven A and B, the main trends were similar to those of station E in this pool, but conditions were less acid than at station E and the second peak of *E. exigua* after 1990 was not present here (VAN DAM *et al.*, 1996).

In Kliplo no clear changes can be found since 1978, although there may be an upward trend of the diatom-inferred pH since 1979 (Fig. 4).

Relationships between diatoms and water chemistry

In the first run of the redundancy analysis (RDA), with 29 environmental variables, including time, the third significant ($p \leq 0.01$) environmental variable was time, after sulphate and sodium. This analysis illustrates that there is a significant trend in our data. However, since time at the scale of months and years is not a relevant variable in the environment and in life cycles of diatoms, with doubling rates of about one day, we excluded time from the following analysis.

The results of the ordination with 28 initial environmental variables are summarized in Fig. 5. As the first two eigenvalues are relatively high, the ordination diagram summarizes the information rather well.

The most important environmental variable is sulphate, which explains 21% of the variation in species composition and has a correlation coefficient (r) of 0.82 with the first axis, as can be read from the plot by projection of the end of the arrow on this axis. The second important variable is sodium (≈ chloride), which has a high correlation ($r = 0.87$) with the second axis. Sulphate and sodium together explain 33% of the total variation. The third major environmental variable is total inorganic carbon (≈ carbon dioxide), which has correlation coefficients with the two first axes of -0.40 and -0.50, respectively, and which explains, together with the two previous variables, 38% of the total variation. Although organic anions (≈ dissolved organic carbon), alkalinity and aluminium are significant environmental variables, they explain only a minor proportion of the variance. The six selected variables explain 44% of the total variation.

Changes in concentrations of sulphate, inorganic carbon species, organic ions, and aluminium and of alkalinity are well known in acidified lakes and pools (LEUVEN *et al.*, 1986; REUSS and JOHNSON, 1986). Thus acidification exerts a major influence on the composition of the diatom assemblages in the pools, although also other chemical variables are important. This is indicated by the significance of sodium. Probably sodium itself is not a decisive variable for the diatoms. Each pool has a unique species composition, which is determined by a large number of (partially unknown) environmental variables. As each of the investigated pools has its own discrete range of sodium concentrations (medians 194, 261 and 304 mmol m^{-3}, Table 1) sodium might be selected by the ordination program to reflect this unique character of each of the pools.

Note that seasonal variation is not among the selected environmental variables. It was used as a passive environmental variable in a subsequent analysis and it is not correlated with any of the ordination axes. Moreover, ammonium and nitrate are not included in the selected variables. Nitrate appears to be especially insignificant for the composition of the diatom assemblages. Ammonium has a somewhat stronger influence, as appeared during the selection procedure. It is correlated ($r = 0.58$) significantly with sulphate. As a passive environmental variable it could be entered in the biplot of Fig. 5 as a vector with the coordinates -0.41 and -0.32, more or less in between aluminium and dissolved inorganic carbon.

The samples from the different pools and periods are arranged in well defined clusters. Nearly all the old (1916-1958) samples are in the lower right section of the diagram. At first sight this could mean that the diatom assemblages of the different pools were very similar at an earlier stage. However, there were historically large differences between the pools, *e.g. Cymbella cesatii* was very abundant in Achterste Goorven A, *Tabellaria flocculosa* in Kliplo and *Eunotia incisa* in Gerritsfles. Most of these characteristic taxa are much rarer in the (active) samples from the period 1979-1994, which were used for the calculation of species scores, from which the position of the (passive) samples in the ordination plot was calculated. There is no reliable modern analogue for the historical samples from Achterste Goorven and Gerritsfles. Nevertheless, the diagram illustrates the discrepancy between the old and recent samples from these pools. Some of the old samples from Gerritsfles are in the upper right part of the diagram: these are relatively recent samples (1950-1958). The recent samples from Kliplo are close to the old ones, while the recent samples from Achterste Goorven are mainly in the lower left part of the diagram.

The diagram shows that most of the old samples, like the present samples from Kliplo, are from water that is rich in organic anions (A), while the recent samples from Achterste Goorven and some recent samples from Gerritsfles are from water that is rich in sulphate and aluminium. The majority of the recent samples from Gerritsfles is opposite to the direction in which the arrow for sodium is pointing. This means that these samples are from water which is relatively poor in sodium. The calculated alkalinity is relatively high for the samples in the right part of the diagram (Kliplo and old samples from other pools).

The most important species determining the first axis is *Eunotia exigua*, which has a score on (and also a correlation with) this axis of -0.80. This can be read from the plot by projecting the head of the arrow on axis 1. This species is well known from acidified surface waters which are rich in sulphate and aluminium (KRAMMER and LANGE-BERTALOT, 1986-1991). *Navicula leptostriata* and the *N. micropunctata*-group have a reverse preference: they prefer humic acid waters and do not tolerate high concentrations of mineral acid. Therefore they are most abundant in the samples from Kliplo and old samples from Achterste Goorven. *E. bilunaris* has approximately a zero score on the first axis and shows an intermediate behaviour with regard to the origin of the acidity (mineral *vs.* organic). Although *Tabellaria quadriseptata* is known from humic acid waters (KRAMMER and LANGE-BERTALOT, 1986-1991), it preferentially occurs in our samples from Gerritsfles, which are poor in organic anions: it is situated in the diagram just opposite of A. The vector for *T. flocculosa* is approximately opposite to the vectors for sulphate, aluminium and total inorganic carbon. This species apparently shuns acidified pools. It is most abundant in samples from the humic-acid pool Kliplo and some recent samples from the clear water pool Gerritsfles. The vector for *Frustulia rhomboides* var. *saxonica* is perpendicular to that of organic anions, which would mean that this taxon is indifferent to the presence of humic matter. According to KRAMMER and LANGE-BERTALOT (1986-1991), it should have a preference for water rich in humic compounds.

Partial recovery of diatom assemblages from acidification

In order to inspect the temporal variation within each of the pools in more detail, separate ordination diagrams are presented in Fig. 6. The same coordinate system as for Fig. 5 is used. As the position of many of the samples coincides, the temporal developments are clarified by adding the schematized time-tracks. It thus becomes clear that in Achterste Goorven A the samples from 1919 are situated in the central part of the bottom of the diagram, with a move to the right until the fifties. After 1953 there is a major shift to the bottom left during the late seventies and early eighties. In the nineties the samples are much more similar to those of 1919. However, it should be noted again that the samples from the nineties do differ from those in 1919, as species such as *Achnanthes minutissima*, *Cymbella cesatii* and *Anomoeoneis vitrea* fo. *lanceolata* were very abundant in the old samples and rare in the recent samples. These developments are a reaction to the chemical changes in the pool, from presumably being a humic-acid pool in 1919, to an acidified pool, rich in sulphate, around 1980 and a decline of sulphate and increase of humic acids since 1980.

The same general pattern is found in Achterste Goorven B. The samples from 1925-1929 are located closer to the bottom than in Achterste Goorven A. A sample from 1975 is also available from this station. Its location in the diagram suggests a clear water pool which is not very rich in sulphate, in agreement with the direct observations made by KWAKKESTEIN (1977). After the extreme drought of 1976 there was a shift to the lower left, with a dominance of *Eunotia exigua*, as a consequence of the mineralization of sulphur compounds from the sediments. During the post-drought phase there was an intitial recovery in the approximate direction of the situation in 1975, but the direction reversed after the increase of sulphate concentration resulting from the minor drought of 1989-1991. After 1991 there was a similar development as in Achterste Goorven A.

The pattern in Achterste Goorven E is a variant of the pattern in Achterste Goorven B. The samples from the period 1919-1926 are situated in a cluster on the lower right hand side. There is no distinct change in the composition of the diatom

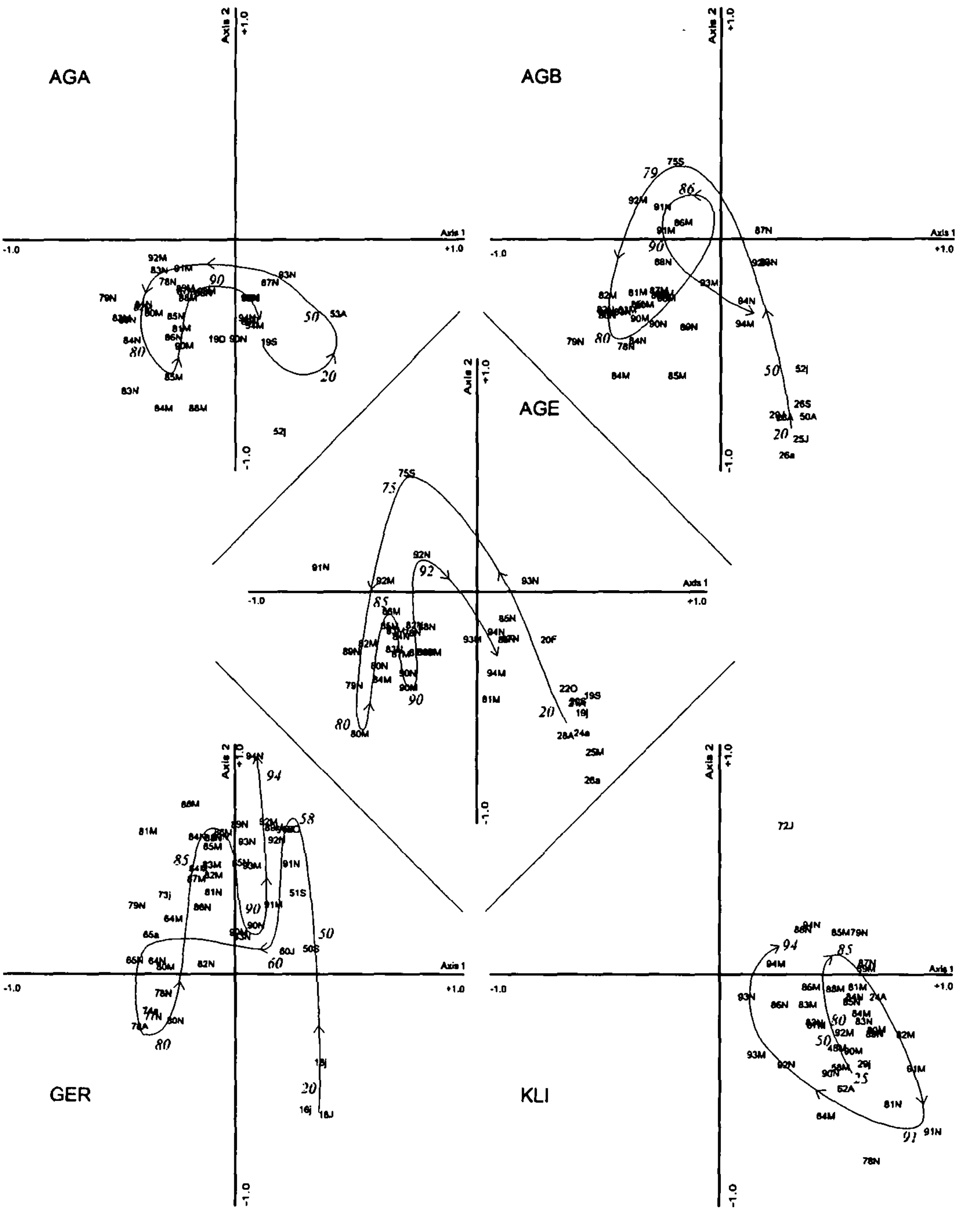

Fig. 6: Long-term changes of diatom assemblages in the same co-ordinate system as Fig. 5. For each pool (AGA, AGB, AGE = Achterste Goorven A, B and E. respectively, GER = Gerritsfles, KLI = Kliplo) the position of the samples is indicated by their labels, where the two digits denote the year in the present century and the letter the month (F = February, a = April, M = May, j = June, J = July, A = August, S = September, O = October, N = November, D = December). The curves represent the schematized time-tracks: the direction is indicated by arrowheads and the approximate time by the large-size year-numbers.

assemblages after the severe drought of 1921 (SCHUURMANS, 1977), when a large part of the sediment of Achterste Goorven was exposed to the atmosphere (VAN DAM, 1988). In 1975, and later years, the developments were similar to those in Achterste Goorven B. After the minor drought of 1989-1991 there was a temporary shift to lower scores on the second ordination axis through the increase of *E. exigua.*

In Gerritsfles there is a large increase of the score on the second ordination axis from 1916-1918 until 1958. This should correspond with a decrease of sulphate concentrations (the sulphate concentrations measured in 1925 and 1930 were not particularly low, VAN DAM, 1988). After the extremely dry year 1959 (SCHUURMANS, 1977), it is likely that a large part of the bottom was exposed to the atmosphere and reduced sulphur compounds were oxidized to sulphate. This is reflected by the shift of the sample station in the diagram in the lower left direction in the period 1960-1965. From 1964 until the mid eighties the Gerritsfles pool was dominated by *E. exigua.* With the steady decline of sulphate in this pool this taxon also declined and other taxa (*Tabellaria quadriseptata, T. flocculosa* and *Frustulia rhomboides* var. *saxonica*) became abundant. A remarkable development is the increase of *E. naegelii* since 1992, which was not found in earlier samples from Gerritsfles. In contrast to Achterste Goorven, there is no return to the situation of the beginning of this century, in spite of the partial recovery from acidification.

The similarity between old and recent samples from Kliplo in the ordination diagram is a real similarity, as there have been no significant changes in the species composition. After 1985 there is a spiral move of the station through the ordination. The high score on the first axis and the low score on the second axis in 1991 might be caused by the peak of dissolved organic carbon in that year (Fig. 3). The subsequent shift to the left might be a consequence of the increase of sulphate, particularly in 1993 (Fig. 3). The sample from 1972 has an isolated position in the upper right part of the diagram. Most species in this sample were similar to those of other samples, but *Frustulia rhomboides* var. *saxonica* was very abundant, while *Navicula micropunctata*-group was absent. The sample was one of over a hundred samples which were taken by SMIT (1976) in similar moorland pools, and was probably confused with another sample. There are clear signs of long-term eutrophication of Kliplo due to atmospheric deposition, *e.g.* increase of ammonium, increase of frequency of blooms of the green alga *Coenochloris helvetica.* This is not reflected by the species composition of diatoms in this pool, although diatoms are generally known to be reliable indicators for trophic state in moorland pools (VAN DAM and KOOYMAN-VAN BLOKLAND, 1978; VAN DAM and BUSKENS, 1993).

The recovery of diatom assemblages in lakes by decreasing acid atmospheric deposition has been clearly shown with paleolimnological techniques in the Sudbury area (Canada), after a dramatic deline of the local emission by melters (DIXIT *et al.*, 1992). Recovery of the diatom assemblages by reductions of sulphur emissions on a national scale was demonstrated with palaeolimnological techniques in some Scottish and Welsh lakes (ALLOTT *et al.*, 1992; JUGGINS *et al.*, 1996), but not by real-time monitoring, because the available series of five years was too short (LANCASTER *et al.*, 1996). Thus our series is probably the first one where partial recovery of acidification is shown by real-time monitoring of diatom assemblages.

Future developments

Probably, further changes due to recovery from acidification are to be expected in moorland pools. The emissions of SO_x in The Netherlands declined with 70% between 1980 and 1994, and further reductions down to 15% of the value in 1980 are to be achieved in 2010 (NIJPELS, 1989; RIVM, 1993, 1995). Also large emission reductions are to be realized in neighbouring countries (UNITED NATIONS, 1994). The average (potential) acid atmospheric deposition over The Netherlands should decline from 7 kmol H^+ ha^{-1} y^{-1} in 1980 via 4.0 kmol H^+ ha^{-1} y^{-1} in 1994 to 1.4 kmol H^+ ha^{-1} y^{-1} in 2010, where the long-term target value (critical load) for moorland pools is about 0.5 kmol H^+ ha^{-1} y^{-1} (NIJPELS, 1989; VAN DAM & BUSKENS, 1993). Probably the expected changes in deposition will be followed by changes in chemistry and diatom assemblages of moorland pools.

The emissions of NO_y declined with 10% between 1980 (when it was 538 10^6 kg y^{-1}) and 1994, while there are large uncertainties about future emissions. Probably, there will be no long-term decrease at all. The emissions of NH_x declined with 27% between 1980 (250 10^6 kg y^{-1}) and 1994 and further reductions down to 30% of the value in 1980 are to be achieved in 2010. The major decrease should be realized between 1990 and 2000 (NIJPELS, 1989; RIVM, 1993, 1995). These changes in emissions are difficult to convert into depositional changes, as the rate of nitrogen deposition above moorland pools is very uncertain (VAN DAM *et al.*, 1996). The current nitrogen deposition above the three monitored pools is 2-10 times higher than the critical deposition value of 0.3-0.6 kmol ha^{-1} y^{-1} (NIJPELS, 1989; BOBBINK and ROELOFS, 1995). With the ecosystem model AquAcid inorganic nitrogen concentrations in moorland pools can be predicted, but the model has still to be extended and improved (VAN DAM *et al.*, 1996).

CONCLUSIONS

The present study has shown that the efforts for the reduction of SO_x-deposition, particularly between 1985 and 1990, in The Netherlands have had positive effects on the condition of moorland pools: sulphate concentration in the surface water has decreased, acid-tolerant diatoms decreased and acid-sensitive diatoms increased in two of the three pools sampled. The deleterious effects of acidification by atmospheric deposition are particularly obvious after extremely dry years, due to oxidation of reduced sulphur compounds from the sediments. Similar effects were observed in other moorland pools, which were monitored once in every four years or less frequently (AQUASENSE TEC, 1995a,b).

It was demonstrated that diatoms are very suitable biological indicators for monitoring the recovery from acidification by sulphur compounds (see also BATTARBEE *et al.*, 1988 and ALLOTT *et al.*, 1992). Sulphate was the most important environmental variable for their distribution. The diatoms respond faster to improved conditions than aquatic macrophytes, which have a longer life cycle than the unicellular algae and are more dependent on the sediment. Hitherto, no spontaneous improvements of macrophyte vegetation in acidified moorland pools in The Netherlands have been observed, although soft-water macrophytes have returned in pools from which sediments were removed and buffering substances were added (BELLEMAKERS *et al.*, 1993).

These results could be obtained because a high-quality data set, obtained by systematical monitoring over a period of more than fifteen years, supplemented by a large amount of data from the beginning of this century, was available. This is a longer record than in most other countries in Europe and North-America, where the impact of acid atmospheric deposition on surface waters has been monitored, in the network of the International Cooperative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes (SKJELKVÅLE *et al.*, 1994).

Further monitoring of the effects of acidification on moorland pools is necessary, as these waters have proved to be excellent ecosystems to monitor the effects of changes in atmospheric deposition and can be used to test the effectivity of measures for the abatement of air pollution, in connection with climate-induced changes (VAN DAM and BELTMAN, 1992).

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