

# Impact of acidification on diatoms and chemistry of Dutch moorland pools

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## Abstract

Old (ca 1920) and recent (1978) diatom assemblages from sixteen pristine moorland pools are compared by analysis of pH-spectra, diversity (Hill's index), (dis)similarity (number of species in common, Dyer dissimilarity) and principal component analysis. The pH-spectra of clear water pools indicate that the formerly wide range of pH (4–6) is very narrow now (3.7–4.6). No significant change of pH (ca 4.5) is observed in brown water moorland pools. Diversity significantly declines in clear water pools and has a tendency to rise in brown water pools. The number of species in common does not change. The Dyer dissimilarity significantly decreases in clear water lakes, no change is found in brown water lakes. The first principal component (PC 1) explains 61% of total variance between samples and is nearly completely determined by *Eunotia exigua*. PC 1 has a strong positive correlation with both the absolute and relative sulfate concentration and other factors related to acidification ( $\text{Ca}^{2+}$ ,  $\text{Al}^{3+}$ ,  $\text{Mg}^{2+}$ , electrical conductivity). PC1 has a negative correlation with factors characterizing humic acid waters ( $\text{KMnO}_4$ -consumption, Fe,  $(\text{Na}^+ + \text{K}^+)/(\text{Ca}^{2+} + \text{Mg}^{2+})$  etc.). Old samples have low scores on PC 1. Recent samples from clear waters have high scores on PC 1. The original variation, caused by regional factors, is replaced by a sulfate controlled variation. The factors which are responsible for the recent differences in sulfate concentrations between pools are discussed (sulfate reduction, accumulation by dry deposition in adjacent pine forests and drought). Some observations contribute to the opinion that acidification may be considered as eutrophication and not as oligotrophication.

## 1. Introduction

The growing use of fossil fuels has caused an increase of atmospheric sulfur in the last decennia. The deleterious effects of acid rain upon aquatic ecosystems have been studied in Scandinavia and North America (Braekke 1976; Almer *et al.* 1978). The extinction of fish populations is very obvious, but also other components of the ecosystem, *viz.* the species composition of plankton and periphyton changes thoroughly by acidification.

The Netherlands are in the very centre of the acid rain area of north-western Europe (Granat 1978). World's lowest annual mean pH yet recorded is 3.8 in 1967 at De Bilt (Fig. 1). The present pH of

precipitation is about 4.3 (Anonymous 1979).

In spite of this severe pollution no investigations focussed on this problem have been carried out in the Netherlands up to now, although Coesel *et al.* (1978) and Van Dam & Kooyman-van Blokland (1978) present strong evidence for the adverse effects on desmids and diatoms in moorland pools (Dutch: ven; German: Heidetümpel). These waters are in a geologic environment that is highly resistant to chemical weathering and have relatively low concentrations of major ions and hence a low buffer capacity. Such waters are extremely vulnerable to inputs of acid precipitation (Wright & Gjessing 1976).



Fig. 1. Location of the study areas. x = rainwater gauge (Anonymous 1979).

Diatoms are widely used as pH-indicators in actuo- and palaeolimnology since the conception of the pH-classification scheme of freshwater diatoms by Hustedt (1939). Miller (1973) and Berge (1979) used the diatom assemblages in cores of Scandinavian lake sediments as a record of past pH-conditions to assess the impact of acid precipitation. Berge (1976) compared diatom samples from 1949 and 1975 to establish a decrease of pH in some south Norwegian streams and ascribed this shift to the acidification of precipitation.

Many moorland pools are shallow and poor in sediments. Therefore we used old samples to estimate the past environmental conditions. A large number of algal samples in moorland pools have been collected between 1916 and 1955 for studies on desmids (see *e.g.* Heimans 1969). In 1978 new samples were taken at the same sites.

According to the theories of Thienemann (1920), Franz (1953), Van Leeuwen (1966), Margalef (1975) and May (1975) complex and delicately balanced ecosystems are confined to a predictable environment (stable conditions) with a low input of energy and matter. Complex natural ecosystems (with a

high diversity) are fragile, *i.e.* very sensitive to perturbation. Perturbation means a relatively unpredictable environment, with a high input of energy and matter. These unstable conditions only permit structurally simple, robust ecosystems (with a low diversity) to exist.

The pH of unpolluted rainwater, in equilibrium with CO<sub>2</sub> in the atmosphere is 5.6–5.7. The present pH of many moorland pools is in between 3.5 and 4.0. This is a difference of about 200  $\mu\text{eq l}^{-1} \text{ H}^{+}$ . With this increase in hydrogen ion concentration all original pH-values of the moorland pools between about 4 and 6 will be concentrated in the narrow range between 3.5 and 4.0. So, similarity of the environment in the moorland pools will increase by acidification.

It may be expected therefore that acidification causes a decrease of diversity and dissimilarity of the diatom assemblages in moorland pools. We will test these hypotheses. Besides, the acidification is shown by comparison of pH-spectra of old and recent diatom samples. Multivariate analysis of data gives insight to changes in species composition, diversity, dissimilarity and their relationships with water chemistry.

Also other processes than pollution of rain water may be responsible for the acidification of moorland pools, *e.g.* the severe drought of 1976 and the reafforestation of the catchments. Their significance will be discussed.

## 2. Study sites

Moorland pools are shallow (depth less than ca 3 m) soft water lakes with no visible in- and efflux. They are part of the heath (podzol) landscape with poorly buffered sandy soils. The surface area of the sixteen pools we studied (Fig. 1; Table 1) varies from about 0.1 to 15 ha.

Eight pools are situated in the province of North-Brabant, near Oisterwijk (B), two in the Veluwe area (V), and six in the province of Drenthe (D).

The bottom of most pools in the B- and V-districts is sandy and often covered with a thin layer of detritus and peat. No detailed knowledge about the hydrology of most pools is available. V1 (Fig. 2) and V2 are situated some tens of meters above the aquifer. Their watershed is hardly larger than their surface and sealed by an impermeable iron-pan

Table 1. Physico-geographical data.

DISTRICT/ name of pool	Number	Surface area (ha) approx.	Bottom <sup>a</sup>	Basin	Brown or clear water <sup>b</sup>	Surrounding landscape ca. 1920 <sup>c</sup>	Surrounding landscape 1978 <sup>c</sup>
<b>BRABANT</b>							
Galgeven	B1	14,7	sand	probably unsealed	clear	heath with regrowth of forest	forest
Schaapsven	B2	1,4	sand	unknown	clear	heath with regrowth of forest	forest, heath with regrowth of forest
Diaconieven	B3	1,3	sand	rather sealed	clear	forest planted before 1900	forest
Brandven	B4	1,4	sand	rather sealed	clear	forest planted before 1900	forest
Achterste Goorven	B5	1,4	sand	probably unsealed	clear	forest planted before 1900	forest
Klein Aderven	B6	1,0	sand	slightly sealed	clear	heath, forest planted before 1900	forest
Middelste Wofspuiven	B7	0,3	sand	unknown	brown	forest planted before 1900	forest
Groot Huisven	B8	2,9	sand	unsealed	clear	heath	heath, forest
<b>VELUWE</b>							
Gerritsfles	V1	5,0	sand	sealed	clear	heath, sand dunes	heath, locally with regrowth of forest;
Kempesfles	V2	0,4	sand	sealed	clear	sand dunes with regrowth of forest	sand dunes sand dunes with regrowth of forest
<b>DRENTHE</b>							
Kliplo	D1	0,4	sand	possibly unsealed	brown	forest planted after ca 1920, heath	forest
Poort II	D2	0,1	peat	unknown	brown	bog surrounded by forest planted after ca. 1920	bog surrounded by forest
Schurenberg	D3	1,1	sand	possibly unsealed	clear	forest	forest
Diepveen	D4	0,9	peat	unknown	brown	forest planted after ca. 1915	forest
Reeënveen	D5	0,1	peat	unknown	brown	bog surrounded by heath	bog surrounded by forest
Pool in Echtenerzand	D6	0,3	peat	unknown	clear	sand dunes, heath	sand dunes, heath

<sup>a</sup> Sand bottoms are often covered with detritus.

<sup>b</sup> Based on chemical data from table 5 (section 4.6). These do not always correspond with the actual water colour which is seen in the field. The water of D1 looks clear and that of D6 looks brown.

<sup>c</sup> Forests consist mainly of Scots pine, sometimes mixed with some birch and/or oak.

(Schimmel & Ter Hoeve 1952). Also B3 and B4 have sealed basins (Ter Hoeve 1949). B5 is aquifer fed and consequently has a larger basin than the previous moorland pools. Transitions between these extreme types occur.

D1 and D3 have a sandy bottom. The other D-pools are tarns in small patches of bog, which are

situated in depressions between inland sand dunes (Fig. 3). In some pools the peat has been excavated partially in former times.

The vegetation of most pools belongs to the oligo-dystrophic type (Van Dam & Kooyman-van Blokland 1978). The vegetation of some of the pools in the B- and V-districts belongs to the



*Figs. 2a & 2b.* Part of the clear water moorland pool V1 (Gerritsfles) about 1920 (2a) and in October 1979 (2b). In 1920 the pool was in an open landscape of heath and inland sand dunes, with *Juniperus communis* as the only tree. The open water was poor in macrophytes (some *Carex rostrata*). In 1979 the pool is situated in a *Molinia caerulea* 'heath' with regrowth of *Betula pubescens* and *Pinus sylvestris*. Around the pool is a dense belt of *Juncus effusus* (foreground), followed by a girdle of *Sphagnum* spp., *Eriophorum angustifolium* and *J. bulbosus* (with duck tracks) and a zone with *C. rostrata*, colonizing the formerly open water. Fig. 2a from Thijsse (1926), Fig. 2b photo by Roel van Beek.



**Figs. 3a & 3b.** Part of the brown water moorland pool D4 (Diepveen) before 1920 (3a), and in May 1980 (3b). Before 1920 the pool was still in the open landscape of inland sand dunes, with *Pinus sylvestris* as the solitary tree on the left and a girdle of Scots pines around pool D3 (background right). In 1980 the pool is situated in plantations of Scots pines. Dominant aquatic macrophytes in both figures are *Carex rostrata* and *Eriophorum angustifolium*. Photo: W. Beijerinck (3a), Roel van Beek (3b).

oligotraphentic type, as described by these authors. The most prominent species of this type (*Lobelia dortmanna* L., *Isoetes lacustris* L.) were rather abundant about 1920, but are nearly extinct now.

After 1840 the heathlands and inland sand dunes, which surrounded all the moorland pools were gradually reafforested with Scots pine (*Pinus sylvestris*). The environs of B3 to B6 were already reafforested before 1900 while V1 is still in the open landscape.

### 3. Methods

#### 3.1. Diatoms

Sampling, slide preparation, identification and counting have been described in detail by Van Dam & Kooyman-van Blokland (1978).

Heimans sampled the moorland pools about 1920 with a plankton net, but he certainly did not collect pure plankton. When he hauled his net, he also gathered parts of macrophytes and some bottom material, as can be seen in the samples preserved. The old and recent samples were taken in the same way. The net plankton gives a picture of the microcenoses of the whole pool, which is shallow and well mixed by wind. Sometimes squeezings of *Sphagnum* were gathered.

The most important references for identification were Van der Werff & Huls (1957–1974), Hustedt (1927–1930, 1930, 1931–1959, 1961–1966), Patrick & Reimer (1966, 1975), Cleve-Euler (1951–1955) and Foged (1977). In general the nomenclature of Hustedt (l.c.) was followed. The lanceolate *Nitzschiae* were identified according to Lange-Bertalot (1976) and Lange-Bertalot & Simonsen (1978). In each slide 400 valves were counted, while the whole slide was scanned with a low magnification ( $\times 125$ ) in order to list the most prominent species outside the count.

The pH-spectra were calculated with the ecological data from the literature mentioned by Van Dam & Kooyman-van Blokland (1978). Data about the geographical distribution of species were also borrowed from the literature.

#### 3.2. Chemistry

pH and conductivity were measured on the spot with a Metrohm E 488 pH-meter and a Yellow Springs Instruments model 33 conductivity meter

respectively. The pH-meter was calibrated before and after each measurement. Water samples were collected in polyethylene bottles and stored at 4 °C. Conductivity, pH, nitrite, ammonium, and bicarbonate were analysed the day after sampling; other ions within one week in the laboratory of the 'Waterleidingbedrijf Midden-Nederland'. All analyses were made in unfiltered water, with the exception of sulfate, which was determined gravimetrically after filtration through ordinary filter paper and permanganate consumption (filtered), which was determined after running through an asbestos filter.

Iron, calcium and magnesium were measured by atomic absorption spectrometry (AAS) using a Perkin Elmer model 703. Aluminium was analysed by flameless AAS with a Perkin Elmer model 306. Nitrate was determined according to Müller & Widemann (1955). All other determinations were carried out according to 'Nederlands Normalisatie Instituut' (1965–1968). In the laboratory conductivity was measured at room temperature and converted to conductivity at 25 °C according to Golterman (1969).

#### 3.3. Statistics

Increase and decrease of individual species, ecological and geographical classes were tested by the Wilcoxon signed rank test (SRT), according to Lehmann (1975). Differences of these parameters between the areas (B + V) and D were tested by the Wilcoxon rank sum test (RST), according to Lehmann (1975). These tests were carried out two-tailed.

Principal component analysis was performed with a program written in APL, using an eigenvalue and eigenvector algorithm based on Jacobi's method (Wilkinson 1965). Programs for diversity analysis were written in Fortran IV and for dissimilarity analysis in APL. Batch runs were done at the CDC Cyber 72 computer of TNO (The Hague), interactive computerwork was done at the Cyber 171 of TNO.

### 4. Results

#### 4.1 Taxonomy, ecology and distribution of diatom taxa

The taxonomy and ecology of freshwater diatoms

are relatively rather well known. Most European, and especially Dutch diatoms can be identified easily with the floras mentioned in section 3.1. However, some variable forms are difficult to identify. Therefore the 24 most common taxa are listed here with some annotations about their taxonomy and ecology. All taxa which could be found are listed in the appendix.

*Achnanthes minutissima* Kützing

*A. microcephala* (Kützing) Grunow is included in this taxon because H. Lange-Bertalot (pers. comm.) concludes the type material of both species to be identical. The species occurs from a pH as low as 4.4 (Besch *et al.* 1972) up to 8.4 (Schoeman 1973) and is considered to be pH-indifferent by Foged (1964) and Meriläinen (1967). This seems to be one of the few diatoms which is rather indifferent with respect to pH; in the Netherlands however, high abundances are only observed in slightly acid to alkaline waters and the species is therefore to be classified as circumneutral. Occurs only with small numbers in most of the pools studied.

*Anomoeoneis exilis* f. *lanceolata* Mayet

This taxon is sometimes difficult to distinguish from *A. serians* var. *brachysira* f. *thermalis* (Grunow) Hustedt, which occurred rather rarely. The latter taxon has about 27 transapical striae in 10  $\mu\text{m}$ , the former one more than 30. *A. exilis* f. *lanceolata* is considered to be circumneutral by Foged (1964) and Meriläinen (1967). Renberg (1976) found this taxon abundantly in some lakes with pH 5–6. The distribution in our samples suggests that it does not thrive well in the most acid pools (pH lower than 4.5 to 5).

*Anomoeoneis serians* var. *brachysira* (De Brébisson) Cleve

This variety is considered to be acidophilous by many authors (*e.g.* Jørgensen 1948; Foged 1964; Meriläinen 1967). According to Hustedt (1957) rarely in the North Sea Lowlands and more common in the northern countries. Most abundant in B5, especially old samples.

*Cocconeis placentula* Ehrenberg

We include the varieties in the species, because these are not ecologically different from the typical species (Cholnoky 1968). The species is alkali-

philous and is very common in meso-eutrophic waters (Van der Werff & Huls 1957–1974), although it may be found in small numbers at a pH as low as 4.7 (Jørgensen 1948). In accordance with the literature data the species occurs mostly in small numbers in the pools studied.

*Cymbella microcephala* Grunow

Characteristic for eutrophic alkaline lakes and ponds and therefore alkaliphilous (Jørgensen 1948; Van der Werff & Huls 1957–1974). Occurred sparsely in most moorland pools, but codominant with *Achnanthes minutissima* in an old sample from B5.

*Eunotia alpina* (Nägeli) Hustedt

This species is sometimes difficult to distinguish from long and slender forms of *E. lunaris* (Ehrenberg) Grunow, especially when the valve has a width of ca 3  $\mu\text{m}$ , which is beyond the limit of Hustedt (1931–1959), who gives a maximum width of 2.5  $\mu\text{m}$  for *E. alpina*, but below the upper limit of 3.5  $\mu\text{m}$ , given by Patrick & Reimer (1966) for *E. naegelii* Migula, a synonym for *E. alpina*. In case of doubt we favoured *E. alpina* when the length-to-breadth ratio was more than about 30, in accordance with both Hustedt and Patrick & Reimer.

*E. alpina* is acidophilous and is more common in northern and mountainous areas than in the lowlands (Jørgensen 1948; Hustedt l.c.; Cleve-Euler 1951–1955; Meriläinen 1967). Occurred frequently, but most times with low abundance in the B- and D-samples and rarely in the V-samples. Rather abundant in some recent D-samples.

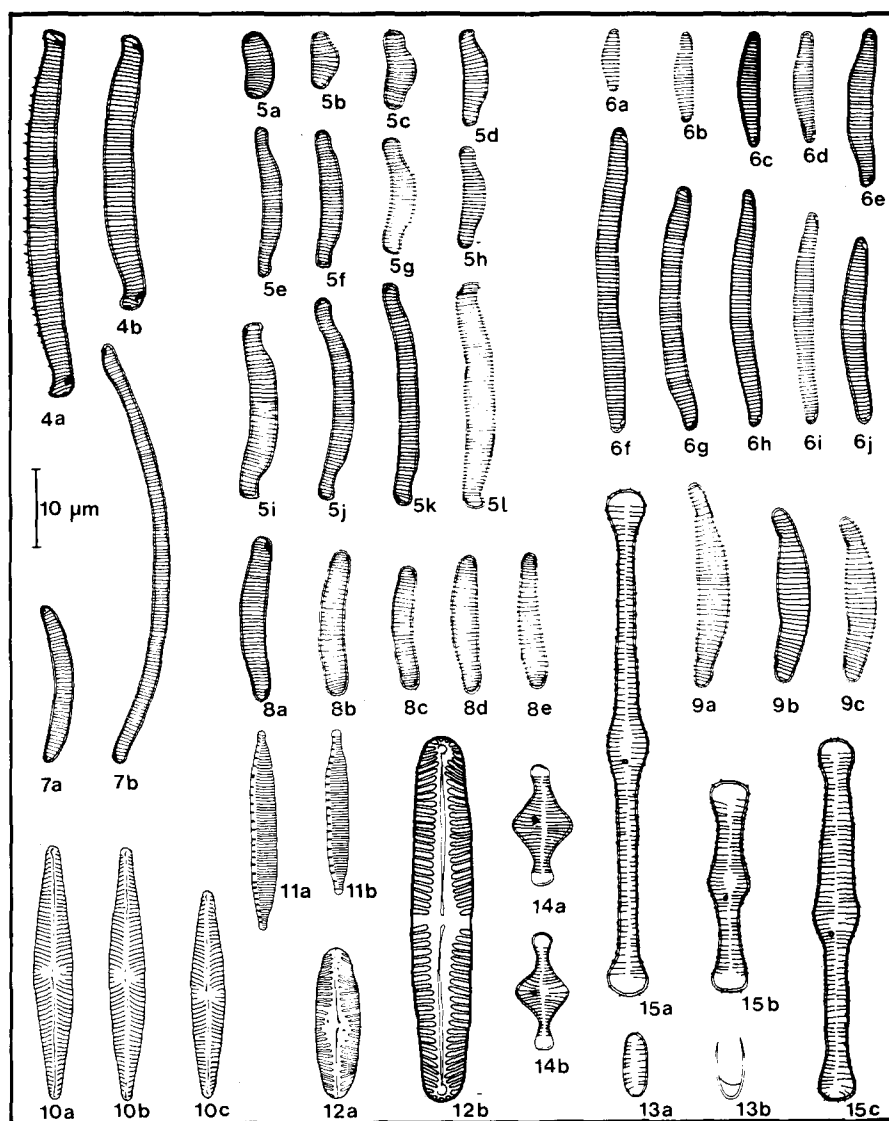
*Eunotia denticulata* (De Brébisson) Rabenhorst

If the small spines are present (Fig. 4a) this diatom is easy to identify. Often some spines or even all have been lost (Fig. 4b). With some experience the last forms can be distinguished by their shape and stronger silification from similar forms, like *E. exigua*.

*E. denticulata* is acidophilous (Meriläinen 1967; Cholnoky 1968) and has its main distribution in northern and mountainous regions. Occurred rather abundant in some old B-samples, recently only in small numbers in a few B- and D-samples.

*Eunotia exigua* (De Brébisson) Rabenhorst

Our concept of this highly variable species is in



Figs. 4–15. 4. *Eunotia denticulata*, 5. *E. exigua*, 6. *E. exigua/tenella* - complex, 7. *E. lunaris*, 8. *E. tenella*, 9. *E. veneris*, 10. *Navicula heimansii*, 11. *Nitzschia perminuta*, 12. *Pinnularia microstauron*, 13. *Tabellaria binalis*, 14. *T. flocculosa*, 15. *T. quadriseptata*.

accordance with Hustedt (1931–1959). The very wide range of this taxon is depicted in Fig. 5. The dimensions of these specimens are: length 6.7–28.9  $\mu\text{m}$ , breadth 2.5–3.8  $\mu\text{m}$  with (18) 20–24 (26) striae in 10  $\mu\text{m}$ . In Fig. 6 the complex of *E. exigua* and *E. tenella* is drawn (length 7.8–39.6  $\mu\text{m}$ , breadth 2.5–3.0  $\mu\text{m}$ , 18–22 striae in 10  $\mu\text{m}$ ). This form is rather similar to *E. tenella*, but the striation is finer than is common for this species (16–20 striae in 10  $\mu\text{m}$ ).

*E. exigua* in natural waters is strongly bound to acid habitats (pools, lakes, bogs and streams) and is therefore acidobiontic (Jørgensen 1948; Symoens 1957; De Graaf 1957; Van der Werff & Huls 1957–1974; Meriläinen 1967; Cholnoky 1968). Such habitats generally have a low mineral content, but this does not mean that *E. exigua* is oligotraphentic. The species thrives equally well in acid mine drainage with a high mineral content and a high load of metals like Mn, Fe, Al, Pb, Co, Cu, Zn and



Ni (Stjerna-Pooth 1953, 1954; Besch *et al.* 1972, Hargreaves *et al.* 1975). It is probably the most resistant diatom for heavy metals. Mass occurrence is reported at a sulfate level of  $1150 \text{ mg l}^{-1}$ , while the upper limit is about  $4000 \text{ mg l}^{-1}$ . No records are made below  $\text{pH} = 2.5$ . Hargreaves *et al.* (l.c.) and Stjerna-Pooth (l.c.) mention that in such stressing environments anomalic and small forms (Fig. 5a, b) are abundant.

The species is present in all but three samples. In old samples it had a mean occurrence of 28 valves, in the recent samples it was present with a mean of 248 valves. In the recent samples the abundance (a) is significantly correlated with the sulfate concentration as follows:  $a = 378 \times \log [\text{SO}_4^{2-}] - 255$  ( $r^2 = 0.67$ ) and more strongly with the quotient  $b = [\text{SO}_4^{2-}] / ([\text{SO}_4^{2-}] + [\text{Cl}^-] + [\text{HCO}_3^-])$ .  $a = 703b - 139$  ( $r^2 = 0.89$ ).

#### *Eunotia lunaris* (Ehrenberg) Grunow

See also the remarks on *E. alpina*. In the counts of this taxon (Fig. 7) the forms belonging to the var. *subarcuata* (Naegeli) Grunow have been included.

*E. lunaris* is circumneutral, but has a slight preference for weakly acid waters (Jørgensen 1948; Hustedt 1957; Patrick & Reimer 1966). The species decreased significantly from a mean of 43 valves in the old samples to a mean of 17 valves in the recent samples.

#### *Eunotia rhomboidea* Hustedt, *E. tenella* (Grunow) Hustedt and *E. veneris* (Kützinger) O. Müller

See also the remarks on *E. exigua*. The three species are closely related and not always easy to distinguish.

One of the first questions which must be solved in order to separate the species in this group properly is whether asymmetry is to be considered as a species character. Most authors depict only symmetrical forms of *E. veneris* (e.g. Hustedt 1931–1959), but in some cases asymmetrical forms are found (e.g. Manguin 1942; Meriläinen 1969). The asymmetrical specimens are rhomboid in girdle view. Van der Werff & Huls (1957–1974) picture symmetrical and asymmetrical forms from the Netherlands. Recently the asymmetrical *E. faba* f. *rhomboidea* Foged, which is commonly found mixed with symmetrical forms of this species, was described (Foged 1972).

Hustedt (1950: 435, T. 34, Fig. 28; T. 36, Fig. 34–41) describes *E. rhomboidea*, which is very similar to *E. tenella*. In Hustedt's type material of *E. rhomboidea* the asymmetry was a constant feature. However, in our material gradual transitions between the two species frequently occurred (Fig. 8). Symmetrical and asymmetrical valves were often rather abundant in the same sample. Any separation of the two 'species' would have been arbitrary. Therefore we include *E. rhomboidea* in *E. tenella*. *E. rhomboidea* cannot be maintained as a species.

Accepting that asymmetry is not a constant feature in some species the second question that must be solved is the distinction between *E. tenella* and *E. veneris*. The terminal nodules, which are removed from the ends of the valve (Fig. 9) are very characteristic for the latter species (Hustedt 1931–1959; Cleve-Euler 1951–1955; Patrick 1958). These nodules are very distinct and at some depths of focus they appear as a notch in the ventral margin (Patrick l.c.; Cleve-Euler l.c.). The ends of the valve are acute to acutely rounded (Hustedt l.c.; Patrick, l.c.). In *E. tenella* (Fig. 8) the terminal raphe nodules are close to the apex, which is rounded (Hustedt l.c.; Petersen 1950; Van der Werff & Huls 1957–1974; Patrick & Reimer 1966). As a thumb rule one may state that *E. veneris* has the nodule on about  $1/8$  of the valve length and *E. tenella* on about  $1/12$  of the valve length. The dimensions in our material are for *E. veneris*: length  $18\text{--}30 \mu\text{m}$ , width  $3.0\text{--}4.8 \mu\text{m}$ ,  $16\text{--}18$  striae in  $10 \mu\text{m}$ , *E. tenella*: length  $10\text{--}21 \mu\text{m}$ , width  $2.9\text{--}4.8 \mu\text{m}$ ,  $19\text{--}20$  striae in  $10 \mu\text{m}$ .

At least two authors do not separate the asymmetrical forms in this group according to the above mentioned criteria. Foged (1950, T. 3, Fig. 8, 9; 1977, T. 10, Fig. 27; in Berge, 1979, T. 1, Fig. 23, T. 3, Fig. 18) and Wuthrich (1975, T. 9; Fig. 1, 4–12) depict asymmetrical specimens as *E. rhomboidea* which fit in our concept of *E. veneris*. The specimens of Foged (1950, T. 3, Fig. 6, 7, 11; 1977, T. 10, Fig. 26) and Wuthrich (1975, T. 9, Fig. 2, 3) fit in our concept of *E. tenella*.

*E. tenella* (including *E. rhomboidea*) is acidophilous (Jørgensen 1948; Foged 1950; Hustedt 1957; Meriläinen 1967, 1969). The asymmetrical forms are found in acid, oligotrophic to dystrophic small *Sphagnum*–*Carex rostrata* – and *Lobelia* – lakes in north-western Germany and southern

Scandinavia (Foged 1950; Hustedt 1950; Behre 1956). Such lakes are very similar to our moorland pools. *E. veneris* is also acidophilous and has a mountainous tendency (Hustedt, 1931–1959; Jørgensen 1948; Symoens 1957; Van der Werff & Huls 1957–1974; Meriläinen 1967). The optimal pH is probably slightly higher than for *E. tenella*.

Both species were found in most samples of the study area. *E. tenella* is slightly (not significantly) less abundant in the recent samples than in the old ones. *E. veneris* decreased significantly from a mean of 57 valves in the old to three valves in the recent ones.

#### *Fragilaria capucina* Desmazières

Alkaliphilous (Jørgensen 1948; Hustedt 1957; Van der Werff & Huls 1957–1974). Occurs erratically. Rather abundant (39 valves) in a recent B1-sample.

#### *Fragilaria virescens* Ralfs

Circumneutral (Hustedt 1957; Meriläinen 1967), but best development in slightly acid waters (Schroeder 1939; Scheele 1952; Salden 1978). Most abundant in the alpine and northern areas of Europe (Hustedt 1957; Symoens 1957). This species decreased significantly in the (B + V)-samples between 1920 and 1978. It was not present in the old D-samples, but occurred rather abundantly in the recent samples D1 and D3.

#### *Frustulia rhomboides* var. *saxonica* (Rabenhorst) De Toni

This taxon is considered to be acidobiontic by Hustedt (1957) and De Graaf (1957) but acidophilous by other authors (e.g. Jørgensen 1948; Van der Werff & Huls 1957–1974; Meriläinen 1967). We share the first opinion, because the taxon is reported to be most abundant in humic acid waters. Cholnoky (1958), Harrison (1958) and Bennett (1968) found mass development of *F. rhomboides* var. *saxonica* in rivers that were polluted with acid mine drainage. Silfversparre (1937), Valin (1958) and Eloranta & Kettunen (1979) mention either the species or the variety, which behave ecologically very similar, to be very common in regions of sulfite discharge from wood pulp factories. Thus one would predict this taxon to increase with growing acidity of the water, like *Eunotia exigua*.

However, in the study area the mean abundance decreases significantly from 89 valves in old samples to 39 in the recent ones. It is more abundant in the samples of the Drenthian district, that is generally rich in humic acids, than in the other samples, which are from relatively humic poor water.

#### *Navicula heimansii* Van Dam & Kooyman

This species (Fig. 10) is described by Van Dam & Kooyman (in prep.). The dimensions are: length 28–35  $\mu\text{m}$ , breadth 4.5–6  $\mu\text{m}$ , 16–18 striae in 10  $\mu\text{m}$ .

The species was not found in the V-samples. Occurs with low numbers in some B- and D-samples. Abundant in an old sample of B5 and a recent sample of D2. Possibly acidophilous. To avoid circular reasoning it would have been better to omit this species from the calculations of the pH-spectra, but this does not influence the results significantly.

#### *Navicula subtilissima* Cleve

This typical inhabitant of bogs and nutrient poor lakes is often found with *Frustulia rhomboides* var. *saxonica* and small species of *Eunotia* (Hustedt, 1961–1966) and therefore classified as acidobiontic by De Graaf (1957) and Meriläinen (1967). After having done the main body of the research it unfortunately appeared that the rather similar *Navicula hoefleri* Cholnoky was included in the counts of *N. subtilissima*. *N. hoefleri* is acidobiontic (Hustedt l.c.).

Both species decrease significantly in the B- and V-samples between 1920 and 1978. No significant changes occurred in the D-samples.

#### *Nitzschia gracilis* Hantzsch

Circumneutral according to Jørgensen (1948), Hustedt (1957), Foged (1964) and Meriläinen (1969). Occurred in low abundances in a number of samples; has a tendency to decrease from 1920 to 1978. Most abundant in an old sample of V2.

#### *Nitzschia perminuta* Grunow

Fig. 11. Valves slightly constricted in the middle, 20–25  $\mu\text{m}$  long, about 3  $\mu\text{m}$  wide. Transapical striae very finely punctated, about 30 in 10  $\mu\text{m}$ . Keel very excentric, with 9–11 fibulae in 10  $\mu\text{m}$ . Central fibulae equidistant. The specimens correspond well with those of Hustedt (1943: 231, Fig. 80, 82–87) and specimens of the type slide of Grunow (Lange-

Bertalot & Simonsen 1978: 85, Fig. 160a, b).

Considered to be alkaliphilous (Hustedt 1943). The species occurred erratically in some B-, D- and V-samples and was commoner in the old B-samples than in the recent ones. Abundant in an old sample of B5.

*Pinnularia appendiculata* (Agardh) Cleve

Recorded by Jørgensen (1948) as acidophilous. Cholnoky (1968) gives an optimal pH of 6.5–6.8. Regarded as circumneutral by other authors (Hustedt 1957; Foged 1964) as the species may be present in alkaline waters, but generally in low numbers (Schoeman 1973). In our experience the species has its highest abundance in acid waters and therefore we consider the taxon to be acidophilous.

Relatively scarce in the samples, but rather abundant in an old sample from V1. Decreases significantly from 1920 to 1978, especially in the D-samples.

*Pinnularia microstauron* (Ehrenberg) Cleve

Fig. 12. This species is extremely variable (see e.g. Hustedt 1930; Lund 1946; Meriläinen 1969; Foged 1974, 1977). Fig. 12b represents a typical form of the species, while Fig. 12a is related to var. *brebissonii* (Kützing) Hustedt. All intergradations between these two extremes were found.

The species and its variety are circumneutral (Hustedt 1957; Foged 1964) but have an optimal development in weakly acid waters (Jørgensen 1948; Van der Werff & Huls 1957–1974; Patrick & Reimer 1966; Cholnoky 1968).

Scattered occurrence over the whole study area, has a tendency to decrease from 1920 to 1978. Most abundant (73 valves) in V1 in 1960.

*Tabellaria binalis* (Ehrenberg) Grunow

Fig. 13. The Dutch material is elliptical in valve view and similar to the Danish forms (Jørgensen 1948). However, there is no reason to separate this form from the typical constricted form as figured by Hustedt (1931–1959), because there is a gradual transition from constricted to elliptical forms (Cleve-Euler 1951–1955).

*T. binalis* is a typical inhabitant of *Lobelia*-lakes in the former heath- and moorland area of north-western Europe and is also found in Scandinavian *Lobelia*-lakes. Acidobiontic (Hustedt 1931–1959; Jørgensen 1948; Cleve-Euler 1951–1955). The rela-

tive abundance in some cores from acidified Scandinavian lakes increased during the last decennia (Miller 1973; Berge 1979).

Regularly present in the old B-samples. Mass occurrence in B8. Recently only a few frustules in the study are.

*Tabellaria flocculosa* (Roth) Kützing

Fig. 14. Acidophilous (Jørgensen 1948; Hustedt 1957, Van der Werff & Huls 1957–1974). Patrick & Reimer (1966) state that the short individuals (as in our figure) are more often found in acid water of bogs and ponds, whereas the longer ones seem to be more often found in oligotrophic to mesotrophic water.

In rather small quantities in most of the samples, rather abundant in the old samples from B2 and D1.

*Tabellaria quadrisepitata* Knudson

Fig. 15. This species is often confused with *T. fenestrata* (Lyngbye) Kützing (see e.g. Van Dam & Kooyman-van Blokland 1978). Characteristic features of *T. quadrisepitata* are: the gradually tapering terminal inflations, the peripheral position of the mucilage pore and the marginal spines, which are visible in the light microscope (Knudson 1952; Patrick & Reimer 1966; Koppen 1975).

The species is confined to acid (dystrophic) water with a low mineral content (Knudson 1954; Florin 1957; Patrick & Reimer l.c.; Plinski & Witek 1976; Sims 1978) and we therefore classify the species as acidobiontic. In the study area the species was present in nearly all samples and had a tendency to decrease in the B-samples from 1920 to 1978.

In total 194 species and varieties, belonging to 36 genera were found. All taxa are listed in the appendix, together with their pH-class and notes about geographical distribution. The distribution of 24 taxa occurring at least with 20 valves in one sample, is listed in Table 2. The samples are in the order of their score on the first principal component, and the species in the order of their loadings on this component (see section 4.5).

The increase or decrease of the species from 1920 to 1978 was tested by SRT. When two or more samples of one pool at one moment were available, one sample was drawn at random. The selected samples are indicated in Table 2. The species with a

Table 2. Number of valves per species in each sample. + = observed outside the count, - = not observed, BA = *Batrachospermum*-squeezing, BM = bottom mud, BS = bottom sand, EP = epiphytes from *Littorella uniflora*, *Lobelia dortmanna* or *Juncus bulbosus*, NP = net plankton, SP = sedimentation plankton, SQ = squeezing of *Juncus bulbosus* and *Sphagnum* spec., SS = *Sphagnum*-squeezing, UT = squeezing of *Utricularia minor*. Closed symbols: old samples used for pairwise comparison, open symbols: recent samples used for pairwise comparison.

pairwise comparison	type of sample	month and year	pool number	<i>Frustulia rhomboides</i> var. <i>saxonica</i>	<i>Eunotia veneris</i>	<i>Eunotia lunaris</i>	<i>Eunotia tenella</i>	<i>Tabellaria quadriseptata</i>	<i>Navicula subtilissima</i>	<i>Tabellaria binalis</i>	<i>Navicula heilmannii</i>	<i>Tabellaria flocculosa</i>	<i>Anomooneis exilis</i> f. <i>lancoolata</i>	<i>Eunotia denticulata</i>	<i>Achnanthes minutissima</i>	<i>Eunotia alpina</i>	<i>Fragilaria virescens</i>	<i>Cymbella microcephala</i>	<i>Pinnularia appendiculata</i>	<i>Pinnularia microstauron</i>	<i>Nitzschia gracilis</i>	<i>Nitzschia perminuta</i>	<i>Fragilaria capucina</i>	<i>Eunotia tenella</i> / <i>exigua</i> complex	<i>Cocconeis placentula</i>	<i>Anomooneis seriata</i> var. <i>brachysira</i>	<i>Eunotia exigua</i>
■	NP	8.22	D3	232	63	36	2	28	7	-	9	7	1	-	-	-	-	-	1	2	-	-	-	-	-	-	1
■	NP	8.33	D6	239	47	50	6	10	+	-	1	-	-	-	4	-	-	-	7	16	-	-	-	7	-	-	1
	BA	6.16	V1	66	9	310	-	-	14	-	-	-	-	-	-	-	-	-	1	-	-	-	-	-	-	-	-
■	NP	8.24	D4	33	255	55	1	-	7	-	-	+	-	-	9	4	-	1	+	-	-	-	-	-	-	-	2
	NP	6.16	V1	18	289	45	17	2	3	-	-	13	-	-	-	+	+	-	-	+	-	-	+	-	-	-	6
▼	NP	7.18	V2	41	187	13	-	28	60	-	-	9	2	-	3	-	-	-	-	-	25	-	-	-	-	-	3
■	NP	8.24	D1	56	96	35	33	22	61	-	2	72	1	-	10	-	-	-	+	-	-	+	-	-	-	-	+
■	NP	6.29	D5	283	+	56	+	-	-	-	+	+	-	-	-	+	-	-	-	-	-	1	-	17	-	-	26
	NP	10.21	B5E	113	41	14	4	70	22	-	92	1	17	+	2	11	-	-	+	-	+	-	-	-	-	-	3
●	NP	8.22	B7	122	-	10	8	75	2	-	1	+	-	156	5	10	-	-	-	-	1	-	-	-	+	-	4
●	NP	7.25	B2	50	39	93	8	58	68	-	-	53	-	-	3	1	2	1	-	+	6	-	-	-	-	-	7
	NP	7.18	V1	25	76	60	114	3	52	-	-	10	34	-	-	-	-	-	-	2	4	-	-	-	-	-	6
□	NP	11.78	D5	202	4	42	3	4	-	1	-	+	-	-	6	109	-	-	-	-	-	1	-	3	-	-	19
	EP	7.29	B8	3	101	-	6	13	1	252	-	1	-	-	-	+	4	-	-	1	-	-	-	-	-	-	3
	EP	7.29	B8	17	53	3	3	22	5	272	3	3	-	-	+	+	1	+	-	+	-	-	-	-	-	-	1
■	NP	8.24	D2	180	88	29	9	29	+	-	-	-	-	-	1	4	-	-	+	1	-	-	-	-	21	-	30
▼	NP	7.18	V1	24	27	99	72	-	65	-	-	23	36	-	8	-	1	-	-	6	6	-	-	-	-	-	5
	NP	8.50	B5B	33	59	22	16	21	11	-	9	5	44	-	6	-	74	4	-	2	5	1	-	-	-	39	-
	NP	9.19	B6	87	11	1	16	131	34	-	7	2	1	55	-	14	1	-	-	+	1	-	-	-	-	4	10
	UT	7.16	V1	24	54	87	105	-	1	-	-	+	-	-	-	1	-	-	90	2	-	-	-	4	-	-	11
	NP	6.52	B5A	10	27	132	33	2	2	-	4	3	11	-	44	5	13	4	3	+	6	1	1	-	-	-	6
□	NP	11.78	D1	28	20	10	31	4	114	-	98	9	22	-	-	-	30	-	-	+	1	-	-	-	+	-	1
	NP	9.50	V1	36	80	10	203	10	6	-	-	15	6	-	-	-	-	-	+	1	-	-	-	-	+	-	16
●	NP	9.20	B6	88	7	4	15	221	8	+	-	-	-	25	-	7	-	-	1	+	-	-	-	-	-	-	16
□	NP	11.78	D2	99	2	41	25	135	8	-	-	1	-	3	5	42	-	-	+	-	1	-	-	-	+	-	19
●	NP	9.26	B5B	29	5	4	6	1	4	-	120	6	53	-	2	2	64	3	+	+	6	48	-	-	+	6	-
●	NP	6.19	B1	52	2	3	83	33	2	75	-	21	-	-	8	3	-	-	-	2	1	+	39	-	-	-	15
	BM	7.18	V1	20	-	65	131	9	61	-	-	20	33	-	1	-	-	-	-	-	+	-	-	-	-	-	18
	NP	7.60	V1	49	37	25	159	7	2	+	-	10	3	-	3	-	-	-	1	73	-	-	-	-	-	-	24
	NP	12.19	B5A	3	+	+	1	1	1	-	+	1	38	4	189	2	-	145	-	-	-	+	2	-	-	-	10
	NP	9.50	V1	121	43	7	113	10	-	+	-	17	+	-	1	1	-	-	-	-	-	-	-	+	-	-	86

Table 2. (Continued).

pairwise comparison	type of sample	month and year	pool number	<i>Frustulia rhomboides</i> var. <i>saxonica</i>	<i>Eunotia veneris</i>	<i>Eunotia lunaris</i>	<i>Eunotia tenella</i>	<i>Tabellaria quadrisepiata</i>	<i>Navicula subtilissima</i>	<i>Tabellaria binialis</i>	<i>Navicula heimansii</i>	<i>Tabellaria flocculosa</i>	<i>Anomooneis exilis</i> f. <i>lanceolata</i>	<i>Eunotia denticulata</i>	<i>Achnanthes minutissima</i>	<i>Eunotia alpina</i>	<i>Fragilaria virescens</i>	<i>Cymbella microcephala</i>	<i>Pinnularia appendiculata</i>	<i>Pinnularia microstauron</i>	<i>Nitzschia gracilis</i>	<i>Nitzschia perminuta</i>	<i>Fragilaria capucina</i>	<i>Eunotia tenella</i> / <i>exigua</i> complex	<i>Cocconeis placentula</i>	<i>Anomooneis seriatus</i> var. <i>brachysira</i>	<i>Eunotia exigua</i>
□	NP	11.78	D4	97	1	70	1	90	2	-	1	1	-	-	2	4	-	-	-	-	-	1	-	-	6	-	111
□	NP	11.78	D3	48	8	11	113	21	16	-	12	5	4	-	-	-	4	-	-	+	10	1	-	5	+	+	111
●	NP	6.16	B3	6	+	+	29	196	+	+	1	+	-	-	+	-	1	-	-	+	-	2	-	+	-	-	163
○	NP	11.78	B2	14	+	+	+	143	-	-	-	5	-	-	8	-	+	2	1	1	-	-	-	2	-	-	174
●	NP	4.26	B4	33	-	2	45	13	2	+	8	1	-	24	+	26	1	-	-	7	-	-	23	-	-	-	177
	NP	7.73	V2	50	40	12	44	18	1	-	-	2	-	-	-	10	-	-	-	3	6	-	-	-	-	-	210
	NP	11.78	B5A	19	5	4	21	19	12	-	+	2	7	-	1	-	20	+	-	-	-	+	-	+	19	-	256
	BM	11.77	V1	11	12	5	52	11	6	1	-	4	11	-	-	-	-	-	-	1	1	-	-	-	-	-	272
○	NP	11.78	B7	26	2	6	46	10	1	-	-	2	-	-	5	3	+	-	-	1	-	-	-	-	2	+	276
	NP	11.78	B5E	12	12	2	31	12	4	-	3	1	4	+	-	-	+	-	-	1	-	-	-	-	+	13	291
□	NP	11.78	D6	62	1	1	13	2	-	-	+	1	-	+	2	+	-	-	-	-	-	-	-	-	+	+	311
○	NP	11.78	B1	21	-	+	15	+	-	1	-	-	-	-	1	-	-	-	-	-	-	-	-	-	2	-	307
○	SQ	11.78	B8	6	+	+	3	+	-	-	-	-	-	-	21	-	-	-	-	+	-	-	1	-	1	-	343
○	NP	11.78	B5B	5	3	3	5	5	+	-	2	1	4	-	+	-	7	1	-	+	+	2	-	-	+	3	352
	NP	11.78	V1	-	6	1	10	5	2	1	-	3	2	-	2	-	-	-	-	-	-	2	-	-	-	-	355
	BS	11.78	V1	5	2	-	19	5	-	-	-	2	+	-	-	-	-	-	+	+	-	-	-	-	-	-	364
	NP	11.77	V1	7	-	3	7	1	1	-	-	+	6	-	1	-	-	-	-	2	-	-	-	-	+	-	371
	NP	11.78	B8	2	+	+	12	8	-	-	-	-	-	-	6	-	-	-	-	+	-	-	+	-	+	-	372
○	NP	11.78	B4	3	+	+	4	1	-	-	1	+	-	-	-	-	+	+	-	+	-	-	-	-	-	-	375
	NP	10.65	V1	1	+	-	7	15	-	-	-	-	-	-	-	-	-	-	-	1	-	-	-	-	-	-	376
	NP	11.78	V1	3	2	+	11	+	2	-	-	2	-	-	+	-	-	-	+	+	-	-	-	-	-	-	378
○	SS	11.78	B3	14	-	+	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	386
○	NP	11.78	B6	+	3	-	5	+	1	+	-	1	+	-	-	1	-	-	+	+	-	-	-	-	+	+	385
	SP	11.78	V1	1	1	-	4	+	-	-	-	+	-	-	-	-	-	-	-	+	-	-	-	-	-	-	394
▼	NP	11.77	V2	+	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	+	-	400
▼	NP	11.78	V1	+	-	-	-	+	-	-	-	-	-	-	-	-	-	-	-	+	-	-	-	-	-	-	400

Table 3. Species which significantly changed in abundance between 1920 and 1978.

Increased	P	Decreased	P
<i>Anomoeoneis exilis</i>	0.026	<i>Eunotia lunaris</i>	0.016
<i>Cocconeis placentula</i>	0.036	<i>Eunotia veneris</i>	0.034
<i>Cyclotella meneghiniana</i>	0.025	<i>Frustulia rhomboides</i>	
<i>Eunotia exigua</i>	0.001	var. <i>saxonica</i>	0.004
<i>Eunotia polydentula</i>	0.046	<i>Surirella arctissima</i>	0.008
<i>Fragilaria vaucheriae</i>	0.007		
<i>Navicula minima</i>	0.042		
<i>Nitzschia gandersheimiensis</i>	0.024		
<i>Rhoicosphenia curvata</i>	0.029		
<i>Stauroneis phoenicenteron</i>	0.044		

significant change in distribution are listed in Table 3. *Eunotia denticulata*, *Fragilaria virescens* and *Tabellaria binalis* occurred rather abundantly in some pools about 1920 and were very rare in 1978, when they occurred sparsely in some pools in which they were absent before. As the test is based on ranking, the remarkable decrease is not reflected by the results of the test.

It is noticeable that most of the increased species are typical for eutrophic, circumneutral or alkaline waters. Only *Eunotia polydentula* and *E. exigua* are species of acid water. From the description of the autecology of the latter species it appears that also this species is often found in eutrophic waters. All the decreased species are indicative for an acid, oligo-mesotrophic environment.

In each sample the abundance of the species with an alpine, boreo-alpine, boreal or mountainous distribution in Europe (marked by M in the appendix) was added. These abundances were compared pairwise. Again with the SRT and the hypothesis that there was no change in the abundance of M-species there is a significant ( $P < 0.01$ ) decrease of these species. The optimal development of M-species is in slightly acid water that is poor to moderately poor in nutrients. A decline of these species means either a deviation from the optimal pH or eutrophication or both.

#### 4.2 pH-spectra

pH-spectra of one old and one recent sample of each moorland pool are given in Fig. 16. When two or more samples of one pool in the same period were available, one sample was drawn at random.

Nygaard's (1959) method to estimate the pH quantitatively from the pH-spectrum according to Hustedt (1939) was gauged in Finnish lakes by Meriläinen (1967). From the latter publication it appears that Nygaard's indices  $\alpha$ ,  $\epsilon$  and  $\omega$  are not suitable for the determination of the pH from pH-spectra when the pH is less than about 5.

Nevertheless it is useful to summarize the whole pH-spectrum in one number, say R. If we assign the numbers 1, 2, 3, 4 and 5 to the pH-classes acidobiontic, acidophilous, circumneutral, alkaliphilous and alkalibiontic respectively and suppose the number of valves in each of these classes to be  $n_1$ ,  $n_2$ ,  $n_3$ ,  $n_4$  and  $n_5$  respectively, then  $R = \sum n_i / \sum n_i$ . The values for  $R_o$  and  $R_r$  (o = old, r = recent) are printed in the top right corner of each diagram in Fig. 16.

All B- and V-pools acidified more or less. In pool B8 R decreases only very slightly. However, acidobiontic species increase considerably in this pool too, at the cost of acidophilous species. This increase is counterbalanced by an increase of species from neutral or alkaline, eutrophic waters (e.g. *Achnanthes lanceolata*, *Navicula seminulum* and *Rhoicosphenia curvata*). The recent pH-spectra of B8, B2 and B4 are distinctly bimodal.

All old B- and V-spectra are unimodal. This may indicate that the water contains enough nutrients for eutraphentic species, but these cannot thrive well because of the acid conditions.

Old and recent pH-spectra of D1 and D2 are very similar: apparently no acidification occurred. In D3 and D5 the spectra shift to the right. These pools are now less acid than in earlier days. D4 and D6 are more acid now than in former times.

The differences  $R_o - R_r$  were tested by SRT. In all pools together R decreased significantly ( $P = 0.002$ ). In the group (B + V) the decrease is even more significant ( $P = 0.001$ ), but not in D ( $P = 0.34$ ). From RST it appears that  $R_o$  differs not significantly between the two groups and that  $R_r$  is significantly lower in (B + V) than in D ( $P < 0.05$ ).

The old pH-spectra are more diverse than the recent ones. The latter ones are very similar. In earlier days the diversity of pH-spectra was greater within the (B + V)-group than within the D-group. Now the situation is reversed.

For the present it is not possible to estimate the pH with R. From visual inspection of Fig. 16 one gets the impression that the pH has fallen 1 to 2

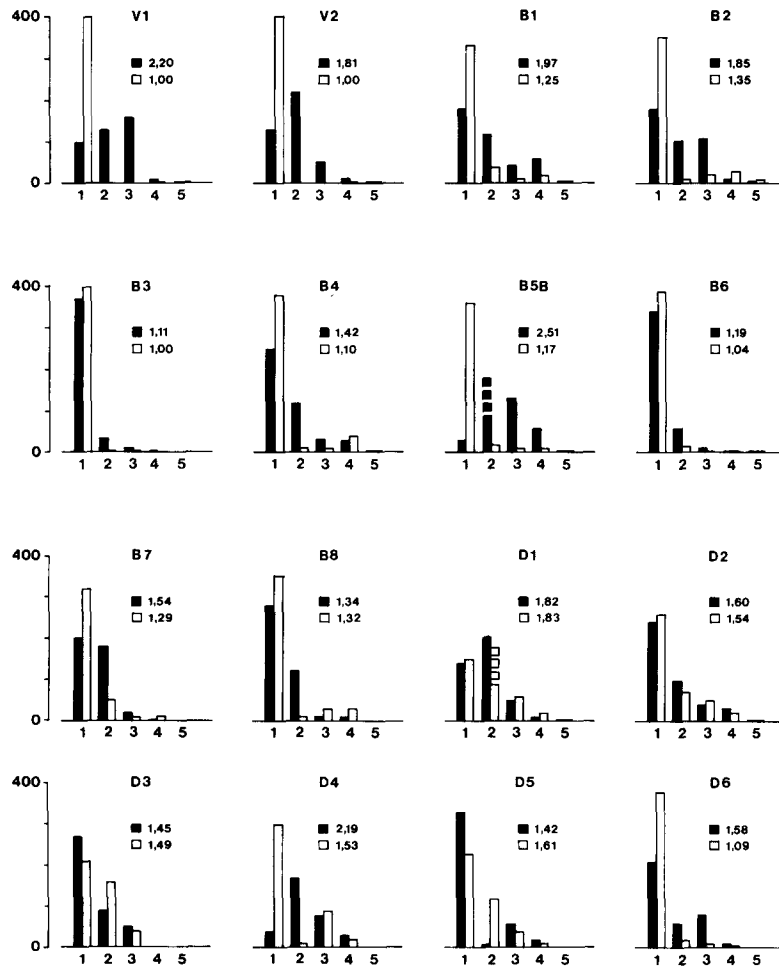


Fig. 16. Histograms of pH-spectra. Ordinate: number of valves in each pH-class. Abscissa: pH-classes (1 = acidobiontic, 2 = acidophilous, 3 = circumneutral, 4 = alkaliphilous, 5 = alkalibiontic). Black bars ca 1920, white bars 1978. Broken bars represent *Navicula heimansii* of which the ecological position is still uncertain. In top right corner of each diagram  $R_o$  (upper number) and  $R_r$  (lower number).

points in the most acidified pools (V1, V2 and B5). In the other B- and V-pools the decline is 0.5 to 1 point.

From several pools more than one recent and old sample is available. For B5 and V1 the pH-spectra are given in Fig. 17 and 18. Data for the period between ca 1920 and 1978 have been included in these figures too. The stations A, B and E in B5 are arranged in a transect from west to east. In about 1920 there was a clear pH-gradient in this direction. This gradient seems to persist until ca 1950. However, this is difficult to assess because of the lack of data at station B. In 1950 the pH was already lower

than in 1920. The decrease continues after 1950. Now the differences between the stations are definitely less distinct than in 1920.

The same process can be observed in V1 (Fig. 18). In contrast with B5, the exact localities of the old sample stations are not known. It is sure that these samples were taken at different places, just as in 1977 and 1978. The increase of acidobiontic species is obvious. There seems to be a turnover point between 1960 and 1965. However, these data have to be interpreted cautiously because of the high variability of the abundance of acidobiontic diatoms in the middle-period.

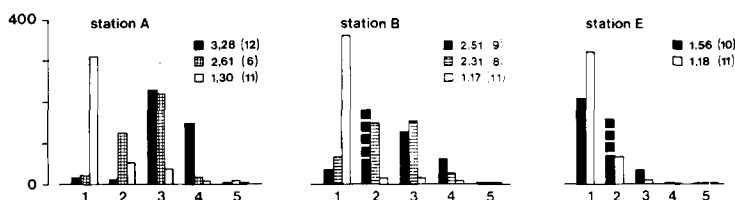


Fig. 17. Histograms of pH-spectra for three stations in B5. Dotted bars ca 1950. Central number in top right corner of each diagram:  $R_{1950}$ . Between parentheses month of sampling. Other symbols as in Fig. 16.

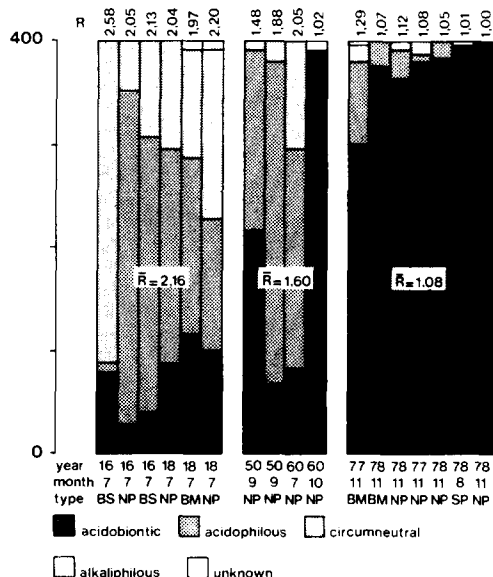


Fig. 18. Histograms of pH-spectra for all samples from VI (1916-1978). Each column represents one sample. The number of valves in each sample is indicated.  $\bar{R}$  is the mean  $R$  for each period. Type of sample: see Table 1.

From Fig. 17 and 18 it also appears that seasonal variations are less important than are changes in the long run.

#### 4.3 Diversity

Indices of diversity are meant to summarize the species-abundance distribution in one number. Some indices, e.g. the number of species in the sample, are especially susceptible to the contribution of rare species; others, e.g. Simpson's (1949) measure of concentration, are more sensitive to the contribution of the most abundant species. The family of diversity indices suggested by Hill (1973) makes it possible to examine these different aspects

of the vector of abundances of a sample in a systematical way (Kempton 1979). Hill's family is defined by  $\Delta_a = (\sum \pi_i^a)^{1/(1-a)}$ , where  $(\pi_1, \pi_2, \dots, \pi_T)$  is the vector of relative abundances of species in the sample;  $a$  may be any real number. Some special cases are:

- $\Delta_{-\infty}$  : the reciprocal of the relative abundance of the rarest species in the sample
- $\Delta_0 = T$  : the number of species in the sample
- $\Delta_1 = e^H$  :  $H = -\sum \pi_i \ln \pi_i$ , the Shannon-index
- $\Delta_2 = (\sum \pi_i^2)^{-1}$  : the reciprocal of Simpson's measure of concentration
- $\Delta_{\infty}$  : the reciprocal of the relative abundance of the commonest species (the dominance index of Berger & Parker, 1970).

Thus  $\Delta_a$  becomes more sensitive for the contribution of dominant species when  $a$  increases. Low values of  $a$  emphasize the richness component of diversity, high values of  $a$  stress the evenness component.

The median diversities of old and recent samples from the (B + V)-pools and D-pools are given in Fig. 19. In addition to  $\Delta_a$  also  $S$ , i.e. the number of all the species recorded in the inspected sample, is given. It appears that  $S$  within the group (B + V) was greater in 1978 than in 1920, although the difference is not significant ( $P = 0.09$ , SRT). For  $a \geq 0$   $\Delta_a$  significantly decreases in this group ( $P < 0.01$ ).  $\Delta_a$  (including  $S$ ) decreases in D for all  $a$ , but not significantly ( $P > 0.25$ ). From RST it appears that the diversity of the groups (B + V) and D did not significantly differ in 1920, but in 1978 the diversity in (B + V) is smaller than in D for  $a \geq 0.25$  ( $P < 0.05$ ).

From the stations B5A and B5B samples are available from ca 1950 (Table 2). The diversities for all  $a$  (including  $S$ ) are higher in 1978 than about



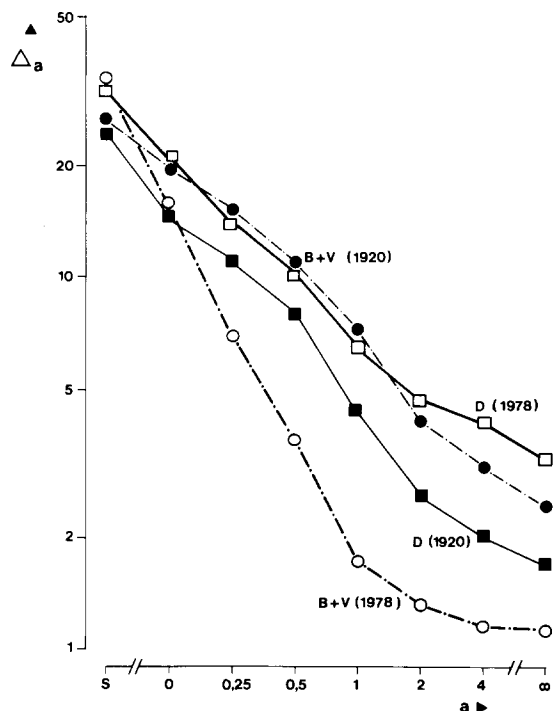


Fig. 19. Logarithmic plot of median Hill diversity ( $\Delta_a$ ). S = all species observed in slide.

1920. The diversities of the period in between are higher than in other periods. In V1 several samples for all periods are available (Table 2). In this pool  $\Delta_a$  gradually declines with time. The differences between ca 1920 and 1978 are significant for  $a \geq 0.50$  ( $P < 0.05$ , RST).

#### 4.4 Dissimilarity

As Dyer (1978) stresses, a major drawback to most applications of (dis)similarity indices has been the general inadequacy or complete omission of formal statistical analysis. He develops a method to estimate the variance of the mean of dissimilarities by allowing for the covariance between dissimilarities that have one sample in common. This method will be applied on two measures which differ in their sensitivity for the contribution of rare species.

The number of species in- and outside the count that is common to both samples in examination, say P, weighs all the species, regardless of their abundance, equally well. Strictly speaking P is a similarity measure, but the statistics can also be

used in this case (Dyer 1978). Q is the index as suggested by Dyer (1978, formula 13), which is related to Simpson's (1949) measure of concentration and stresses the contribution of the dominant species. Q has an expected value of zero if the organisms are dispersed randomly.

Four different kinds of mean dissimilarities are of interest. They are the mean (o) of the dissimilarities of old samples, with a typical element denoted by  $d_{KL}$ , the mean (r) of the dissimilarities of recent samples, with a typical element denoted by  $d_{kl}$ , the mean (o-r) of the changes in dissimilarity, i.e. the differences  $d_{KL} - d_{kl}$  for all K, k, L and l, and the mean (z) of the values  $d_{Kl} + d_{kL} - d_{kl} - d_{KL}$  for all K, k, L and l. z may be interpreted as the shift of the centroid of species composition with time. The variances of these means are calculated by Dyer's method and tests are based on these variances.

The results of the calculations for the 16 pairs of samples which are marked in Table 2 are given in Table 4. Let us denote the area by a superscript and the group by a subscript.  $P^{B+V}$  does not change significantly with time, because  $\hat{\beta}_{o-r}^{B+V}$  is smaller than twice the square root of its variance. Nevertheless there are changes, because  $(\hat{\sigma}^2)^{B+V}_r > (\hat{\sigma}^2)^{B+V}_o$ . The increase of  $\hat{\rho}^{B+V}$  with time indicates that two dissimilarities with one sample in common are more interdependent in the recent (B + V)-samples than in the old (B + V)-samples. Besides, there is a significant change in the place of the centroid of species composition. For P no significant changes are found in the area D.

The Dyer dissimilarity Q decreases significantly in the (B + V)-pools. Also z is significant. Q does not significantly increase in the D-pools, but the change in species composition is significant.

Thus, changes are most obvious in Q, which stresses the differences which are seen at a rather rough inspection of the slides. The differences fade away with more careful inspection, as is the case with P.

#### 4.5 Principal component analysis

Principal component analysis (PCA) visualizes the position of diatom samples in the vector space of species in a graphical display. Dissimilarity and diversity can approximately be read from the graph (Ter Braak, in prep.).

Table 4. Dissimilarity statistics.

$\hat{\beta}$ : estimate of mean dissimilarity (Dyer 1978, formula 9)  
 var ( $\hat{\beta}$ ): estimate of variance of  $\hat{\beta}$  (Dyer 1978, formula 12)  
 $\hat{\sigma}^2$ : variance of the dissimilarities (Dyer 1978, formula 10)  
 $\hat{\rho}$ : estimate of covariance between two dissimilarities which have one sample in common  
 \*: significant at  $P < 0.05$  level

Area	Group	$\hat{\beta}$	Var ( $\hat{\beta}$ )	$\hat{\sigma}^2$	$\hat{\rho}$
P: number of species in common					
B + V	o	13.36	1.304	7.12	1.84
	r	10.98	6.427	32.11	9.24
	o-r	2.37	5.614	31.75	7.84
	z	-2.27*	1.084	10.72	1.23
D	o	12.40	6.382	15.44	2.86
	r	16.67	3.067	13.02	0.67
	o-r	-4.27	8.271	26.86	2.85
	z	1.93	-0.040(1.52) <sup>a</sup>	9.13	-1.17
Q: Dyer dissimilarity					
B + V	o	0.20	0.0011	0.0071	0.0016
	r	0.05	0.0010	0.0048	0.0015
	o-r	0.15*	0.0032	0.0160	0.0045
	z	0.59*	0.0115	0.0498	0.0170
D	o	0.13	0.0033	0.0125	0.0009
	r	0.18	0.0038	0.0102	0.0016
	o-r	-0.05	0.0095	0.0293	0.0035
	z	0.10*	0.0022	0.0113	0.0002

<sup>a</sup> between parentheses var ( $\hat{\beta}$ ) with the assumption that  $\hat{\rho} = 0$

The Dyer dissimilarity  $Q$  (section 4.4) is an unbiased estimator for half the squared Euclidian distance, that is used as a distance measure in PCA. As the real  $Q$  is approximated rather well by projection of the samples in a few dimensions it is possible to estimate  $Q$  from the graph.

A diatom assemblage that is poor in species and dominated by one or two species will be far from the origin in the vector space of species. A diverse assemblage, with many species in low abundances will be close to the origin. In noncentred PCA the distance of the sample to the origin of the graph is a measure for Simpson's (1949) measure of concen-

tration, *i.e.* the inverse of Hill's (1973) index for  $a = 2$ . Thus  $\Delta_2$  can be read approximately from the graph for each sample. In centred PCA, however, the origin of the graph is not the real origin of the vector space of species, but the centroid (the point representing the mean abundance of each species). The real origin can be calculated and added to the graph.

To examine which species are responsible for the position of the samples in the vector space of species the loadings of the species are plotted simultaneously with the scores of the samples in a 'biplot' (Gabriel 1971). The loadings are scaled to a sum of squares of unity, while the scores are scaled to a sum of squares equal to the corresponding eigenvalues. These scalings correspond to an Euclidian distance biplot, which can be interpreted in terms of species counts, the diversity index  $\Delta_2$ , and the Dyer dissimilarity  $Q$ .

PCA was done according to Orloci (1966), after centering the species, but without any further standardization. Only the 24 species that are present with at least 20 valves in one of the 57 samples (Table 1) were used. According to Whittaker & Gauch (1978) it is a corollary that, for a vegetation fairly rich in species, reduction of the number of species used for ordination only slightly reduces ordination efficiency. In our case this can also be argued theoretically. From section 4.4 it appears that differences are best seen with a dissimilarity measure that is not sensitive for the contribution of rare species, *i.e.* the Dyer dissimilarity  $Q$ . As already noted, the Euclidian distance, used in PCA, has a simple relationship with  $Q$ .

The purpose of our ordination is not only to visualize the data of diatom counts in a convenient graph, but also to correlate principal components with environmental parameters. As species often have a non-linear relationship with environmental parameters a simple Gaussian ordination, based on an eigenvector method and a suitable data transformation (Kooijman 1977) was carried out.

The first and most important principal components of both ordinations are very similar; the other principal components are different. Because the first component has a very significant correlation with acidification, as will be discussed in the next section, only the linear PCA is dealt with here.

The results are given in Fig. 20. Threedimensionally the ordination has about the shape of a

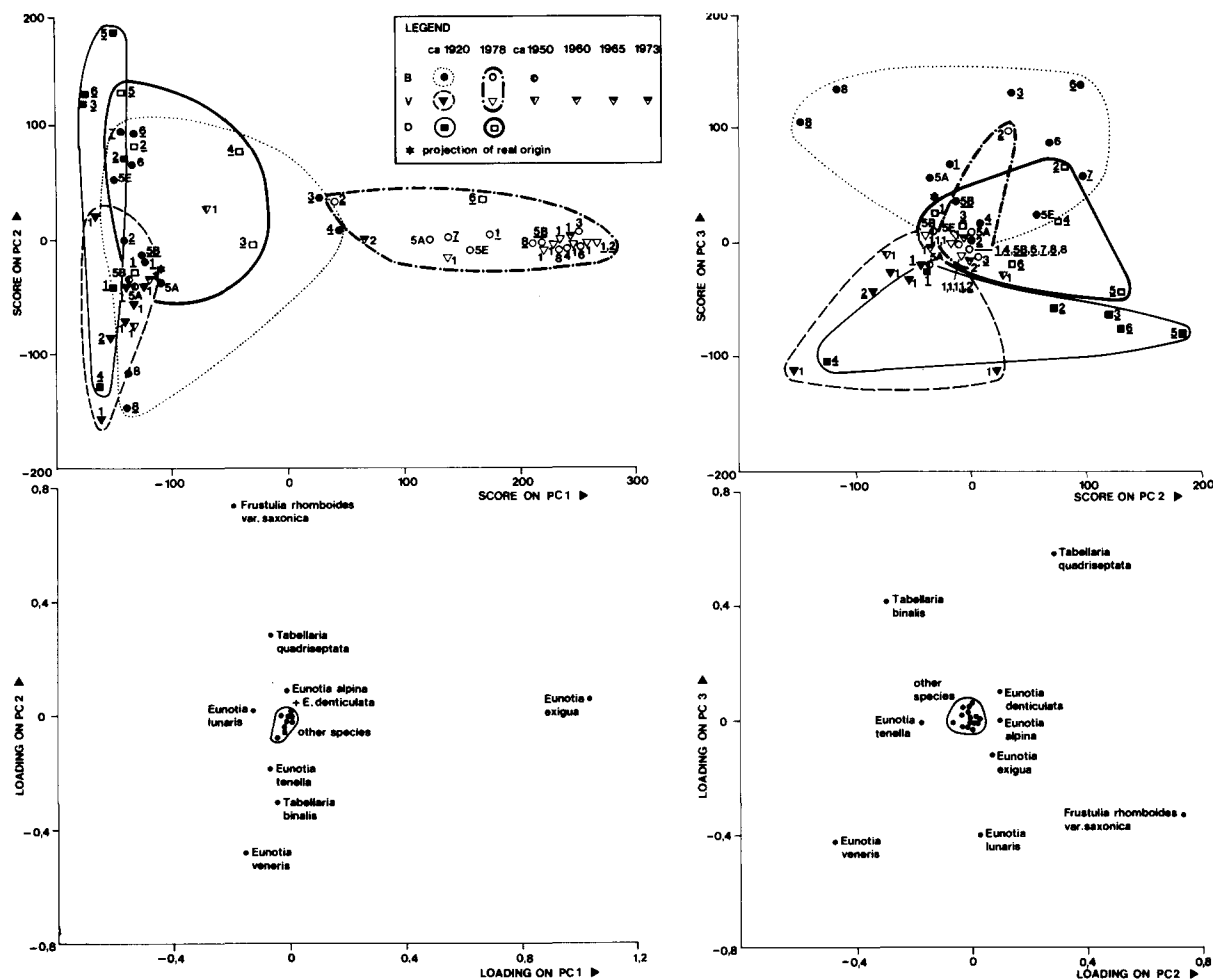


Fig. 20. Biplots of principal component analysis. Samples used for pairwise comparisons are underlined.

toad-stool. In the first two dimensions we look at the stem and the side-projection of the umbrella; in the second and third dimension we look at the bottom of the umbrella, in which the stem is projected as only a small circle. The first principal component (PC 1) represents 61.3% of total variance; PC 2 and PC 3 represent 9.1 and 6.3% of total variance respectively. Thus Fig. 20 explains 76.7% of the total variance. The projection of the real origin is indicated by  $\star$  in the plane of PC 1 and PC 2. To approximate the inverse of  $\Delta_2$  one has to measure the distance of each sample to the point at a distance of 116.2 units from  $\star$  perpendicular above the plane of the paper. The distance of the real origin to the space of PC 1, 2 and 3 is 110.0 units.

PC 1 completely separates the recent (B + V)-samples from the old ones, mainly by *Eunotia exigua*, which is dominant in the modern samples (at least 174 valves in each sample). This cluster includes D6. The old B3- and B4-samples are very close to the recent (B + V)-cluster. All other old B- and V-samples have a low score on PC 1, and thus a low abundance of *E. exigua*. It is easily seen that the species composition differs among these samples, e.g. B8 has a relatively high proportion of *E. veneris* and *Tabellaria binalis*, while B6 and B7 are rich in *T. quadrisepata* and *Frustulia rhomboides* var. *saxonica*. The cluster of old V-samples partly overlaps the old B-cluster in the first two dimensions, but is separated in the third dimension.

The old V-samples are relatively rich in *Eunotia lunaris*, *E. tenella* and *E. veneris*. Old B-samples are differentiated by *Tabellaria binalis* and *T. quadrisepata*.

The recent (B + V)-samples are very close to each other, so that their mean Dyer dissimilarity is low. In contrast, the old (B + V)-samples are more spread out, so that their mean Dyer dissimilarity is high (see also Table 4). Above that there is a considerable shift of the centroid of the cluster with time.

All samples with a high abundance of *Eunotia exigua* are far from the real origin and thus have a low diversity. This is also the case for other samples with dominance of other species, e.g. old B6 with *Tabellaria quadrisepata*, old B8 with *T. binalis* and an old VI-sample with *E. veneris*. Old (B + V)-samples are closer to the origin than recent ones, therefore the former ones are more diverse.

Also recent D-samples have a higher score on PC 1, and thus a higher proportion of *E. exigua*, than old D-samples. There is a shift on PC 2 and PC 3 too, which is caused by a decrease of *E. veneris* and *Frustulia rhomboides* var. *saxonica*. Also *Tabellaria quadrisepata* and *Eunotia tenella* contribute to the move of the D-cluster with time. As the intersample distances within the two D-clusters are about the same, the Dyer dissimilarity does not change with time, although the shift of the centroid of the cluster is significant (see also Table 4). The recent D-samples have another direction to the origin than the old ones, but their distance from the origin does not change (with the exception of D6); neither does diversity.

Furthermore it is interesting to see that the samples of V1 and B5 from about 1950 are very close to the old samples. The V1-sample of 1965 is in the cluster of recent samples, while the V1-sample of 1960 was still close to the old samples. So, major changes apparently occurred after this year. Also note the increase of *Eunotia exigua* in V2 between 1973 and 1978, which will be discussed in section 5.2.

The general conclusions from PCA are that the variation within recent samples, especially in the Veluwe- and Brabant-districts, totally differs from the old variation. While there was a great dissimilarity between the old samples, with generally high diversities, there is only a rather small dissimilarity in the recent samples, with low diversities. The variation that is left, is mainly caused by

differences in the relative abundance of only one single species: *Eunotia exigua*. In the Drenthian samples there is also a shift in species composition, among other things an increase of *E. exigua*, but no clear change in dissimilarity and diversity.

#### 4.6 Chemistry and diatoms

The results of chemical analyses are given in Table 5. The analyses of some old samples from moorland pools and some measurements of precipitation chemistry have been included. This table gives not only the absolute amounts of major ions, but also some ratios. The relative composition of the cations  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $(\text{Na}^+ + \text{K}^+)$  and the anions  $\text{HCO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  is visualized in Fig. 21. This diagram comprises cation- and anion-triangles, which are combined for economy of space.

The old pH-measurements have been made in the field by colorimetry. Almer *et al.* (1974) report that measurements with color-indicator solutions are close to electrometric measurements, the color-indicator values seldom being more than two-tenths of a unit higher. Thus the magnitude of the old measurements is correct. Calculated pH-values from the  $\text{CO}_2\text{--HCO}_3^-$  equilibrium (see e.g. Golterman 1969) are 6.5 and 6.8 for the old samples from B6 and B5B respectively. This is not in agreement with the direct measurements, which are about 4.5 and 5.5 respectively.  $\text{CO}_2$  was measured in the field and  $\text{HCO}_3^-$  was determined after one or more days in the laboratory. pH and the concentration of inorganic carbon compounds may change in this time. Also in the recent samples pH has a tendency to increase, when measurements in the field and the laboratory are compared. Probably the figures for  $\text{HCO}_3^-$  are too high, especially in the old samples.

The chemical parameters have been divided in seven groups, depending on the degree of correlation with the first principal component (PC 1). PC 1 has a very strong positive correlation with the parameters of group 1 in Table 5 and a very strong negative one with the parameters of group 7. The parameters in these groups are very strongly inter-correlated too. PC 1 is strongly correlated with group 2 and 6, well correlated with group 3 and 5 and not correlated with group 4.

The strongest correlations are not found between PC 1 and the absolute concentrations of ions, but

Table 5. Chemical data of moorland pools and precipitation. Recent samples are in decreasing order of score on first principal component (PC1). Physico-chemical parameters are in decreasing order of correlation with PC1.  $r_{PC1}$  = Spearman rank correlation coefficient between chemical data and score on PC1 of recent samples. tr = trace, n.d. = not detectable.

				Recent samples															Old samples <sup>a</sup>					Precipitation <sup>b</sup>				
Station number				V2	B6	B3	B4	V1	B5B	B8	B1	D6	B7	B2	D3	D4	D2	D1	D5	B6	V1	B5B	B	V	D			
Year (19..)				77	78	78	78	78	78	78	78	78	78	78	78	78	78	78	78	78	20	30	19	78	78	78		
Month				11	11	11	11	11	11	11	11	11	11	11	11	11	11	11	11	11	4	4	11					
group	$r^2_{PC1}$	Score on PC1		266	251	250	242	221	218	210	173	168	138	39	-31	-43	-133	-135	-144	-137	-140	-127						
1	0.85	$SO_4^{2-}/(Cl^- + SO_4^{2-} + HCO_3^-)$		0.78	0.64	0.69	0.70	0.64	0.77	0.73	0.77	0.58	0.54	0.44	0.44	0.37	0.23	0.19	0.31	0.23	0.41	0.23	0.68	0.67	0.59			
	0.79	$Ca^{2+}/(Ca^{2+} + Cl^-)$		0.46	0.44	0.39	0.45	0.34	0.53	0.48	0.44	0.25	0.35	0.29	0.19	0.10	0.11	0.25	0.16	0.22	0.29	0.23	0.27	0.34	0.28			
	0.76	$Ca^{2+}/(Ca^{2+} + Mg^{2+} + Na^+ + K^+)$		0.31	0.44	0.32	0.37	0.27	0.41	0.35	0.36	0.20	0.31	0.30	0.18	0.09	0.10	0.25	0.14	-	-	0.32	0.29	0.24				
	0.73	$Ca^{2+}$	meq l <sup>-1</sup>	0.20	0.20	0.17	0.25	0.17	0.55	0.35	0.25	0.12	0.15	0.10	0.15	0.05	0.05	0.15	0.07	0.07	0.20	0.17	0.043	0.040	0.034			
	0.72	$SO_4^{2-}$	meq l <sup>-1</sup>	0.69	0.44	0.62	0.69	0.62	1.64	1.00	1.08	0.54	0.37	0.21	0.48	0.29	0.12	0.12	0.19	0.10	0.40	0.21	0.175	0.158	0.127			
2	0.62	$Mg^{2+}/(Ca^{2+} + Mg^{2+} + Na^+ + K^+)$		0.31	0.24	0.20	0.27	0.29	0.28	0.29	0.23	0.25	0.19	0.21	0.23	0.19	0.22	0.18	0.23	-	-	0.18	0.18	0.17				
	0.61	$Al^{3+}$ <sup>d</sup>	mg l <sup>-1</sup>	-	0.4	0.8	1.0	1.0	5.0	2.0	3.5	0.5	0.2	0.1	1.0	0.4	0.1	0.1	0.1	-	-	-	-	-				
	0.55	EC <sub>25</sub> (field)	mS m <sup>-1</sup>	-	9.5	14.8	12.9	15.4	28.5	19.7	15.2	14.3	7.4	6.3	12.8	10.1	6.5	6.4	11.4	-	-	-	-	-				
	0.53	NH <sub>4</sub> <sup>+</sup> -N-in./ (NH <sub>4</sub> <sup>+</sup> -N-in. + NO <sub>3</sub> <sup>-</sup> -N)		0.88	1.00	1.00	1.00	0.91	0.99	0.75	1.00	0.96	0.71	1.00	0.66	0.98	0.83	0.57	0.78	-	-	-	-	-				
	0.47	EC <sub>25</sub> (lab.)	mS m <sup>-1</sup>	14.3	8.6	11.6	12.7	12.7	27.5	19.3	18.7	12.7	7.9	5.7	13.8	10.2	6.9	7.6	9.8	4.6	9.7	9.7	6.4	5.7	5.3			
3	0.32	$Mg^{2+}$	meq l <sup>-1</sup>	0.17	0.12	0.11	0.18	0.19	0.38	0.29	0.16	0.16	0.09	0.07	0.19	0.10	0.11	0.12	0.13	0.08	0.20	0.16	0.024	0.025	0.025			
	0.01	K <sup>+</sup>	meq l <sup>-1</sup>	0.01	0.02	0.07	0.03	0.03	0.04	0.03	0.03	0.01	0.04	0.01	0.03	0.06	0.03	0.02	0.03	-	-	0.004	0.005	0.005				
	0	NO <sub>3</sub> <sup>-</sup> -N	μg l <sup>-1</sup>	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	<3	n.d.	n.d.	n.d.	-	-				
4	-0.05	NH <sub>4</sub> <sup>+</sup> -N-inorganic	mg l <sup>-1</sup>	0.23	0.04	0.66	0.14	0.70	2.9	0.47	1.17	0.47	0.27	0.23	0.39	3.7	0.70	0.12	0.18	tr.	0.30	tr.	1.7	2.0	1.3			
	-0.11	HCO <sub>3</sub> <sup>-</sup>	meq l <sup>-1</sup>	0	0	0	0	0	0	0	0	0	0.03	0.03	0	0.05	0	0.07	0	0.08	0.10	0.15	0	0	0			
	-0.45	PO <sub>4</sub> <sup>3-</sup> -P (ortho)	μg l <sup>-1</sup>	3	7	7	3	7	7	3	3	20	7	3	52	10	10	7	n.d.	n.d.	n.d.	11	10	7				
	-0.48	KMnO <sub>4</sub> -cons. (filtr.)	mg l <sup>-1</sup>	3	3	4	4	4	8	3	4	1	35	9	2	45	5	7	10	11	13	14	-	-				
	-0.49	HCO <sub>3</sub> <sup>-</sup> /(Cl <sup>-</sup> + SO <sub>4</sub> <sup>2-</sup> + HCO <sub>3</sub> <sup>-</sup> )		0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.04	0.06	0.00	0.06	0.00	0.11	0.00	0.19	0.10	0.17	0.00	0.00	0.00			
5	-0.49	pH (lab.)		3.8	4.3	4.0	3.9	4.1	3.9	3.8	3.7	3.8	4.7	4.6	4.0	5.1	4.4	5.6	4.0	-	-	4.2	4.4	4.4				
	-0.49	Na <sup>+</sup>	meq l <sup>-1</sup>	0.20	0.13	0.20	0.22	0.26	0.37	0.33	0.26	0.35	0.20	0.15	0.46	0.33	0.30	0.33	0.33	-	-	0.063	0.070	0.078				
	-0.50	KMnO <sub>4</sub> -cons. (unfiltr.)	mg l <sup>-1</sup>	12	10	4	7	7	15	9	4	1	70	9	4	75	55	35	25	-	-	-	-	-				
	-0.51	NO <sub>3</sub> <sup>-</sup> -N	mg l <sup>-1</sup>	0.03	0.00	0.00	0.00	0.07	0.02	0.16	0.00	0.02	0.11	0.00	0.20	0.07	0.14	0.09	0.05	n.d.	n.d.	n.d.	0.8	0.9	0.8			
	-0.57	Fe	mg l <sup>-1</sup>	0.21	0.18	0.09	0.06	0.07	0.42	0.14	0.09	0.50	1.4	0.33	0.15	0.35	0.55	0.75	0.22	<0.1	0.15	<0.1	-	-				
6	-0.58	pH (field)		3.7	4.2	3.9	3.8	3.8	3.7	3.6	4.0	3.7	4.6	4.6	3.8	4.5	4.5	4.6	4.3	~4.5	~5.5	-	-	-				
	-0.62	Cl <sup>-</sup>	meq l <sup>-1</sup>	0.20	0.25	0.28	0.30	0.35	0.49	0.38	0.32	0.39	0.28	0.24	0.62	0.44	0.41	0.45	0.42	0.25	0.47	0.54	0.082	0.079	0.087			
	-0.64	NH <sub>4</sub> <sup>+</sup> -N-organic	mg l <sup>-1</sup>	0.31	0.02	0.14	0.16	0.14	0.14	0.27	0.16	0.27	0.54	0.16	0.16	0.74	0.58	0.43	0.31	0.28	0.18	0.22	-	-				
	-0.77	EC <sub>25</sub> (Cl <sup>-</sup> )/EC <sub>25</sub> (field)		-	0.20	0.15	0.18	0.17	0.13	0.15	0.16	0.21	0.29	0.29	0.37	0.33	0.48	0.54	0.28	-	-	-	-	-				
	-0.82	EC <sub>25</sub> (Cl <sup>-</sup> )/EC <sub>25</sub> (lab.)		0.11	0.23	0.19	0.18	0.21	0.14	0.15	0.13	0.24	0.27	0.32	0.34	0.33	0.45	0.45	0.33	0.42	0.37	0.42	0.10	0.11	0.13			
7	-0.82	Cl <sup>-</sup> /(Cl <sup>-</sup> + SO <sub>4</sub> <sup>2-</sup> + HCO <sub>3</sub> <sup>-</sup> )		0.23	0.36	0.31	0.30	0.36	0.23	0.28	0.23	0.42	0.41	0.50	0.56	0.56	0.77	0.70	0.69	0.58	0.49	0.60	0.39	0.33	0.41			
	-0.82	(Na <sup>+</sup> + K <sup>+</sup> )/(Ca <sup>2+</sup> + Mg <sup>2+</sup> + Na <sup>+</sup> + K <sup>+</sup> )		0.38	0.33	0.48	0.37	0.44	0.31	0.36	0.41	0.55	0.50	0.49	0.59	0.67	0.67	0.57	0.63	-	-	0.50	0.54	0.59				
	-0.83	(Na <sup>+</sup> + K <sup>+</sup> )/(Ca <sup>2+</sup> + Mg <sup>2+</sup> )		0.62	0.48	0.93	0.58	0.78	0.44	0.56	0.71	1.24	1.00	0.94	1.44	2.60	2.06	1.46	1.71	-	-	1.01	1.16	1.42				
	-	CO2	mmol	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0.07	0.11	0.05	-	-				

<sup>a</sup> Borrowed from Redeke & de Vos (1932), Redeke (1948) and J. Heimans (pers. comm.)

<sup>b</sup> Yearly averages of the stations Gilze-Rijen and Eindhoven (B), Deelen (V) and Witteveen (D) taken from Anonymous (1979). The figures for B are mean values of Gilze-Rijen and Eindhoven

<sup>c</sup> Only recent samples have been used for calculation of  $r_{PC1}$  (no correction for ties). Significance levels:  $r = 0.51$ :  $P = 0.05$ ;  $r = 0.67$ :  $P = 0.01$ ;  $r = 0.85$ :  $P = 0.001$ .

<sup>d</sup> All Al-species assumed to be present as Al<sup>3+</sup>

between PC I and some ratios.  $(\text{Na}^+ + \text{K}^+)/(\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^+ + \text{K}^+)$ ,  $\text{Ca}^{2+}/(\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^+ + \text{K}^+)$  and  $\text{Mg}^{2+}/(\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^+ + \text{K}^+)$  are the relative concentrations of  $(\text{Na}^+ + \text{K}^+)$ ,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  referred to their sum (= sum of major cations) and define the points in Fig. 21. The ratio  $\text{Ca}^{2+}/(\text{Ca}^{2+} + \text{Cl}^-)$  can be used as an easy-to-measure surrogate for the relative  $\text{Ca}^{2+}$ -concentration, because the proportion of  $\text{Mg}^{2+}$  is rather constant in surface and ground waters, the proportion of  $\text{K}^+$  is fairly small and  $\text{Na}^+$  is highly correlated with  $\text{Cl}^-$  (Van Wirdum 1980).  $(\text{Na}^+ + \text{K}^+)/(\text{Ca}^{2+} + \text{Mg}^{2+})$  is the ratio of univalent to divalent cations, which was recognized to be an important factor for the spatial distribution of algae by Pearsall (1921). It is closely related to the relative  $(\text{Na}^+ + \text{K}^+)$ -concentration. In the same way the relative concentrations of the anions  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$  and  $\text{HCO}_3^-$  are defined. The ratio of the calculated electrical conductivity of the  $\text{Cl}^-$ -ions present in the sample ( $\text{EC}_{25}(\text{Cl}^-)$ ) and the electrical conductivity of the sample ( $\text{EC}_{25}$ ) is an easy-to-measure surrogate for the relative  $\text{Cl}^-$ -concentration, as the electrical conductivity is correlated very strongly with  $(\text{Cl}^- + \text{SO}_4^{2-} + \text{HCO}_3^-)$  (Van Wirdum 1980). Because  $\text{HCO}_3^-$  is of minor importance or absent at all in the moorland pools, this ratio may be used as a surrogate for the relative  $\text{SO}_4^{2-}$ -concentration too.

For a first inspection of the data Spearman rank correlation coefficients have been used, because we had no *a priori* model of the relationships between environmental and species composition parameters. Product moment correlation coefficients before and after logarithmic transformation of chemical data have been calculated too. Then the most salient relationships are those between PC I and  $\log [\text{SO}_4^{2-}]$  ( $r^2 = 0.69$ ;  $P < 0.001$ ) and the relative concentrations of  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  (both  $r^2 = 0.90$ ;  $P < 0.001$ ). The first two relations are visualized in Fig. 22 and 23.

Other positive correlations are between PC I and the relative  $\text{Mg}^{2+}$ -concentration,  $\text{Al}^{3+}$ ,  $\text{EC}_{25}$ , and somewhat weaker between PC I and organic  $\text{NH}_4^+\text{-N}$ ,  $\text{Cl}^-$ , pH, Fe,  $\text{NO}_3^-\text{-N}$ ,  $\text{Na}^+$ ,  $\text{KMnO}_4$ -consumption,  $\text{PO}_4^{3-}\text{-P}$  and the relative  $\text{HCO}_3^-$ -concentration.

$\text{Al}^{3+}$ ,  $\text{Ca}^{2+}$  and to a lesser extent  $\text{Mg}^{2+}$  often have relative high concentrations in acidified waters, because these ions are easily leached from

the soil in watersheds that are exposed to acid precipitation (Dickson 1975, 1978; Likens & Bormann 1975; Gjessing *et al.* 1976; Wright & Gjessing 1976; Almer *et al.* 1978; Cronan & Schofield 1979). The increase in concentration of these ions and of  $\text{SO}_4^{2-}$  and  $\text{H}^+$  causes an increase of electrical conductivity. Thus PC I may be interpreted as an acidification component. Acidity also restrains the activity of nitrifying bacteria (Hutchinson 1957; Wetzel 1975), which may be the cause of the good correlation between PC I and the ratio inorganic  $\text{NH}_4^+\text{-N}/(\text{inorganic } \text{NH}_4^+\text{-N} + \text{NO}_3^-\text{-N})$ .

Dystrophic and bog waters have a high  $\text{KMnO}_4$ -consumption and are often rich in iron, which forms complexes with humic substances (Wetzel 1975; Gjessing 1976). These waters may be also moderately rich in  $\text{PO}_4^{3-}\text{-P}$  (Wetzel 1975; Prakash *et al.* 1975), which is correlated negatively with PC I, like  $\text{KMnO}_4$ -consumption and Fe. Organic  $\text{NH}_4^+\text{-N}$  and  $\text{NO}_3^-\text{-N}$  are correlated negatively too with PC I. Wetzel (1975) states the N-level to be 'moderate' for dystrophic lakes and 'low' for clear water oligotrophic lakes. Bog lakes are fed by rain water, which is relatively rich in  $\text{Na}^+$  and  $\text{Cl}^-$  and poor in  $\text{Ca}^{2+}$ . Moreover, peatmosses exchange hydrogen ions more strongly with divalent cations than with univalent ones (Anschütz & Gessner 1954; Clymo 1967). Therefore the relative concentration of  $(\text{Na}^+ + \text{K}^+)$  and the absolute concentration of  $\text{Na}^+$  is high in dystrophic and bog waters (Wetzel 1975).  $\text{Cl}^-$ , which is like  $\text{Na}^+$  of marine origin in precipitation, is often closely associated with  $\text{Na}^+$  in bog waters (Tolpa & Gorham 1961). Therefore the absolute and relative  $\text{Cl}^-$ -concentrations are also negatively correlated with PC I. All variables which have a negative correlation with PC I are connected with ombrogenic, dystrophic waters. High humus content apparently counteracts man-made acidification.

Thus acidification causes a shift from ombrogenic to soligenic environment, which can be easily seen from the cation triangle in Fig. 21. Ombrogenic (often dystrophic) moorland pools of the Drenthian district are in the right corner, soligenic (mostly clear water) pools are in the centre of Fig. 21. Ombrogenic pools have a high proportion of chloride, soligenic pools have a high proportion of sulfate, which can also easily be seen in Fig. 21. In this Figure and in Fig. 22 and 23 also the data of

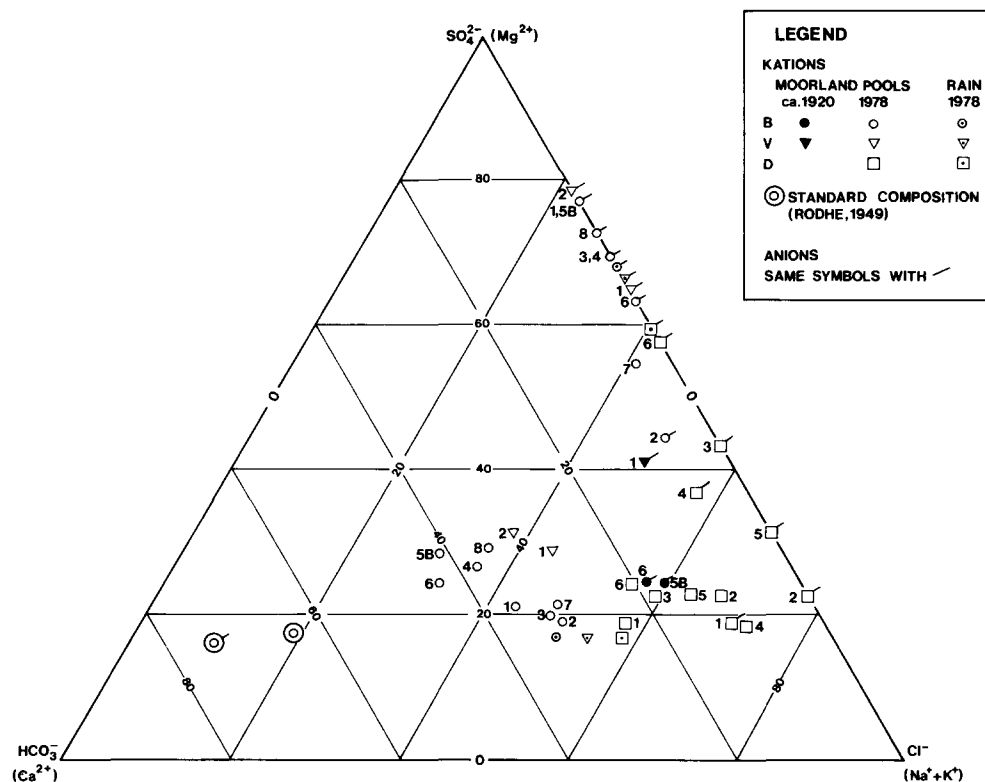


Fig. 21. Combined anion-cation triangular diagram.

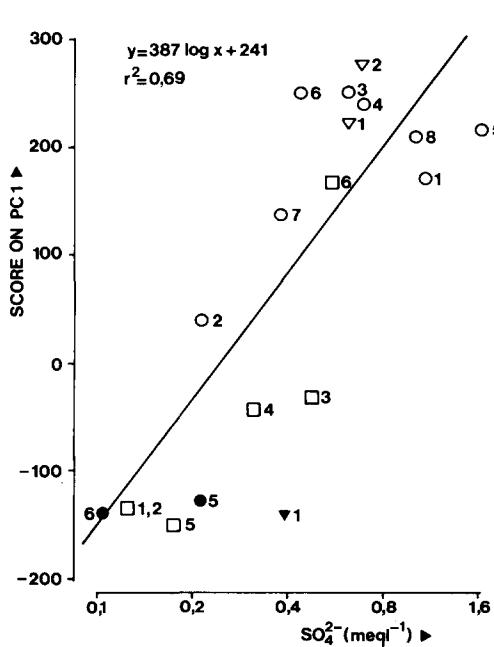


Fig. 22. Semi-logarithmic plot of absolute sulfate concentration and score on first principal component. Symbols as in Fig. 20.

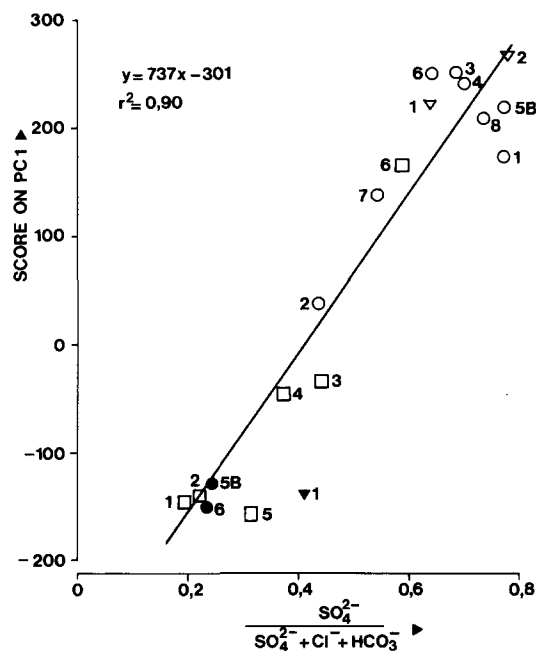


Fig. 23. Plot of relative sulfate concentration and score on first principal component. Symbols as in Fig. 20.

three old samples are plotted. The chemical and diatom samples of pools B5B and B6 were taken in about the same year. The chemical sample of V1 was taken twelve years after the diatom sample. The relative and absolute sulfate concentrations increased considerably between about 1920 and 1978, as well as the score on PC 1. The absolute chloride concentration is about the same in 1978 as in the old samples, which means that the changes in chemistry very probably are caused by increase of the sulfate concentration.

*Eunotia exigua* has a high loading on PC 1 (Fig. 20) and therefore has a high rank correlation ( $r = 1$ ) with this component. The species has nearly the same correlations with chemical parameters as PC 1 and is an excellent indicator species for acidification. Especially *Frustulia rhomboides* var. *saxonica* and *E. lunaris* have high negative loadings on PC 1 and are therefore highly negatively rank correlated with this component ( $r = -0.88$  and  $0.79$  respectively). Good development of these species is indicative for non-acidified moorland pools.

The most significant rank correlation ( $r = 0.84$ ) between diversity indices and chemical parameters is between  $\Delta_2$  and  $EC_{25}(Cl^-)/EC_{25}(\text{field})$ . The rank correlation coefficient between  $\Delta_2$  and the relative sulfate concentration is  $0.74$ . As may be expected from the projection of the origin in the PCA, the correlation between  $\Delta_2$  and PC 1 is also high ( $r = -0.88$ ).  $\Delta_2$  and the other indices of the Hill-family decrease with increasing acidity.

PC 2 and 3 are less easy to interpret than PC 1, because chemical analyses are, with a few exceptions, only available for the recent samples which have relatively low scores on PC 2 and 3. Presumably the variation in the old samples, which have high scores on these components, is connected with small differences in chemistry, caused by regional factors (soil, hydrology) and the presence of humic substances.

The absolute and relative concentrations of rain water constituents is given in Table 5 and Fig. 21. It appears that the absolute and relative ( $Na^+ + K^+$ )-concentrations are increasing in the order B, V, D, which corresponds with this sequence in the water of the moorland pools. The increase of the relative ( $Na^+ + K^+$ )-concentration in the Drenthian pools and the decrease of this concentration in the Brabantine and Veluvian pools can be nicely seen in Fig. 21. The absolute concentration of rain water

sulfate increases again in the order B, V, D; the relative concentration is highest in V and equal in B and D.

The chemistry of precipitation determines the chemical composition of moorland pool water to a considerable extent, because the sandy subsoil is extremely poor in nutrients and other soluble minerals. Also dry deposition of gases contributes to the ionic content of the water. Assuming an approximate deposition velocity of  $1.1 \text{ cm s}^{-1}$  for gaseous  $SO_2$  on fresh water (McMahon & Denison 1979) and using the data of the national air pollution network (Anonymous 1978) the dry deposition of  $SO_2$ -S is about twice as much as the wet deposition of  $SO_4^{2-}$ -S at the nearest gauges to the moorland pools in the three areas. This can only be a very rough estimate, because the deposition velocities of  $SO_2$  as given by various authors differ from  $0.4$  to  $2.2 \text{ cm s}^{-1}$  (McMahon & Denison 1979). Nevertheless the calculated ratio of wet and dry deposition is in accordance with the value that is given by Fowler (1980) for agricultural areas  $30$ – $300 \text{ km}$  from sources. The dry deposition of particulate (aerosol) S is only a few percents of gaseous deposition (Husar *et al.* 1978).

## 5. Discussion

### 5.1 Comparison with other diatom studies

The acidification of humic poor moorland pools causes a reduction of diversity ( $\Delta_a$  for  $a \geq 0.25$  (section 4.3)). This agrees with the hypothesis set in the introduction and the results of Berge (1976). He found a reduction of the Shannon-index, which is  $\ln \Delta_1$ , in diatom assemblages of some Norwegian rivers between 1949 and 1975.

By acidification the Dyer dissimilarity decreases but this is not the case with the number of species in common (section 4.4). This corresponds with the hypothesis set in our introduction and with the results of Berge (1976, 1979). This author emphasizes that acidification primarily causes quantitative changes in the diatom assemblages, while the floristic composition remains rather constant.

Acidification of humic poor pools gives rise to a huge increase of the proportion of *Eunotia exigua*, at the cost of *E. veneris*, *Fragilaria virescens*,



*Frustulia rhomboides* var. *saxonica*, *Navicula subtilissima*, *N. heimansii*, *Surirella arctissima*, *Tabellaria binalis*, *T. flocculosa* and *T. quadri-septata*.

No changes of diversity and dissimilarity can be detected in the humic pools in Drenthe. However, the species composition changes: *Eunotia exigua* increases at the cost of *E. veneris*, *Pinnularia appendiculata* and - to a lesser extent - *Frustulia rhomboides* var. *saxonica*. Berge (1979) found no clear changes in the diatom assemblages in a core of Högkleivvatn, a humic acid lake in southern Norway; which may indicate the buffering action of humic substances.

Berge (1976) finds a clear rise of *Eunotia exigua* with increasing acidity of rivers between 1949 and 1975, but the relative abundance does not exceed 18.5% (equivalent to 74 valves). The pH of the rivers where this species was abundant is between 4.8 and 6.1. He finds a decline of *Frustulia rhomboides*, which is ecologically very similar to its variety *saxonica*, between 1949 and 1975. According to Berge (1979) acidification promotes the growth of *Tabellaria binalis* and probably *Eunotia denticulata*, as recorded in the sediment of some Norwegian lakes. He found no clear relationship between the acidity of the lakes (recent pH 4.7–5.7) and the abundance of *E. exigua*.

Miller (1973) found an increase of *E. veneris* and *Tabellaria binalis* in the recent sediments of Stora Skarsjö, an acidifying clear water lake near the Swedish west coast. She found no significant change in the relative abundances of *Eunotia exigua* and *Frustulia rhomboides* (including varieties). The pH of this lake was 6.3 in 1947 and 4.5 in 1971.

The most striking difference between the Scandinavian and the Dutch situation is the difference in the behaviour of *Tabellaria binalis*, that increases in the northern countries, but decreases in the Netherlands. This may be connected with the pH which is now 3.7–3.8 in those moorland pools where *T. binalis* was abundant about 1920. At the above cited Scandinavian localities the pH is now between 4.5 and 5.7. Furthermore the northern localities have generally a lower electrical conductivity ( $<8 \text{ mS m}^{-1}$ ) than the Dutch ones ( $>6 \text{ mS m}^{-1}$ ). According to the literature the optimal development of this species is found in waters with pH 4.0–6.8 and low conductivity. One may speculate

that *T. binalis* will disappear from Scandinavia when acidification continues.

Perhaps the same reasoning can be applied to *Eunotia denticulata* and *E. veneris*. *Frustulia rhomboides* has the same behaviour in Norway as its variety *saxonica* in the Netherlands. Also the less dominant role of *Eunotia exigua* in the northern countries may probably be ascribed to the less acidified and mineralized waters in Scandinavia, as compared with the Dutch situation.

## 5.2 Sulfur metabolism

As the recent sulfate concentration varies considerably from pool to pool (Table 5) this concentration can be changed by processes occurring in the moorland pools or their watershed. A model for the prediction of the sulfate concentration from dry and wet sulfur deposition requires quantitative information about hydrological, physico-chemical and biological processes in the moorland pools and their watersheds. As this information is not available, sulfur metabolism can be discussed only in general terms. In addition to acid precipitation also afforestation and drought may cause acidification and they are included in this discussion.

Differences in the water balance of the pools will be reflected in different concentrations of major ions, e.g. chloride, which is biologically and hydrologically conservative. Differences in retention time and rate of evaporation are reflected by the chloride concentration (Table 5). The ratio  $\text{SO}_4^{2-}/\text{Cl}^-$  is more suited to trace the changes of the sulfate concentration by biological and chemical processes than the absolute sulfate concentration. This ratio is closely related to the relative sulfate concentration (RSC), as the bicarbonate concentration is negligibly small (Table 5). When the RSC in a pool is equal to the RSC of incident rain this does not mean that sulfate production and consumption in this pool are in equilibrium, because the dry deposition of sulfur is about twice the wet deposition (section 4.6).

RSC is lowest in the Drenthian pools (except D6), which are generally rich in humus. Baas Becking & Nicolai (1934) investigated a small dystrophic pool, close and similar to D5. They measured very high concentrations of sulfuretted hydrogen and pointed out the importance of sulfate reduction in this type of moorland pools. Hongve

(1978) obtained strong experimental evidence for sulfate reduction taking place in sediments of dystrophic lakes in Norway.

Schindler *et al.* (1980) measured a strong increase of sulfate reduction in the hypolimnion of Experimental Lake 223 in a whole lake acidification experiment. Thus the mineralization of organic matter is intensified by the activity of sulfate reducing bacteria. Possibly the released nitrogen and phosphorus compounds will eutrophy the moorland pools. (The mechanism of eutrophication by sulfate addition to alkaline waters has been described by Ohle (1954). In this case the iron from ferriphosphate precipitates as ferrosulfide, because ferrosulfide is insoluble in neutral and alkaline waters, while phosphate is released. In contrast, ferrosulfide is soluble in acid bog waters (Baas Becking *et al.* 1960).)

No old chemical samples are available to test this hypothesis of eutrophication in brown water pools. The increase of the diatom *Eunotia exigua* might be interpreted as an increase of the trophic state. Coesel & Smit (1977) investigated the recent desmid assemblages in the Drenthian pools (except D6) and compared their results with the inventories of Beijerinck (1926) and Wartena (1954). The trophic ranges of desmids are fairly well known (Coesel, 1975). It appears that the number of mesotraphentic desmid species gradually increased during the past sixty years in D1, D2, D4 and D5. The rise is most spectacular in D5, that was the most oligotrophic moorland pool in this district sixty years ago. Typical oligo-mesotraphentic desmids disappeared from D3. This pool seems to be more oligotrophic than about 1920. The data in Table 5 suggest that the pools with an increased trophic state are fairly rich in humus (high permanganate consumption and high levels of iron, organic nitrogen and phosphorus), while D3 is relatively poor in humus and rich in sulfate. These observations support the hypothesis of eutrophication by sulfate reduction in dystrophic moorland pools.

B7, the only dystrophic pool among the Brabantine localities, has relatively low sulfate and high phosphate levels, as compared with the other pools in this district. The diatom assemblage is less affected by acidification than in most Brabantine pools (Fig. 20). Nevertheless the desmid flora of B7 greatly impoverished between 1920 and 1975 (Coesel *et al.* 1978). In Brabant the air is more

severely polluted than in Drenthe, that is rather remote from urban and industrial areas. This may indicate that the buffering capacity of humic pools for acidification ends when the sulfur burden exceeds a certain level.

In some pools, notably V2, B5B and B1, the RSC is rather high. This may be caused by processes which accumulate sulfate. Many authors (*e.g.* Lillieroth 1950; Malmer 1961; Ohle 1965) emphasize that lowering of the water table by drought or drainage promotes the mineralization of organic sulfur compounds, by which sulfate is released. The year 1976 was extremely dry and very low water levels were observed in the moorland pools. Large parts of the bottom were left dry. Also 1975 was rather dry. After 1976 the water level gradually rose again.

This process is reflected by the RSC in V1. In April 1974 the RSC was 0.66; in November of the years 1977, 1978 and 1979 RSC was respectively 0.73, 0.64 and 0.56. In V2 the RSC was 0.70 in July 1973 and 0.78 in November 1977. In B5B these values were for November 1978 and 1979 respectively 0.77 and 0.68 (data for 1973 and 1974 from Notenboom-Ram, 1976; for 1977 and 1979 Van Dam, unpublished). The high RSC levels in Table 5 are at least partially caused by the dry summer of 1976. As a consequence the abundance of *Eunotia exigua* increased between 1973 and 1977 in V2 (Fig. 20) and between 1975 and 1978 in B5 (compare with pool A1 in Van Dam & Kooyman-van Blokland 1978).

The effect of drought may be compared with that of snow melt. The snow pack accumulates sulfur that is released in the spring. In Scandinavian rivers sometimes fish killings are observed in this time of the year (Almer *et al.* 1978). In dry summers the reduced sulfur, that has been accumulated in the sediments, is released as sulfate.

Also the plantations of Scots pine around many Brabantine moorland pools may be responsible for the high RSC in this area. The heathlands and inland sand dunes were reafforested between 1840 and 1920. Afforestation enhances evapotranspiration (Noirfalise 1967) and consequently the concentration of ions in the soil solution and in ground water. Conifers are known to acidify soil and soil water because the litter is relatively poor in cations and exchanges hydrogen ions for cations (Malmer 1974; Odén 1976). Also the uptake of ammonium

nitrogen contributes to the 'biological acidification' by pine forests (Odén 1976). An increase of ammonium in rain water by air pollution will increase biological acidification.

In conifer stands throughfall is enriched with sulfate in comparison with incident rain. This is caused by leaching of sulfur from foliage and by dry deposition of gaseous and particulate sulfur species (e.g. Nihlgård 1970; Mayer 1974; Abrahamsen *et al.* 1977; Heinrichs & Mayer 1977; Mayer & Ulrich 1978). The dry deposition of sulfur species increases in the order bare soil, deciduous forest, coniferous forest. The filtering action of pines causes an increase of sulfate concentration in throughfall, stemflow and seepage water below the root zone.

The increased acidity promotes the leaching of metals; especially calcium and aluminium are found in high concentrations in seepage water. According to Malmer (1974) there seem to be no close connections between the acidification of lakes and rivers on the one hand and the acidification or any other changes in the soils of their watersheds on the other hand. Nevertheless Likens & Borman (1975) suppose that increased leaching of soil by acid precipitation will enrich lakes and rivers in the long run. As stated in section 4.6 enrichment with aluminium, calcium and magnesium has been found in Scandinavian and North-American waters that are affected by acid rain.

Linzon (1978) and Laaksovirta & Olkkonen (1979) found an increase of sulfur in pine needles with an increase of  $\text{SO}_2$  in ambient air. Thus, when air pollution increases the sulfur content of the foliage, which directly falls in the moorland pools, will increase too.

Apart from V2 which is not taken into account, as this moorland pool was sampled in 1977, when the drought of 1976 had still a larger impact than in 1978, the highest RSC's are found in B1, B5B and B8. These pools have contact with the aquifer and are surrounded by more or less dense stands of Scots pine, especially B5B. B3 and B4 are surrounded by pines too, but these pools are not aquifer fed and have a lower RSC than the previous pools. The relatively low RSC for B2 is difficult to explain, because this moorland pool is situated in a depression with pines. This pool is supposed to be aquifer fed.

### 5.3 Metals

We found a very good correlation between the absolute and relative sulfate concentration on the one hand and the composition of the diatom assemblages on the other hand. Also other chemical parameters are highly correlated with PC 1 (Table 4). So it is very difficult to assess which factor is responsible for the spatial and temporal differences between the diatom assemblages.

It can be argued that sulfate governs the chemistry of the moorland pools. In the pools where old chemical samples are available the absolute chloride concentration remained rather constant in the last 60 years, whereas the absolute sulfate concentration increased considerably. Also other ions increased between 1920 and 1978, e.g. calcium, but this may be a consequence of enhanced cation leaching by rise of the acidity, caused by sulfate.

Schroeder (1939) found that sulfate below  $4.2 \text{ meq l}^{-1}$  is practically without influence on the distribution of diatoms. Stokes & Hutchinson (1975) tested the growing ability for the green algae *Chlorella vulgaris* and *Scenedesmus quadricauda* for sulfate concentrations in the range 0–5.2  $\text{meq l}^{-1}$  in a bioassay. Growth of these algae was independent of sulfate concentration. This supports the hypothesis that the change of the diatom assemblages is not a direct consequence of increased sulfate levels.

Aluminium and heavy metals (Cd, Cu, Hg, Mn, Pb, Zn) are often found in high concentrations in acidified waters, not only by increased fallout in polluted areas, but also because their solubility increases with acidity (Dickson 1975; Stokes & Hutchinson 1975; Beamish & Van Loon 1977; Almer *et al.* 1978; Henriksen & Wright 1978). The maximal content of  $\text{Al}^{3+}$  reported from acidified lakes in Scandinavia is  $0.68 \text{ mg l}^{-1}$  at pH 4.0 (Almer *et al.* 1978). In B5B  $5.0 \text{ mg l}^{-1}$  at pH 3.7 was measured. According to Almer *et al.* (1978) a concentration of ca  $0.1 \text{ mg l}^{-1}$  is toxic to fishes. (Fishes do not naturally occur in moorland pools. In those pools where fishes have been introduced in the past, fish killings are observed, especially in the last five or ten years).

No systematic research on the impact of heavy metal pollution on many species of diatoms has been carried out, but it appears from some case studies that diatoms are sensitive to heavy metals

(Besch *et al.* 1972; Patrick 1977). *Eunotia exigua*, which takes most advantage from the acidification, is reported in literature to be very resistant to pollution with acids, aluminium and heavy metals and is often found in acid mine drainage. Probably the elevated levels of aluminium and heavy metals, caused by increased sulfate concentrations are more important for the distribution of diatoms than sulfate *per se*.

#### 5.4 Oligotrophication or eutrophication?

Hydrogen ion concentration controls the solubility of aluminium, and consequently that of phosphate. Phosphates have a minimal solubility at pH 5–6. The solubility increases when pH becomes less than five or more than six (Stumm & Morgan 1970; Dickson 1978; Almer *et al.* 1978). Thus if pH drops from 4.5–5.0 to below 4.0, as is the case in many clear water moorland pools, more phosphorus is expected to become available for primary production.

Almer *et al.* (1978) showed that the relationship between biomass of phytoplankton and pH in 58 Swedish lakes is similar to the relationship of experimentally determined phosphorus solubility and pH, although they could not distinguish in phosphorus levels in the field, because of the overall low concentrations (less than  $10 \mu\text{g l}^{-1}$ ), just as in our clear water pools. According to Almer *et al.* (1978) it is not unlikely that the most acid lakes were actually richer in phosphorus and therefore had the largest biomasses.

Grahn *et al.* (1974), Grahn (1977) and Coesel *et al.* (1978) observed a strong increase of the biomass of *Mougeotia*, other filamentous algae and macrophytes, mainly *Sphagnum*, in lakes and moorland pools, which are exposed to acid precipitation. Because of the ion exchange capacity of peat-mosses this is interpreted by these authors as oligotrophication. On the other hand one may consider the mere increase of the biomass of the macrophytes as eutrophication.

No old and recent data about primary production of the moorland pools are available. Nevertheless we can get some idea about the change of the trophic state by using the species as trophic indicators, as has been done already in section 5.2. It appears from section 4.1 that especially eutraphentic diatom species increased from about 1920

to 1978, while oligotraphentic species decreased. No luxuriant growth of the former category can be observed, because most eutraphentic diatoms prefer circumneutral or alkaline waters. However, the acidobiontic *Eunotia exigua* can thrive in both oligotrophic and eutrophic environments.

It is a matter of experience that the adverse effects of human impact in wetland ecosystems are best observed in dry years. During dry summers in the beginning of this century elements of the alliance Nanocyperion, especially the association Cicendietum filiformis (*e.g.* *Cicendia filiformis*, *Juncus mutabilis*, *Peplis portula*, *Echinodorus ranunculoides*) occurred abundantly on the shores of pristine moorland pools (Beijerinck 1929; Goffart & Sternon 1936). In contrast, in the very dry summer of 1976 seedlings of species of the Bidention and related alliances (*e.g.* *Polygonum hydropiper*, *Lycopus europaeus*, *Eupatorium cannabinum*) were observed frequently (Van Dam & Kooyman-van Blokland 1978). The latter alliance requires more nutrients than the first one. During 1976 and after this year the amphiphyte *Juncus bulbosus* showed an explosive expansion in many oligotrophic moorland pools (see also Sykora, 1979). It is generally found in acid, oligotrophic waters, but it has a wide ecological amplitude as it may occur abundantly even in brackish and acid mine water (Schoof-van Pelt 1973; Sand-Jensen & Rasmussen 1978). So the autecology of *J. bulbosus* and *Eunotia exigua* is remarkably similar. *Juncus effusus*, a typical indicator of disturbance (Reichelt 1964), was still absent from the investigated moorland pools a few decennia ago, but it now forms a massive belt along the shore of some pools, notably B1 and VI. The species greatly increased after 1976 (see also Sykora 1979).

We agree with Almer *et al.* (1978) that acidification may not be interpreted as oligotrophication. They use the term 'acidotrophication'. Perhaps it is grammatically more correct to speak about 'aciditrophication', analogous to acidification.

#### Summary

In November 1978 ten moorland pools in the province of Brabant (B), the Veluwe area (V) and six pools in the province of Drenthe (D) were sampled for chemistry and diatoms. Slides were

prepared from this material and from samples which were taken about 1920 at the same stations. 194 taxa were listed, of which 24 are more or less common. One species, *Navicula heimansii*, is new for science.

The wide range of pH between (B + V)-pools in 1920 (estimated from diatom spectra 4–6) is very narrow in 1978 (measured 3.7–4.6). Most (B + V)-pools are clear water lakes. No significant decline of the pH (ca 4.5) in most D-pools is observed, although some of them are very acid now (pH ca 3.7). Most D-pools are brown water lakes.

The diversity indices  $\Delta_a$  (Hill 1973) decline significantly in the (B + V)-pools for  $a \geq 0$ .  $\Delta_a$  has a tendency to increase with time in the D-pools, although this change is not significant. The number of species which two pools have in common does not change with time. The Dyer (1978) dissimilarity shows a significant decrease in the (B + V)-pools; no changes are found in the D-pools.

The first three components of the principal component analysis (PCA) explain 61, 9 and 6% of the total variance respectively. Old samples have lower scores on the first principal component (PC 1) than recent ones. Recent samples have lower scores on the second and third component than old ones. PC 1 is correlated well with  $\log [\text{SO}_4^{2-}]$  ( $r^2 = 0.69$ ) and even better with the relative sulfate concentration ( $r^2 = 0.90$ ). Thus, the original variation, caused by regional factors is substituted by a sulfate controlled variation.

PC 1 is also positively correlated with a number of other factors ( $\text{Ca}^{2+}$ ,  $\text{Al}^{3+}$ ,  $\text{Mg}^{2+}$  and electrical conductivity, of which is known that they increase by acid precipitation, and negatively with parameters which are characteristic for dystrophic bog waters (organic  $\text{NH}_4^+\text{-N}$ ,  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{Fe}$ ,  $\text{PO}_4^{3-}\text{-P}$ , ( $\text{Na}^+ + \text{K}^+$ ) ( $\text{Ca}^{2+} + \text{Mg}^{2+}$ ),  $\text{KMnO}_4$ -consumption).

PC 1 is nearly completely determined by *Eunotia exigua*, which is known to inhabit natural acid waters as well as acid mine waters. The species is very resistant to pollution by aluminium and heavy metals. Besides *E. exigua* some eutraphentic species increase by acidification. These are present in small quantities, because the low pH is an unfavourable environment for these generally circumneutral and alkaliphilous species. Typical oligotraphentic species (e.g. *Frustulia rhomboides* var. *saxonica*, *Tabellaria binalis*, *Eunotia denticulata*) decline with increasing acidity.

The range of sulfate concentration between the moorland pools (0.12–1.64 meq  $\text{l}^{-1}$ ) is larger than would be expected from the sulfur deposition pattern in the Netherlands. In humic acid pools the sulfate concentration is low by sulfate reduction. Highest sulfate concentrations are found in moorland pools in depressions, that have been afforested with Scots pines. The high dry deposition of sulfur compounds on conifers adds to the sulfate concentration of seepage to these pools.

Another factor which may explain partially the high sulfate concentrations in 1978 is the severe drought of 1976. By drought and drainage organic sulfur species are mineralized and sulfate is released.

The oligotrophication - eutrophication controversy is discussed. We agree with Almer *et al.* (1978) that acidification does not cause oligotrophication and that aciditrophication is a more correct term to describe the process.

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## Appendix

List of all species found with pH-indication class (see text p. 431)  
M = alpine, boreo-alpine, boreal or mountainous distribution within Europe.

acf	<i>Achnanthes altaica</i> (Poretzky) A. Cleve-Euler
alb	<i>A. brevipes</i> Agardh
cir	<i>A. delicatula</i> (Kützing) Grunow
alf M	<i>A. exigua</i> Grunow
cir M	<i>A. flexella</i> (Kützing) Brun
alf	<i>A. hungarica</i> Grunow
alf	<i>A. lanceolata</i> (De Brébisson) Grunow
cir	<i>A. linearis</i> (W. Smith) Grunow
cir	<i>A. minutissima</i> Kützing
cir	<i>A. peragalli</i> Brun & Heribaud
alb	<i>Actinopteryx undulatus</i> (Bailey) Ralfs
alf	<i>Amphipleura pellucida</i> Kützing
alf	<i>Amphora ovalis</i> Kützing
alf	<i>A. ovalis</i> var. <i>lybica</i> (Ehrenberg) Cleve
alf	<i>A. pediculus</i> (Kützing) Grunow
alb	<i>A. veneta</i> Kützing
-	<i>A. veneta</i> var. <i>capitata</i> Haworth
alf M	<i>Anomoeoneis exilis</i> (Kützing) Cleve
cir	<i>A. exilis</i> f. <i>lanceolata</i> A. Mayer
-	<i>A. intermedia</i> Østrup (with some doubt)
acb M	<i>A. serians</i> (De Brébisson) Cleve
acf	<i>A. serians</i> var. <i>brachysira</i> (De Brébisson) Cleve
acf	<i>A. serians</i> var. <i>brachysira</i> fo. <i>thermalis</i> (Grunow) Hustedt
alb	<i>A. sphaerophora</i> (Kützing) Pfitzer
alb	<i>Bacillaria paradoxa</i> Gmelin
alf	<i>Caloneis amphishaena</i> (Bory) Cleve
	<i>C. lagerstedtii</i> (Lagerstedt) Cholnoky
alf	<i>Cocconeis placentula</i> Ehrenberg
-	<i>Coscinodiscus</i> spec.
alf	<i>Cyclotella comta</i> (Ehrenberg) Kützing
alf	<i>C. meneghiniana</i> Kützing
cir	<i>C. stelligera</i> Cleve & Grunow
alf	<i>Cymatopleura solea</i> (De Brébisson) W. Smith
alb	<i>Cymatosira belgica</i> Grunow
cir M	<i>Cymbella aequalis</i> W. Smith
alf	<i>C. aspera</i> (Ehrenberg) Cleve
acf M	<i>C. cesatii</i> (Rabenhorst) Grunow
alf	<i>C. cistula</i> (Hemprich) Kirchner
acf M	<i>C. gracilis</i> (Rabenhorst) Cleve
alf M	<i>C. hebridica</i> (Gregory) Grunow
alf	<i>C. microcephala</i> Grunow
cir	<i>C. naviculiformis</i> Auerswald
acf	<i>C. perpusilla</i> A. Cleve
alf	<i>C. prostrata</i> (Berkeley) Cleve
alf	<i>C. turgida</i> Gregory
cir	<i>C. ventricosa</i> Agardh
alf	<i>Diatoma vulgare</i> Bory
alf	<i>Diploneis interrupta</i> (Kützing) Cleve
alb	<i>Epithemia sorex</i> Kützing
alb	<i>E. zebra</i> (Ehrenberg) Kützing
acf M	<i>Eunotia alpina</i> (Nägeli) Hustedt
cir	<i>E. arcus</i> Ehrenberg
acf	<i>E. denticulata</i> (De Brébisson) Rabenhorst

- acb *E. exigua* (De Brébisson) Rabenhorst  
 acf M *E. faba* (Ehrenberg) Grunow  
 acf *E. fallax* A. Cleve  
 acf *E. flexuosa* (De Brébisson) Kützing  
 cir *E. lunaris* (Ehrenberg) Grunow  
 acf *E. meisteri* Hustedt  
 acf M *E. monodon* var. *bidens* (Gregory) W. Smith  
 acf *E. pectinalis* (Dillwyn) Rabenhorst  
 acf *E. pectinalis* var. *minor* (Kützing) Rabenhorst  
 acf *E. pectinalis* var. *undulata* Ralfs  
 acf *E. pectnalis* var. *ventralis* (Ehrenberg) Hustedt  
 acf *E. polydentula* Brun  
 acf *E. praerupta* var. *bidens* Grunow  
 acf M *E. robusta* var. *diadema* (Ehrenberg) Ralfs  
 acf M *E. robusta* var. *tetraodon* (Ehrenberg) Ralfs  
 acf *E. sudetica* O. Müller  
 acf *E. tenella* (Grunow) Hustedt  
 acb? *E. tenella/exigua* complex  
 acf M *E. valida* Hustedt  
 acf M *E. veneris* (Kützing) De Toni  
 – *E. spec.* 351.1  
 alf *Fragilaria capucina* Desmazières  
 alf *F. construens* (Ehrenberg) Grunow  
 cir *F. lapponica* Grunow  
 alf *F. leptostauron* (Ehrenberg) Hustedt  
 alf *F. pinnata* Ehrenberg  
 alf *F. vaucheriae* (Kützing) Petersen  
 cir M *F. virescens* Ralfs  
 acf *Frustulia rhomboides* (Ehrenberg) De Toni  
 acb *F. rhomboides* var. *saxonica* (Rabenhorst) De Toni  
 alb *Gomphoneis olivaceum* (Hornemann) Dawson ex Ross & Sims  
 alf *Gomphonema acuminatum* Ehrenberg  
 alf *G. acuminatum* var. *coronatum* (Ehrenberg) W. Smith  
 alf *G. acuminatum* var. *trigonocephalum* (Ehrenberg) Grunow  
 alf *G. angustatum* (Kützing) Rabenhorst  
 alf *G. angustatum* var. *sarcophagus* (Grunow) Hustedt  
 alf *G. constrictum* Ehrenberg  
 cir *G. gracile* Ehrenberg  
 alf *G. intricatum* Kützing  
 cir *G. parvulum* (Kützing) Grunow  
 cir *G. subclavatum* Grunow  
 cir *Hantzschia amphioxys* (Ehrenberg) Grunow  
 acf M *Melosira distans* (Ehrenberg) Kützing  
 alf *M. varians* Agardh  
 alf *Meridion circulare* (Greville) Agardh  
 alf *Navicula accomoda* Hustedt  
 alf *N. atomus* (Kützing) Grunow  
 alf *N. avenacea* De Brébisson  
 acf? *N. brevissima* Hustedt (with some doubt)  
 alf *N. cryptocephala* Kützing  
 alf *N. dicephala* (Ehrenberg) W. Smith  
 alf *N. exilis* Kützing  
 acb M *Navicula festiva* (Östrup) Krasske  
 alf *N. gracilis* Ehrenberg  
 alf *N. gregaria* Donkin  
 acf? *N. heimansii* Van Dam & Kooyman  
 acb *N. hoefleri* Chohnoky  
 alf *N. hungarica* Grunow  
 alb? *N. hungarica* var. *lueneburgensis* Grunow  
 cir *N. integra* (W. Smith) Ralfs  
 acf *N. mediocris* Krasske  
 alf *N. minima* Grunow  
 alf *N. modica* Hustedt  
 cir *N. mutica* Kützing  
 alf *N. pelliculosa* Hilse  
 alf *N. peregrina* fo. *minor* Kolbe  
 alf *N. pseudohalophila* Chohnoky  
 cir *N. pupula* Kützing  
 cir *N. radiosa* Kützing  
 alf *N. rhynchocephala* Kützing  
 alf *N. seminulum* Grunow  
 acb M *N. subtilissima* Cleve  
 – *N. variostrata* Krasske (with some doubt)  
 alf *N. veneta* Kützing  
 alf *N. viridula* Kützing  
 – *N. spec.* 351.1  
 cir *Neidium affine* (Ehrenberg) Cleve  
 alf *Ne. affine* var. *amphirhynchus* (Ehrenberg) Cleve  
 acf *Ne. affine* var. *longiceps* (Gregory) Cleve  
 cir *Ne. iridis* (Ehrenberg) Cleve  
 cir *Ne. iridis* fo. *vernalis* Reichelt  
 acb *Ne. ladogensis* var. *densestriata* (Cleve) Foged  
 alf *Nitzschia acula* Hantzsch  
 alf *Ni. amphibia* Grunow  
 alf *Ni. dissipata* (Kützing) Grunow  
 alf *Ni. frustulum* (Kützing) Grunow  
 – *Ni. gandersheimiensis* Krasske  
 cir *Ni. gracilis* Hantzsch  
 alf *Ni. hantzschiana* Rabenhorst  
 alf *Ni. hungarica* Grunow  
 alf *Ni. inconspicua* Grunow  
 cir *Ni. intermedia* Hantzsch ex Cleve & Grunow  
 alf *Ni. levidensis* (W. Smith) Grunow  
 cir *Ni. palea* (Kützing) W. Smith  
 alf *Ni. paleacea* Grunow  
 alf *Ni. perminuta* Grunow  
 cir *Ni. perpusilla* (Kützing) Grunow emend. Lange-Bertalot  
 – *Ni. tenuis* W. Smith  
 cir *Ni. umbonata* (Ehrenberg) Lange-Bertalot  
 (= *Ni. thermalis* (Ehrenberg) Auerswald)  
 – *Ni. spec.*  
 acf *Peronia heribaudii* Brun & Peragallo  
 acf M *Pinnularia appendiculata* (Agardh) Cleve  
 acf *P. biceps* Gregory  
 acf *P. borealis* Ehrenberg  
 acf *P. braunii* var. *amphicephala* (A. Mayer) Hustedt  
 cir *P. gibba* Ehrenberg  
 cir *P. microstauron* (Ehrenberg) Cleve  
 cir *P. microstauron* var. *brebissonii* (Kützing) Hustedt  
 acf *P. nobilis* Ehrenberg  
 acf *P. silvatica* Petersen  
 acf *P. streptoraphe* Cleve  
 acf M *P. undulata* Gregory  
 cir *P. viridis* (Nitzsch) Ehrenberg  
 – *P. spec.* 293.1  
 – *P. spec.* 363.1  
 alf *Rhoicosphenia curvata* (Kützing) Grunow

alf *Stauroneis anceps* fo. *gracilis* (Ehrenberg) Cleve  
 cir *S. kriegeri* Patrick  
 cir *S. phoenicenteron* (Nitzsch) Ehrenberg  
 alf *S. smithii* Grunow  
 – *S. spec.*  
 acf M *Stenopterobia arctica* Cleve-Euler  
 acf *S. intermedia* Lewis  
 alb *Stephanodiscus astraea* (Ehrenberg) Grunow  
 alb *S. dubius* (Fricke) Hustedt  
 alb *S. hantzschii* Grunow  
 alf *Surirella angusta* Kützing  
 acf M *S. arctissima* Schmidt  
 alf *S. biseriata* De Brébisson

– *S. didyma* Kützing  
 cir *S. linearis* W. Smith  
 cir *S. linearis* var. *constricta* (Ehrenberg) Grunow  
 alf *S. ovata* Kützing  
 alf *Synedra pulchella* (Ralfs) Kützing  
 cir *S. rumpens* Kützing  
 alf *S. tabulata* (Agardh) Kützing  
 cir M *S. tenera* W. Smith  
 alf *S. ulna* (Nitzsch) Ehrenberg  
 acb M *Tabellaria binalis* (Ehrenberg) Grunow  
 acf *T. flocculosa* (Roth) Kützing  
 acb *T. quadriseptata* Knudson