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IMPACT OF ACIDIFICATION AND EUTROPHICATION ON MACROPHYTE COMMUNITIES IN SOFT WATERS IN THE NETHERLANDS
I. FIELD OBSERVATIONS

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ABSTRACT

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During the last decades a strong decline has been noticed in the number of waters dominated by "*Littorellion*" species, mostly isoetids such as *Lobelia dortmanna* L., *Isoetes lacustris* L. and *Littorella uniflora* (L.) Aschers. Sixty-eight waters, which were known to be dominated by *L. uniflora* after 1950 were investigated. In 1980, *L. uniflora* appeared to be absent or to have strongly decreased in 53 (78%) of the waters. In 41 of them, *Littorella* had been replaced by submerged *Juncus bulbosus* L. and/or *Sphagnum* spp. These changes seem to have been caused by changed inorganic carbon budgets as a consequence of acidification.

In the remaining 12 waters, eutrophication of the water and/or sediment seems to be responsible for the changes in the plant communities. Enrichment with phosphate of the mineral sediment alone, leads to luxurious growth of submerged, rooted macrophyte species such as *Myriophyllum alterniflorum* DC and *Ranunculus peltatus* Schrank, whereas phosphate-enrichment of both sediment and water leads to luxurious growth of pleustophytes such as *Riccia fluitans* L. and *Lemna minor* L. in small, shallow waters, and to plankton bloom and luxurious growth of epiphytes in larger, deeper waters.

In these cases light limitation seems to be responsible for the disappearance or decline of the "*Littorellion*" species.

INTRODUCTION

During the past 20 years the deleterious effects of acid precipitation on aquatic and terrestrial ecosystems have become a growing problem. Reports from The First International Symposium on Acid Precipitation and Forest Ecosystem (Dochinger and Seliga, 1975), the United Nations Conference of the Human Environment (Engstrom, 1971) and of the International Conference on the Impact of Acid Precipitation (Drabløs and Tollan, 1980) emphasize the gravity of the phenomenon.

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Aquatic habitats appear to be very susceptible to the effects of acidification. The severe depletion of fish stocks and the decreased diversity of algae and invertebrates in many Swedish and Norwegian waters can be ascribed to the effects of acidification. It is only fairly recently that a few reports have been published which express alarm about acidifying waters on poorly buffered sediments in The Netherlands (van Dam and Kooyman-van Blokland, 1978; van Dam et al., 1981; van Zellem, 1981) and Belgium (Vangenechten et al., 1980).

The mechanisms of acidification, however, appear to be very complex and are not yet clearly understood. In many cases the impact of acidification cannot be ascribed to pH decrease alone. Brooktrout, *Salvelinus fontinalis* (Mitchell), for instance, seem to suffer mainly from toxic aluminium concentrations mobilized by increasing acidification, as they are able to tolerate substantial additions of sulphuric acid (Daye and Garside, 1975). It is also known that heavy metals are more toxic to algae at low pH (Voigt, 1979). Inhibition of nitrogen-turnover processes (Tamm et al., 1977; Alexander, 1980) and leaching (Stuanes, 1980) are likely to cause future nutrient deficiencies in forest ecosystems.

However, little is known about the impact of acidification on freshwater macrophytes. Grahn et al. (1974) and Grahn (1977) mention a suppression of isoetid species in Scandinavian waters by luxurious *Sphagnum* growth as a result of acidification of the water and Nilssen (1980) mentions a luxurious growth of *Juncus bulbosus* L. in acidified Scandinavian waters.

In The Netherlands there has been a dramatic decline in the number of stands belonging to the phytosociological alliance *Littorellion* during the last 30 years. These stands are characterized by isoetids, such as *Littorella uniflora* (L.) Aschers., *Lobelia dortmanna* L., *Isoetes lacustris* L., etc. (Schoof-van Pelt, 1973). These "*Littorellion* communities" mainly occurred in moorland pools, in small lakes and dune lakes on mineral sandy soils. It is precisely these poorly buffered waters that have become strongly acidified during the last decades (van Dam and Kooyman-van Blokland, 1978; van Dam et al., 1981). The coastal dune lakes appeared to be less susceptible to acidification, due to the higher content of calcium carbonate of the bottom.

Eutrophication has been generally considered to be the main cause of the decline of the *Littorellion* communities (Schoof-van Pelt, 1973; Westhoff et al., 1973; Westhoff, 1979; Pott, 1982; Wittig, 1982a,b; Wittig and Pott, 1982); Westhoff (1979) also mentioned inadequate maintenance in nature reserves as a secondary cause. Acidification was not mentioned by these authors as being a possible cause.

The present study was carried out in order to establish which part of the decline of the "*Littorellion*" communities can be ascribed to acidification, which part is due to eutrophication and possible other causes, and which changes in the composition of the macrophyte communities occur as a result of these processes.

STUDY SITES

Sixty-eight low alkaline waters were selected, where, according to Schoof-van Pelt (1973), Westhoff et al. (1971) and H.M. van der Steeg, C. den Hartog, D.T.A. van der Ploeg, E. Weeda and J. Hofstra (personal communication), after 1950 a plant community occurred, in which *Littorella uniflora* was one of the dominant species.

Low alkaline waters in The Netherlands are restricted to the higher situated, poorly buffered, sandy soils in the southern and eastern part of the country and to the coastal region (Fig. 1). In the provinces of Noord Brabant, Limburg, Gelderland, Overijssel and Drenthe mainly moorland pools and some small lakes were studied and only dune pools in the province of Zuid-Holland and on the isles of Texel, Terschelling and Schiermonnikoog.

Fig. 1. Study areas. One dot may represent more than one investigated water.

MATERIALS AND METHODS

During 1979 and 1980 each water was visited four times. On location, the floral composition was investigated and pH-measurements were carried out with a Metrohm model E 488 pH-meter and a model EA 152 combined electrode. Alkalinity was estimated by titration of 100 ml of water with 0.01 M HCl down to pH 4.2. Water samples were taken in 200-ml iodated polyethylene bottles, immediately passed through a Whatman GF/C filter and fixed with 1 ml of a 200 mg l^{-1} $HgCl_2$ solution. In the micro-habitats of each investigated plant species (Tables III and IV) sediment samples were taken with a brass tube (length 10 cm, diameter 5.6 cm). On each location 6 subsamples were taken and decanted into a 2-l iodated polyethylene bottle. All samples were transported to the laboratory in a refrigerated container.

After arrival at the laboratory, the water samples were immediately frozen and stored at −20°C until use. Redox potential measurements in the sediment samples were carried out within 24 h with a Metrohm model E 488 pH/mV meter and a model EA 217 platinum electrode. For the analysis of the interstitial water 20 g of wet well-mixed sediment were weighed into a 400-ml conical flask and shaken on a Gerhardt model LS 20 shaker during 1 h, after addition of 200 ml bidistilled water. After centrifugation in a Heraeus Crist model 111 labofuge (10 min, 5000 r.p.m.) the acidity was estimated by titration of 50 ml of the supernatant with 0.01 M NaOH up to pH 8.2. The alkalinity was estimated by titration of 50 ml of the supernatant with 0.01 M HCl down to pH 4.2. One-hundred ml of the supernatant was fixed by adding 0.5 ml of a 200 mg l^{-1} $HgCl_2$ solution and stored at −20°C until use. The water content of the sediment samples was estimated by weighing, drying (24 h, 105°C) and reweighing. One-hundred mg of dried sediment were digested in a mixture of 0.5 ml concentrated perchloric acid and 0.2 ml sulphuric acid under pressure (4 h, 170°C), according to Kotz et al. (1972), in order to obtain the elemental composition. For all samples, calcium was estimated with a Beckman model 1272 Atomic Absorption Spectrophotometer, sodium and potassium were estimated flame-photometrically using a Technicon I Auto Analyzer and colorimetrically with a Technicon II Auto Analyzer: orthophosphate according to Hendriksen (1965), nitrate according to Kamphake et al. (1967), ammonia according to Grasshoff and Johannsen (1977), chloride according to O'Brien (1962) and sulphate according to Technicon methodology (1981). The carbon dioxide content was calculated from acidity and that of hydrogen carbonate was calculated from alkalinity, both with a correction for pH.

RESULTS

Macrophyte composition

In 1979, only 15 (~ 22%) of the 68 waters investigated, in which *Littorella uniflora* was one of the dominant species after 1950, were still dominated by this species; most of these stands occurred in the coastal region (Table Ia).

The 53 waters in which *L. uniflora* had disappeared or had strongly decreased can be divided into two groups. The major group (group 1: 41 waters) is characterized by the presence of submerged *Juncus bulbosus* and/or *Sphagnum* spp. and a very low turbidity of the water (Table Ib). Twenty-five of these waters were dominated by *J. bulbosus*, 12 of them by *Sphagnum* spp. and in four of them no submerged plants were present except *J. bulbosus* and *Sphagnum* spp. in very small quantities. The waters of the minor group (group 2: 12 waters) are characterized by the absence of submerged *J. bulbosus* and/or *Sphagnum* spp. (Table Ic). Five of these waters were dominated by *Myriophyllum alterniflorum* DC, three of them by *Riccia fluitans* L. and *Lemna minor* L. and four of them were free of submerged macrophytes. In these last four waters the turbidity was very high as a result of luxurious plankton growth. Considerable differences were also noticed in the composition and coverage of the accompanying species (Tables Ia—c).

Eleocharis acicularis (L.) R. et S. is common and generally covered more than 5% of the bottom in the waters dominated by *Littorella uniflora*, it occurred only sporadically with a low coverage in waters dominated by *J. bulbosus*, and was absent in waters dominated by *Sphagnum* spp. In waters dominated by *M. alterniflorum*, *E. acicularis* showed a mean coverage and a frequency equal to those in waters dominated by *L. uniflora*, whereas *E. acicularis* was absent in waters dominated by *Riccia fluitans* and *Lemna minor*.

Luronium natans (L.) Raf. showed a similar pattern. It is common in waters dominated by *Littorella uniflora*, is less numerous in waters dominated by *J. bulbosus* and is scarce in waters dominated by *Sphagnum* spp. In the waters dominated by *M. alterniflorum*, *Luronium natans* is frequent, whereas in waters dominated by *Riccia fluitans* and *Lemna minor* it is absent.

Potamogeton gramineus L. is restricted to waters dominated by *Littorella uniflora. Polygonum amphibium* L. and *Eleocharis palustris* (L.) R. et S. were both frequent in waters dominated by *Littorella uniflora, M. alterniflorum* and *Riccia fluitans/Lemna minor*; and were scarce in waters dominated by *J. bulbosus* and *Sphagnum* spp.

Carex rostrata Stokes and *Eriophorum angustifolium* Honck. occurred more frequently in the waters dominated by *J. bulbosus. Eleocharis multicaulis* (Sm.) Sm. was absent in waters dominated by *Littorella uniflora* (without *J. bulbosus*), *M. alterniflorum* and *Riccia fluitans. Ranunculus peltatus* Schrank and *Elodea nuttallii* (Planch.) St. John both occurred only in waters dominated by *Myriophyllum alterniflorum.*

TABLE Ia

The floral composition of the waters in which *Littorella uniflora* was one of the dominant plant species in 1979—1980

Number of waters	Dominant species	Accompanying species	Frequency*
11	*Littorella uniflora* (L.) Aschers.	*Eleocharis acicularis* (L.) R. et S.	7 +
		Eleocharis palustris (L.) R. et S.	6
		Hydrocotyle vulgaris L.	6
		Echinodorus ranunculoides (L.) Engelman	5
		Luronium natans (L.) Raf.	5 +
		Potamogeton gramineus L.	5 +
		Ranunculus flammula L.	5
		Carex rostrata Stokes	4
		Elatine hexandra (Lapierre) DC	4 +
		Myosotis scorpioides L.	4
		Peplis portula L.	3
		Polygonum amphibium L.	3 +
		Chara globularis Thuill.	2
		Carex lasiocarpa Ehrh.	2
		Echinodorus repens (Lamk.) Kern et Reichg.	2
		Myriophyllum alterniflorum DC	2
		Eriophorum angustifolium Honck.	1
		Hypericum elodes L.	1
		Menyvanthes trifoliata L.	1
		Nuphar lutea (L.) Sm.	1
		Ranunculus ololeucos Lloyd	1
4	*Littorella uniflora* (L.) Aschers. and *Juncus bulbosus* L.	*Hydrocotyle vulgaris* L.	3
		Sphagnum spp.	3 +
		Carex rostrata Stokes	2
		Eleocharis multicaulis (Sm.) Sm.	2 +
		Eleocharis palustris (L.) R. et S.	2
		Lobelia dortmanna L.	2
		Ranunculus flammula L.	2
		Echinodorus repens (Lamk.) Kern et Reichg.	1
		Hypericum elodes L.	1
		Luronium natans (L.) Raf.	1 +
		Isoëtes lacustris L.	1 +

*Frequency: the number of waters in which the species was present. Species marked with + often had a coverage of >5%.

TABLE Ib

The floral composition of the waters in which *Littorella uniflora* was not one of the dominant species and in which submerged *Juncus bulbosus* and/or *Sphagnum* occurred in 1979—1980

Number of waters	Dominant species	Accompanying species	Frequency
25	*Juncus bulbosus* L.	*Sphagnum* spp.	12 +
		Carex rostrata Stokes	11 +
		Hypericum elodes L.	8
		Eleocharis palustris (L.) R. et S.	8
		Hydrocotyle vulgaris L.	7
		Luronium natans (L.) Raf.	7
		Eriophorum angustifolium Honck.	7 +
		Utricularia minor L.	7 +
		Nymphaea alba L.	6
		Ranunculus flammula L.	6
		Ranunculus ololeucos Lloyd	6
		Peplis portula L.	6
		Potamogeton natans L.	6
		Carex lasiocarpa Ehrh.	2 +
		Echinodorus ranunculoides (L.) Engelman	2
		Eleocharis acicularis (L.) R. et S.	2
		Menyanthes trifoliata L.	1
		Nuphar lutea (L.) Sm.	1
		Polygonum amphibium L.	1
12	*Sphagnum* spp. (most *S. cuspidatum* Hoffm.)	*Carex rostrata* Stokes	10 +
		Eriophorum angustifolium Honck.	9 +
		Hydrocotyle vulgaris L.	8
		Eleocharis multicaulis (Sm.) Sm.	6
		Utricularia minor L.	5 +
		Eleocharis palustris (L.) R. et S.	4
		Menyanthes trifoliata L.	4
		Hypericum elodes L.	4
		Ranunculus flammula L.	4
		Carex lasiocarpa Ehrh.	3
		Juncus articulatus L.	3
		Luronium natans (L.) Raf.	2
		Nymphaea alba L.	2
		Polygonum amphibium L.	2
4	Clear water no submerged macrophytes except *Juncus bulbosus* and *Sphagnum* spp. in very small quantities		

TABLE Ic

The floral composition of the waters in which *Littorella uniflora* was not one of the dominant species and in which no submerged *Juncus bulbosus* and/or *Sphagnum* spp. occurred in 1979—1980

Number of waters	Dominant species	Accompanying species	Frequency
5	*Myriophyllum alterniflorum* DC	*Eleocharis palustris* (L.) R. et S.	5 +
		Hydrocotyle vulgaris L.	4
		Littorella uniflora (L.) Aschers.	4 +
		Luronium natans (L.) Raf.	4 +
		Myosotis scorpioides L.	4
		Ranunculus peltatus Schrank	4 +
		Ranunculus flammula L.	4
		Apium inundatum (L.) Rchb.f.	2
		Chara globularis Thuill.	2 +
		Eleocharis acicularis (L.) R. et S.	2 +
		Echinodorus ranunculoides (L.) Engelman	2
		Elatine hexandra (Lapierre) DC	2 +
		Nuphar lutea (L.) Sm.	2
		Polygonum amphibium L.	2 +
		Nymphaea alba L.	1
		Peplis portula L.	1
		Elodea nuttallii (Planch.) St. John	1
3	*Riccia fluitans* L.	*Lemna trisulca* L.	3 +
	Lemna minor L.	*Eleocharis palustris* (L.) R. et S.	2
		Hydrocotyle vulgaris L.	2
		Myosotis scorpioides L.	2
		Nuphar lutea (L.) Sm.	2 +
		Ranunculus flammula L.	2
		Rorippa amphibia (L.) Besser	2
		Peplis portula L.	1
4	Turbid water (algal bloom) no submerged macrophytes		

Frequency: the number of waters in which the species was present. Species marked with + often had a coverage >5%.

Chemistry of water and sediments

Water chemistry

The results are shown in Table II. Waters dominated by *Littorella uniflora* had a mean alkalinity of 0.50 meq. l^{-1} and those dominated by *J. bulbosus* of 0.02 meq. l^{-1}; however, it has to be noted that most waters with *J. bulbosus* as a dominant species had no buffer capacity at all. The waters dominated by *Sphagnum* spp. had a mean alkalinity of 0.01 meq. l^{-1}, those dominat-

Water chemistry

	n*	pH "average"	Alkalinity (HCO_3^- mmol l^{-1})		SO_4^{2-} (μmol l^{-1})		Cl^- (μmol l^{-1})		Ca^{2+} (μmol l^{-1})	
			Mean	90%	Mean	90%	Mean	90%	Mean	90%
Littorella uniflora	33	6.5	0.50	0.1—1.0	336	100—1000	638	40—4000	262	100— 300
Juncus bulbosus	107	3.9	0.02	0.0—0.2	780	300—2000	666	80—1500	310	100— 500
Sphagnum spp.	59	3.8	0.01	0.0—0.2	530	100—1500	448	80— 500	160	100— 300
Myriophyllum alterniflorum	25	6.9	0.80	0.2—2.0	660	500—1500	795	100—1500	620	400—1500
Ranunculus peltatus	62	6.8	1.10	0.4—2.0	822	200—2000	979	200—2000	710	300—1500
Riccia fluitans	31	6.4	0.80	0.2—2.0	607	200—1000	687	250—1000	564	300—1500
Lemna minor	91	6.5	0.90	0.2—4.0	836	200—2000	1001	80—4000	679	200—1500

*Number of samples investigated.

TABLE IIb

Water chemistry

	n*	NH_4^+ (μmol l^{-1})		NO_3^- (μmol l^{-1})		PO_4^{3-} (μmol l^{-1})		K^+ (μmol l^{-1})	
		Mean	90%	Mean	90%	Mean	90%	Mean	90%
Littorella uniflora	33	5	0— 10	10	0— 50	0.16	0.0— 0.5	134	50—300
Juncus bulbosus	107	40	2— 200	7	0— 50	0.21	0.0— 0.5	163	10—400
Sphagnum spp.	59	46	2— 400	7	0— 50	0.19	0.0— 0.5	79	10—200
Myriophyllum alterniflorum	25	23	0— 50	44	0— 100	0.22	0.0— 0.5	139	50—300
Ranunculus peltatus	62	44	0— 100	162	0— 600	0.44	0.0— 5.0	176	20—400
Riccia fluitans	31	27	2— 50	85	0— 10	2.20	0.1—20.0	190	50—400
Lemna minor	91	68	2—1000	224	0—2000	1.70	0.1—20.0	210	50—750

*Number of samples investigated.

ed by *M. alterniflorum* and *Riccia fluitans* and/or *Lemna minor* had a mean alkalinity of 0.80 meq. l^{-1}, thus higher than that of the *Littorella*-dominated waters. From the investigated plant species, *J. bulbosus* and *Sphagnum* spp. occurred mainly in acid waters (pH on an "average" 3.8) while the other species mostly occur in circumneutral waters.

The mean sulphate concentration was the lowest in waters dominated by *Littorella uniflora* and clearly higher in the waters dominated by *J. bulbosus, M. alterniflorum* and *R. fluitans.* The mean ammonium concentration was low in the waters dominated by *L. uniflora* and clearly higher in the waters dominated by *J. bulbosus* and *Sphagnum* spp. However, the mean nitrate concentration was the lowest in waters dominated by *J. bulbosus* and *Sphagnum* spp., somewhat higher in waters dominated by *L. uniflora,* and much higher in the waters dominated by *M. alterniflorum* and *R. fluitans.*

The mean concentrations of ortho-phosphate were practically identical in the waters dominated by *L. uniflora, J. bulbosus, Sphagnum* spp. and *M. alterniflorum* and much higher in waters dominated by *R. fluitans* and *Lemna minor.*

The mean concentrations of potassium, calcium and chloride did not differ much in the investigated waters, except for waters dominated by *Sphagnum* spp., in which they were lower, and for waters dominated by *M. alterniflorum*, in which they were higher.

Sediment analysis (Tables III and IV)

The mean HCO_3^- concentration of the interstitial water was the lowest for *J. bulbosus,* somewhat higher for *Sphagnum* spp. and *Littorella uniflora* and much higher for *M. alterniflorum* and *R. fluitans.*

The mean CO_2 concentration, however, was the highest for *J. bulbosus* and *Sphagnum* spp., clearly lower for *M. alterniflorum* and the lowest for *L. uniflora.*

The mean concentrations of ortho-phosphate in the interstitial waters were almost equal for *L. uniflora, J. bulbosus* and *Sphagnum* spp. and clearly higher for *M. alterniflorum* and *R. fluitans.* The mean nitrate concentration was the highest for *L. uniflora,* lower for *J. bulbosus* and *Sphagnum* spp. and much lower for *Lemna minor* and *R. fluitans.*

The mean ammonium concentrations, however, were the lowest for *Littorella uniflora,* higher for *J. bulbosus* and *Sphagnum* spp. and the highest for *Lemna minor* and *R. fluitans.*

The mean calcium concentration was almost equal for *Littorella uniflora, J. bulbosus* and *Sphagnum* spp. and much higher for *M. alterniflorum, Ranunculus peltatus, Lemna minor* and *Riccia fluitans.*

The mean total N concentration in the sediment was the lowest for *Littorella uniflora*, somewhat higher for *J. bulbosus* and *M. alterniflorum* and much higher for *Sphagnum* spp., *Lemna minor* and *R. fluitans.*

The mean total P concentration also was the lowest for *Littorella uniflora*, higher for *J. bulbosus, Sphagnum* spp., *M. alterniflorum, R. fluitans* and the highest for *Lemna minor.*

TABLE III

Chemical composition of the interstitial water

	n*	HCO_3^- (mmol l^{-1})		CO_2 (mmol l^{-1})		PO_4^{3-} (µmol l^{-1})		NO_3^- (µmol l^{-1})		NH_4^+ (µmol l^{-1})		Ca^{2+} (µmol l^{-1})	
		Mean	90%	Mean	90%	Mean	90%	Mean	90%	Mean	90%	Mean	90%
Littorella uniflora	9	1.02	0.06— 3.00	0.90	0.09—1.50	5.1	0.0— 7.5	90	15—180	267	150— 390	708	200—1500
Juncus bulbosus	19	0.42	0.00— 2.10	3.33	0.90—9.60	5.4	0.0— 15.0	60	15—180	510	150—1200	627	200—1500
Sphagnum spp.	9	0.78	0.00— 3.00	5.01	1.20—9.90	5.4	0.0— 15.0	51	15— 75	564	150—1500	675	200—1500
Myriophyllum alterniflorum	9	4.80	1.20— 9.00	1.65	0.00—3.90	11.4	1.5— 30.4	73	15—180	381	150— 600	1650	750—3000
Ranunculus peltatus	15	3.60	0.90— 9.90	1.86	0.00—7.50	24.0	3.6— 75.0	81	15—180	333	150— 900	1650	750—3000
Riccia fluitans	10	7.06	1.10—36.20	0.50	0.00—1.30	29.9	3.1—154.0	16	0— 35	1270	182—3790	1700	600—3000
Lemna minor	11	7.20	2.20—18.00	1.05	0.24—2.20	33.0	26.3— 68.8	29	9—106	1250	900—2400	1890	600—4000

*Number of samples investigated.

TABLE IV

Physico-chemical properties of the sediment

	n*	Redoxpotential (mV)	Total N (μmol g^{-1})		Total P (μmol g^{-1})		Total Ca (μmol g^{-1})		Organic weight (% dry weight)	
		Mean	Mean	90%	Mean	90%	Mean	90%	Mean	90%
Littorella uniflora	9	+70	35	14— 90	1.8	0.7— 5.0	7	0— 17	2.9	0.7— 5.0
Juncus bulbosus	19	+102	67	14— 90	4.8	1.0— 10.0	14	1— 80	8.1	0.9—20.0
Sphagnum spp.	9	+194	235	50— 300	7.2	2.0— 25.0	16	0— 82	20.0	5.3—60.0
Myriophyllum alterniflorum	9	−40	65	10— 300	10.5	2.0— 40.0	56	10—170	6.5	0.8—25.0
Ranunculus peltatus	15	+47	109	14— 400	20.0	5.0—100.0	40	5—180	5.6	0.8—20.0
Riccia fluitans	10	−150	544	70—1300	14.2	7.0— 34.0	75	8—180	23.0	4.3—78.0
Lemna minor	11	−152	247	47— 820	22.0	11.0— 33.0	64	8—103	11.4	3.4—41.6

*Number of samples investigated.

The mean total Ca concentrations show similar results: the lowest for *Littorella uniflora* and the highest for *R. fluitans*.

The mean organic weight of the sediment was the lowest for *L. uniflora*, followed by *Ranunculus peltatus*, *M. alterniflorum* and *J. bulbosus* and much higher for *Sphagnum* spp. and *Riccia fluitans*.

DISCUSSION

All observations indicate that acidification of the water caused by acid precipitation leads to a decrease and finally to a disappearance of *Littorella uniflora* and other "*Littorellion*"-species, coincident with the appearance and often luxurious growth of *J. bulbosus*, *Sphagnum* spp., *Eleocharis multicaulis*, *Eriophorum angustifolium*, etc. (Table Ib). The pH of the water declines strongly, on an average from pH 6.5 for *L. uniflora* down to pH 3.8 for *J. bulbosus* and *Sphagnum* spp. Alkalinity decreases on an average from 0.50 meq. l^{-1} for *L. uniflora* down to 0.02 and 0.01 meq. l^{-1} for *J. bulbosus* and *Sphagnum* spp., respectively Wiegleb (1978) mentions similar data for waters in the Federal German Republic: a mean alkalinity of 0.50 meq. l^{-1} for *L. uniflora* and 0.05 meq. l^{-1} for *J. bulbosus*. Sand-Jensen and Rasmussen (1978) also found *J. bulbosus* to be the only vascular plant species in strongly acidified Scandinavian waters and Nilssen (1980) mentions a luxurious growth of *J. bulbosus* in acidified waters in Norway. Grahn et al. (1974) and Grahn (1977) mention a suppression of isoetid species in Scandinavian waters by luxurious *Sphagnum* growth as a result of acidification of the water. Investigations by Spence (1964, 1967) clearly show that "*Littorellion*" species such as *L. uniflora* and *Lobelia dortmanna* mainly occur in non-acid, poorly buffered waters.

Acidification of the water brings about not one, but a number of changes in the physical and chemical properties of water and sediment, and it is difficult to establish which of these is responsible for the changes in the macrophyte composition. For instance, the sulphate concentration in *J. bulbosus*-dominated, acidified waters on an average is more than twice as high as in non-acidified *Littorella uniflora*-dominated waters, probably due to precipitation containing sulphuric acid and a possibly reduced sulphate reduction, though it is doubtful whether these sulphate concentrations have any influence on the observed changes in the macrophyte composition, as the maximum SO_4^{2-} concentration at which *L. uniflora* occurs is considerably higher than the mean SO_4^{2-} concentration for *J. bulbosus*.

There are also changes in the nitrogen and phosphorus concentrations. The total amount of mineral nitrogen in waters dominated by *J. bulbosus* and *Sphagnum* spp. (including the interstitial water) is on an average much higher than in waters dominated by *L. uniflora*, but whether this is of any importance is doubtful, as phosphorus is normally the limiting factor in moorland pools (Table IIb).

In particular, the mean ammonium concentrations of water and interstitial water in acidified *J. bulbosus* and *Sphagnum* spp. dominated water bodies are much higher, undoubtedly due to the strongly decreased nitrification as a result of the low pH.

Whether these high ammonium concentrations are toxic to *L. uniflora* under these circumstances has to be doubted. It is true that ammonium toxicity to various species of aquatic macrophytes has been reported (Glänzer et al., 1977), but these experiments were carried out at a relatively high pH, at which part of the ammonium occurs as toxic NH_3 (Warren, 1962). However, the pH in the acidified waters is low enough to keep all ammonium as non-toxic NH_4^+. Possibly, however, differences among the various plant species may occur in the ability to use ammonium instead of nitrate as a nitrogen source.

The mean concentration of ortho-phosphate in the water as well as in the interstitial water is almost equal in *L. uniflora*-, *J. bulbosus*- and *Sphagnum* spp.-dominated waters. Therefore, it is not to be expected that the changes in the macrophyte composition are due to changes in the phosphate budget, although the mean total P concentration in the upper sediment layer of the acidified waters is clearly higher than in the non-acidified waters. The accumulation of organic material as a result of strongly decreased microbial activity could partly be responsible, just as the strongly increased redox potential in the upper sediment layer, which leads to accumulation of phosphate coming from the deeper, anaerobic sediment layers. The latter process has been described by Damman (1978) for ombrotrophic peat bogs.

It is likely that the tremendous changes in the inorganic carbon content which occur as a result of acidification, play a very important role. In acidifying waters the inorganic carbon content of the water decreases to practically zero and the submerged macrophytes fully depend on the sediment, as the diffusion of CO_2 from the air into stagnant acid water is insufficient. Sand-Jensen and Søndergaard (1979) discovered that the size of *L. uniflora* plants in water poor in CO_2 depends on the CO_2 level in the sediment.

The data presented here clearly show that in acidified waters dominated by *J. bulbosus* the mean CO_2 concentration of the interstitial water is much higher than in non-acidified, *L. uniflora*-dominated waters. The CO_2 concentration of the interstitial water is highest in the *Sphagnum* spp.-dominated waters. This increase of CO_2 is no doubt due to the acid water layer acting upon the HCO_3^- and CO_3^{2-} in the sediment. This leads to the hypothesis that changes in the macrophyte composition are mainly dependent on the changes in the CO_2 concentration in the sediment, which depends on the HCO_3^- content of the interstitial water and the CO_3^{2-} content of the sediment. Field observations support this hypothesis. In acidifying dune pools above relatively CO_3^{2-}-rich sediments (e.g. van Hünenplak, Terschelling) the very tall *Littorella* plants became fully overgrown by *Sphagnum* spp., whereas in acidifying moorland pools above relatively CO_3^{2-} -poor sediments (e.g., Groothuisven near Tilburg) *L. uniflora* and *Lobelia dortmanna* became overgrown by *J. bulbosus*.

In other acidifying moorland pools, above mainly CO_3^{2-}-free sediments (e.g., Staalbergven near Oisterwijk; Galgenven near Tilburg) *Littorella uniflora* and *Luronium natans* gradually became smaller and almost disappeared, without any other macrophyte species appearing in their place.

Carbon dioxide measurements in the interstitial water of these locations also confirmed the above hypothesis. There is also an explanation for the fact that in some places "*Littorellion*" communities can persist for a very long period after acidification, as was observed in a few cases during this study and has also been mentioned by Pietsch (1972) for some waters in Central Europe. Luxurious growth of *J. bulbosus* and *Sphagnum* spp. can only occur after an increased CO_2 level in the sediment when phosphate or nitrogen are not limiting.

Eutrophication, in its classical meaning (increased input of nutrients such as phosphate and nitrogen), often mentioned as the main cause of the disappearance or decline of "*Littorellion*" species (Schoof-van Pelt, 1973; Pietsch, 1977; Wittig, 1980), has been a less important factor in the investigated waters during the last 30 years than acidification.

A change in the phosphate content seems to be responsible for the changes in all 12 waters where "*Littorellion*" species had strongly declined or had disappeared and no submerged *Sphagnum* spp. or *J. bulbosus* occurred. A phosphate enrichment of the mineral soil, with a strongly increased amount of ortho-phosphate in the interstitial water, but hardly any increased amount of phosphate in the water (e.g., environments rich in iron) leads to luxurious growth of submerged macrophytes, mainly *M. alterniflorum* and/or *Ranunculus peltatus* and a suppression of "*Littorellion*" species. However, after a number of years of luxurious plant growth the submerged plant species disappear again because the sediment becomes more or less organic. It is not clearly understood which processes underlie this phenomenon. Possibly the strongly lowered redox potential and the concomitantly strongly decreased CO_2 and strongly increased NH_4^+ concentration of the sediment which are found under these circumstances are responsible (Tables III and IV). A phosphate enrichment of the system with a clearly increased ortho-phosphate concentration not only in the sediment, but also in the water leads to luxurious growth of non-rooted macrophytes such as *Riccia fluitans* and *Lemna minor* in small, shallow waters. In larger and deeper water bodies the water becomes turbid as a result of algal bloom and/or strong development of epiphytes on the macrophytes. In this case, light reduction seems to be responsible for the decrease or disappearance of the submerged macrophytes (Sand-Jensen and Søndergaard, 1981).

In order to support the preceding hypotheses, and to obtain causal relations between the changes in the physico—chemical environment and plant development, "in vitro" eco-physiological and culture experiments have been carried out with the plant species involved. The results of these experiments will be discussed in part II of this study.

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