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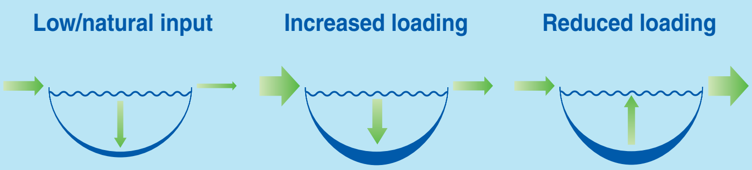
Nutrient dynamics in lakes

– with emphasis on phosphorus, sediment and lake restoration

Doctor's dissertation (DSc), 2007

Martin Søndergaard



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Data sheet

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Abstract: This dissertation including 16 selected English papers and book contributions was accepted by the Faculty of Science, University of Aarhus, for the doctor's degree in natural sciences. The dissertation describes the dynamic interactions between nutrients, the sediment and the biological conditions in lakes. First, Danish lake types are characterised and their development described relative to changes in nutrient loading. After this a more detailed description is given of lake retention of nutrients, including the binding of phosphorus in the sediment and the exchange of phosphorus between the sediment and the water phase. Next follows a section on the multiple mechanisms behind the release and uptake of phosphorus in the sediment. As a user-oriented aspect, lake restoration and the results obtained from the various chemical and biological methods applied in Denmark so far are treated. Finally, reflections are made on future management and research issues for Danish lakes, including the future climate and the implementation of the EU Water Framework Directive.

Keywords: Lakes, lake types, ponds, nutrients, phosphorus, nitrogen, phosphorus fractions, biological structure, internal phosphorus loading, resuspension, redox conditions, submerged macrophytes, sediment, lake restoration, biomanipulation, Water Framework Directive.

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Supplementary note: This doctor's dissertation is available in Danish (excluding the 16 attached papers) entitled Næringsstofdynamik i søer - med fokus på fosfor, sedimentet og restaurering af søer. Doktordisputats.

This dissertation, along with the enclosed 16 original papers, was accepted by the Faculty of Science, University of Aarhus, for public defence for the doctor's degree in natural sciences (DSc). The defence took place on April 27, 14.15 p.m., at the University of Aarhus.

Contents

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Preface

This doctor's dissertation consists of 16 selected associated papers and a compressed account submitted to Aarhus University to assessment for the doctoral degree in natural sciences. The account summarises and places the papers in a broader perspective and may be read as an independent work.

The dissertation, which was submitted in October 2005, was elaborated at the National Environmental Research Institute's Department of Freshwater Ecology in Silkeborg, and it comprises papers published during a 15-year period, from 1990 to 2005. Within the account references are given by numbers to the associated papers as given in the list on page 69.

Besides the National Environmental Research Institute (NERI), I wish to express my gratitude to numerous persons for their contributions to the realisation of this project. First and foremost I wish to thank the lake group dynamo, Erik Jeppesen, who with his great commitment and scientific knowledge has given me invaluable support and inspiration through the long journey of writing this dissertation. I am also grateful to former and present scientific members of the lake group for their hard work and great contribution to our joint efforts for making a difference within lake research: Jens Peder Jensen, Torben Lauridsen, Susanne Lildal Amsinck, Rikke Bjerring Hansen, Asger Roer Pedersen, Lone Liboriussen, Frank Landkildehus, Liselotte Sander Johansson, Peter Kristensen and Ole Sortkjær. The technical members of the lake group deserve much praise and gratitude for their hard work, great skill and dedication ensuring the smooth functioning of all practical aspects: Birte Laustsen, Jane Stougaard-Pedersen, Karina Jensen, Lissa Skov Hansen, Lone Nørgård, John Glargård, Lisbet Sortkjær, Kirsten Thomsen and Ulla Kvist Pedersen. Thanks are also due to Anne Mette Poulsen for linguistic assistance and text layout and to Tinna Christensen, Kathe Møgelvang and Juana Jacobsen from NERI's graphical department for their meticulous drawings and layout assistance. I am also grateful to the past and present research directors, Torben Moth Iversen and Kurt Nielsen, for their positive attitude and encouragement. Special thanks go to Kurt Nielsen and Erik Jeppesen for their valuable comments on earlier versions of the dissertation.

I am also grateful to the Danish counties and their employees for their systematic gathering and processing of data and for their many valuable inputs to integrate science and management. In many respects this has been a prerequisite for writing this dissertation on nutrient dynamics in lakes in more general terms.

Finally, I wish to thank Carlsbergfonden for financial support to the compilation and completion of this dissertation. Over the years the investigations involved have received support from several EU-funded projects (BUFFER, ECOFRAME, EUROLIMPACS) as well as from Danish research projects and institutions (CONWOY, the Strategic Environmental Research Program).

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Danish summary

Afhandlingen er en sammenfatning af 16 udvalgte videnskabelige artikler, der beskriver forskellige aspekter af næringsstofdynamikken og næringsstoffers interaktioner med biologiske forhold i søer.

Beskrivelsen omfatter de fleste danske søtyper, fra småsøer og vandhuller til den største danske sø, Arresø, men dog med hovedvægten lagt på de større og lavvandede søer. I en af artiklerne sammenlignes søer langs en størrelsesgradient, og her identificeres væsentlige forskelle blandt andet betinget af højt humusindhold og fravær af fisk i de mindste søer. Lavvandede søer er den mest udbredte søtype i Danmark og er kendetegnet ved blandt andet en meget tæt kobling mellem vandfase og sediment.

Der er endvidere især lagt vægt på at beskrive forhold vedrørende fosfor, der anses som det vigtigste næringsstof for den økologiske tilstand i de fleste søer. Også aspekter vedrørende kvælstofs, jerns og siliciums kredsløb behandles. Begge de to vigtige næringsstoffer - fosfor og kvælstof - ophobes i organiske forbindelser i både sediment og vandfase, men til forskel fra kvælstof indgår fosfor også i en række kemiske bindinger i uorganisk form i især sedimentet.

Beskrivelsen af fosfordynamikken omfatter blandt andet sedimentforhold og de mange bindingsformer af fosfor i sedimentet, hvoraf ikke mindst bindingen til jernforbindelser er vigtige for det meget dynamiske samspil mellem sediment og vand ved ændrede iltforhold i overfladesedimentet. Endvidere beskrives de mange mulige mekanismer bag udvekslingen af fosfor mellem vandfase og sediment, omfattende mekanismer som resuspension, ændringer i redoxforhold og indflydelsen fra bentiske primærproducenter. Omfanget af denne udveksling kan i flere sammenhænge være afgørende for søernes vandkvalitet – ikke mindst i kraft af den interne fosforbelastning, der ses i mange søer efter en reduceret ekstern fosforbelastning, og som kan bidrage til at fastholde søer i den uklare tilstand.

Den sæsonmæssige variation i vandfasens indhold af næringsstoffer behandles ligeledes, herunder også indsvingningsforløbet efter reduceret ekstern tilførsel af næringsstoffer, typisk opnået via mindsket spildevandsbelastning. Under indsvingningsforløbet mod mindre næringsrige forhold reduceres betydningen af den interne belastning i første omgang ved, at varigheden af negativ fosfortilbageholdelse om sommeren mindskes. I sedimentet kan der ske en frigivelse af fosfor ophobet i 20-25 cm's dybde, og sedimentet kan på den måde være en meget væsentlig fosforkilde i mere end 20 år, selvom det mest almindelige synes at være op til 10-15 år.

I afhandlingen indgår også artikler, der behandler forvaltningsmæssige aspekter såsom restaurering af søer ved anvendelsen af fysiske, kemiske og biologiske principper samt problemstillinger i forbindelse med implementeringen af EU's Vandrammedirektiv. Blandt restaureringsmetoderne er hovedvægten lagt på de biologiske metoder, herunder indgreb i fiskebestanden. Det er den langt hyppigst anvendte metode i Danmark og er gennem de sidste 10-20 år gennemført i omkring 50 søer. Her beskrives bestræbelser og forudsætninger for at opnå permanente klarvandede forhold, og flere artikler beskriver, hvordan skiftet fra uklare til klarvandede forhold i lav-

vandede søer også fører til en markant øget evne til at tilbageholde såvel fosfor som kvælstof i de lavvandede søer. Blandt de kemiske metoder beskrives blandt andet anvendelsen af nitrat til at oxidere sedimentoverfladen for derigennem at øge den redoxfølsomme binding af fosfor til jernforbindelser.

Vandrammedirektivet fordrer, at der opnås en god økologisk tilstand senest i år 2015, men implementeringen rummer en række vanskeligheder, som illustreret ved en analyse af data fra omkring 700 danske søer. Blandt vanskelighederne er fastsættelse af den menneskelige upåvirkede tilstand, hvorudfra 5 økologiske klasser med forskellig kvalitet skal fastlægges og den økologiske kvalitet defineres i velafgrænsede klasser.

1 Nutrients and Danish lakes

Numbering more than 120,000, lakes and ponds constitute an important feature of the Danish landscape. Concurrently with the human exploitation of nature and the increasing expansion of urban communities, their role have changed from being primarily an important food resource to representing important recreational and natural values (*Mathiesen, 1969; Sand-Jensen, 2001; Hofmeister, 2004*). The biological development of lakes has been documented through palaeolimnological investigations and these show that human activities and changed land use practices in lake catchments have produced considerable changes in environmental state (*Jeppesen et al., 2001; Bradshaw, 2001; Johansson et al., 2005*). The most significant changes have, however, taken place during the past approx. 100 years, where increased population density and intensified agriculture have led to enhanced nutrient input and deteriorated water quality in many lakes (*Bradshaw, 2001; Amsinck et al., 2003; Søndergaard et al., 2003a*).

During the past 20-30 years various regional and national action plans have been implemented at a cost of several billion Danish kroner, with particular emphasis on improved wastewater treatment (*Jeppesen et al., 1999a*). At the national level various action plans on the aquatic environment have been introduced, latest Action Plan III, adopted in spring 2004, with primary focus on agricultural runoff control (*Søndergaard et al., 2003b; Nielsen et al., 2005a*). Also at European level within the framework of the European Union initiatives have been launched, the most important being the Habitats Directive and the Water Framework Directive (WFD) of which the latter is to ensure and improve the aquatic environment. In the future the WFD will have a great impact on the state and management of the Danish aquatic environment (see section 5.3). Thus, obtaining a clear understanding of the role of internal nutrient dynamics in lakes is not only interesting from a scientific point of view, it also has extensive, social and financial implications.

1.1 The importance of nutrients

The reason why nutrients play such a decisive role for the environmental state of lakes is the fact that the primary production of lakes is strongly limited by nutrient availability. Increased nutrient input therefore leads to enhanced lake productivity, with cascading effects on the remaining trophic levels and the interactions between these (*Jeppesen, 1998*). Thus, in many lakes, during the 20th century the input of phosphorus and nitrogen from urban communities and agriculture triggered a shift from a clearwater state with submerged macrophytes as the most significant primary producer to a turbid state now with phytoplankton as the dominant primary producer (*Körner, 2002; Jeppesen et al., 2005a*). Simultaneously, marked alterations occurred in fish and zooplankton structure, which again affected the grazing control on phytoplankton (*Jeppesen et al., 1997a, 2000*).

Particularly phosphorus is important among the many nutrients that apart from carbon contribute to plant primary production and, thus form the basis for the other components of the food chain. This is because phosphorus often functions as the limiting nutrient and thereby determines phytoplankton abundance (*Dillon & Rigler, 1974*). Therefore, the availability of phosphorus is frequently considered as being the single most important factor for the overall environmental state of lakes. The importance of phosphorus can be illustrated by the marked changes occurring along a phosphorus gradient for a number of the most central groups of organisms (Fig. 1.1).

Another significant nutrient, nitrogen, typically occurs in concentrations much higher than those of phosphorus and despite that the demand by primary producers for nitrogen is higher than for phosphorus there will often be a nitrogen surplus. The stream inlet concentrations of total nitrogen (TN) to Danish lakes are, on average, 40 times higher (weight basis) than that of total phosphorus (TP) (*Jensen et al., 2004*). Due to the nitrogen removal in lakes, the TN:TP ratio of the lake water is lower (on average 19.5), but may vary substantially from lake to lake (Fig. 1.2). In aquatic systems nitrogen is considered potentially limiting if TN:TP is below ca. 9, while phosphorus is considered potentially limiting at a TN:TP ratio of approx. 22 (*Guildford et al., 2000*). As shown in Fig. 1.2, in most Danish lakes the levels of nitrogen and phosphorus often lie within the interval where both nitrogen and phosphorus may be limiting, the number of potentially phosphorus limited lakes being, however, much higher

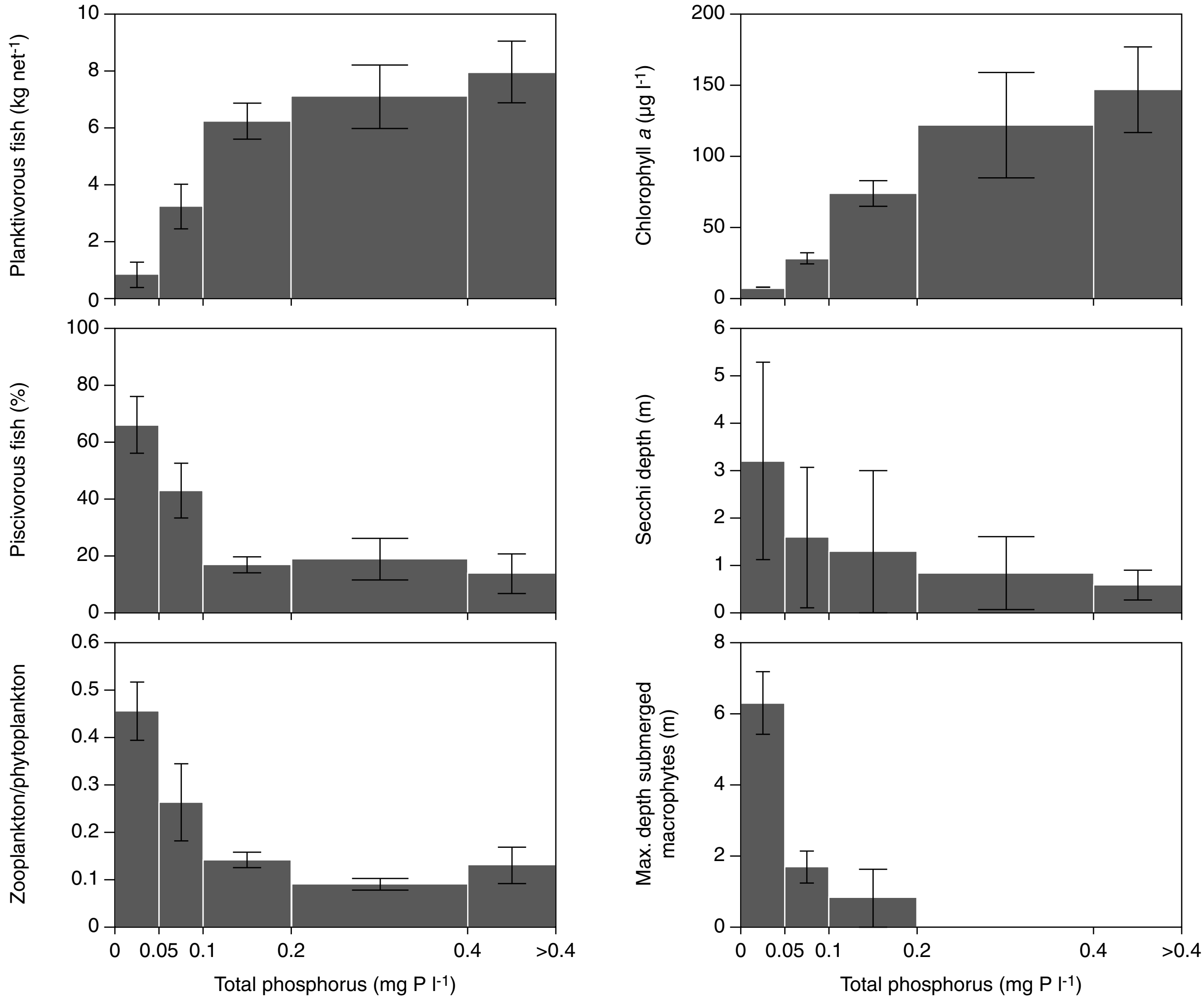

Figure 1.1 Impact of lake water phosphorus concentrations on a number of biological variables. Data from 65 Danish lakes. From Jeppesen *et al.* (1999a) and Søndergaard *et al.* (1999).

than that of potentially nitrogen limited lakes. Also, new available nitrogen may, contrary to phosphorus, be formed in the lakes via cyanobacterial fixation of N_2, although several investigations indicate that this normally constitutes only a minor fraction of the total nitrogen supply (*Jeppesen et al., 1998; Ferber et al., 2004*).

Nitrogen is recognised to play an important role in some lake ecosystems (*Moss 2001; Maberly et al., 2002*). This is true not least for lakes having a small catchment and a relatively modest nitrogen input and where nitrogen leaves the system through denitrification, while phosphorus remains (*James et al., 2002*). A Danish example of this is Lake Esrom, which is nitrogen limited for prolonged periods during the growth seasons (*Brodersen et al., 2001; Jónasson, 2003*). However, the concentration of nitrogen is often strongly correlated with the phosphorus level, implying that differentiation between the impact of the two substances may be difficult within an empirical context (Fig. 1.2).

Recent Danish results and analyses suggest that the significance of nitrogen is stronger than hitherto anticipated (*Jeppesen et al., 2005b*). In a number of enclosure experiments Gonzales Sagrario *et al.* (2005) demonstrated that at nitrogen concentrations above 1.2-2 mg N l^{-1} and phosphorus concentrations exceeding 0.1-0.2 mg P l^{-1}, the risk rose of a shift to turbid conditions. At high phosphorus concentrations but nitrogen concentrations below 1.2 mg N l^{-1}, almost all enclosures were clear and hosted submerged macrophytes. Also empirical analyses have shown that submerged macrophytes tend to disappear at nitrogen concentrations above 1-2 mg N l^{-1} (Fig. 1.3). The mechanisms behind the impact of nitrogen on submerged macrophytes have not yet been fully clarified, but nitrogen limitation of phytoplankton

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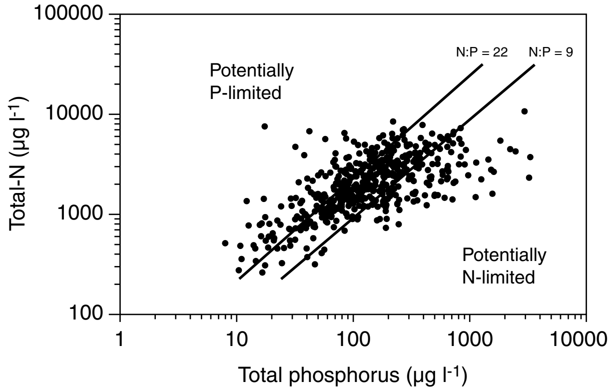


Figure 1.2 The relationship (weight basis) between lake water concentrations of total phosphorus and total nitrogen. Data from 487 Danish lakes (annual averages).

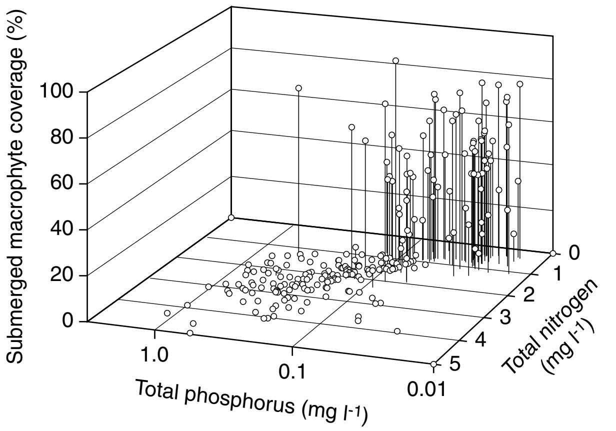


Figure 1.3 Coverage of submerged macrophytes relative to total phosphorus and total nitrogen concentrations. Data from 44 Danish lakes (246 lake years). From Jeppesen *et al.* (2005b).

and epiphytic algae is probably a key influencing factor. Supporting this supposition, Moss (2001) demonstrated that the species richness of plants declines with increasing nitrogen concentrations. Likewise, a high nitrate content favours the development of filamentous green algae that by shading may impede the growth of submerged macrophytes (*Irfanullah & Moss, 2004*).

Several other nutrients play an important role in the nutrient cycle of lakes. One of these is silicate, being an element of the skeleton of diatoms. Diatoms normally account for a major proportion of the phytoplanktonic primary production in lakes, especially during spring. The availability of silicate has not changed in consequence of human activities in the same way as the nitrogen and phosphorus input, but it may have changed, however, due to eutrophication. An example is Lake Michigan where decreased phosphorus loading has led to increased silicate concentrations due to declining abundance of diatoms and reduced silicate uptake (*Barbiero et al., 2002*). Investigations in Danish lakes have shown that also silicate concentrations respond to reduced eutrophication, but negatively so, probably because the sediment release declines at decreasing sedimentation and turnover of organic matter /1/. Finally, reduced phosphorus loading and improved Secchi depth result in increased benthic production, particularly in shallow lakes (*Liboriussen & Jeppesen, 2003*). This augments the uptake of silicate at the sediment surface, which may again lead to decreased silicate levels in the water phase (*Phillips et al., 2005*).

Iron only constitutes a minor proportion of the build-up of organic matter but indirectly has a strong influence on the nutrient turnover in lakes. This is due to the formation of compounds, including especially iron hydroxids, which are important for the inorganic binding of phosphorus in lakes, not least in the sediment's phosphorus pools. Retention and availability of phosphorus are therefore closely linked with the capacity of iron to release or take up phosphorus (see section 2.2).

1.2 Danish lake types

The morphology and water chemistry of Danish lakes differ widely (Table 1.1). Many of these differences have a significant impact on the nutrient dynamics and environmental state of the lakes. Water depth is one of the important morphological features (*Jeppesen et al., 1997a*).

Shallow versus deep lakes

Danish lakes are generally relatively shallow. Only 10% of the large-sized Danish lakes have a mean depth exceeding 5 m (Table 1.1). The shallow conditions imply that most lakes are fully mixed and do not display the summer stratification characteristic of deep and wind-protected lakes (*Wetzel, 2001*). These factors greatly influence the interactions between the sediment and the water phase. In deep lakes, the nutrients potentially released from the sediment are accumulated in the often oxygen-poor hypolimnion during summer and thus become difficult to access for primary producers. In contrast, in shallow lakes the well-mixed conditions create an immediate interaction between the sediment and the photic zone's pools of nutrients and primary producers during the

Table 1.1. Characteristics of Danish lakes based on data from approx. 800 lakes, most with a size larger than 1 ha.

	Characteristics	Min-Max	Median value
Water depth	Shallow, only approx. 10% with a mean depth above 5 m	0 – 38 m	Mean depth 2.9 m
Size	Small, of the 600 Danish lakes larger than 5 hectares, only 6 are larger than 10 km^2	0 – 40 km^2	9 hectares
Retention time	Varies widely from lake to lake, but most larger lakes have a relatively short retention time	few days – many years	0.4 year
Alkalinity	Most are alkaline, only 11% with an alkalinity below 0.2 meq l^{-1}, especially lakes in non-moraine landscapes as in Western Jutland, are more acid	-0,1 – 5,6 meq l^{-1}	2.1 meq l^{-1}
Colour	Most large-sized Danish lakes are not alkaline, but most small-sized lakes are more or less brown-coloured	< 10 – >500 mg Pt l^{-1}	unknown
Secchi depth	Turbid water due to high nutrient input, 75% have a Secchi depth below 1.5 m	<0.2 – 8 m	0.9 m
Phosphorus conc.	Nutrient-rich, 75% of Danish lakes have a phosphorus concentration above 80 µg P l^{-1}	5 – >1000 µg P l^{-1}	150 µg P l^{-1}
Water quality	Only 1/3 of Danish lakes fulfil the water quality objectives required by water managers		

whole growth season (*Nixdorf & Deneke, 1995*). Furthermore, the relatively small water volume relative to the size of the lake bottom in shallow lakes means that a given release rate of, for instance, phosphorus will lead to a more rapid increase in the concentrations of the water phase than in deep lakes with their greater water volumes /4/. The interactions between the sediment, water phase and biological components are thus closely coupled in shallow lakes.

Large versus small lakes

Lake area is another important morphological feature. In Denmark small lakes dominate in size; only 600 of the 120,000 Danish lakes and ponds larger than 100 m^2 are larger than 5 hectares, and only 6 larger than 1,000 hectares (Table 1.2). Despite that, the most intensive lake investigations have all been conducted in large lakes, the knowledge of and data on lakes smaller than 1-5 hectares being much less comprehensive. That this is so will naturally be reflected in many of the analyses presented in this dissertation.

Despite the widely differing sizes of Danish lakes, Lake Arresø with its 4,000 hectares being the largest, there are no indications of major inter-lake differences as long as the size is larger than 1-10 hectares (/3/; *Jensen et al., 2001; Søndergaard et al., 2002*). One difference does, however, exist, namely as to the species numbers of submerged macrophytes and fish, which continues to increase with size also in the large lakes. This is in accordance with the prevailing perception that increasing size results in more niches and, with it, the potential occurrence of more species (*Tonn & Magnusson. 1982; Dodson et al., 2000; Oertli et al., 2002*). By way of example, the average number of submerged macrophyte species in alkaline, nutrient-poor lakes increases from approximately three in lakes sized 1 hectare to around fifteen in lakes sized 100 hectares (/3/; *Søndergaard et al., 2003c*).

Table 1.2 Size distribution of Danish lakes > 100 m^2.

Size	Number	Number (% of total number)	Area (km^2)	Area (% of total lake area)
100-1,000 m^2	87,000	73.0	34	5.8
1,000-10,000 m^2	29,000	24.3	78	13.3
1-10 ha	2,900	2.4	70	11.9
10-100 ha	260	0.2	72	12.3
100-1,000 ha	70	< 0.1	214	36.5
> 1,000 ha	6	< 0.1	119	20.3

Small lakes and ponds less than approx. 10 hectares differ from larger lakes in several respects. An essential difference is the absence of fish from the smallest lakes /3/. The majority of lakes smaller than 1,000 m^2 are fishless, and in an investigation of 83 Danish lakes sized between 25 and 3,400 m^2 92% were devoid of fish (*Henriksen, 2000*). The occurrence or non-occurrence of fish depends on the capacity of each individual species to survive under poor oxygen conditions in connection with ice coverage in winter or during summer draughts, but also the connection with other water areas is important. Thus, small lakes and ponds connected with other water areas will often contain fish due to continuous immigration of new fish /3/. Also plant diversity is impacted, and isolated lakes are therefore usually more species-poor (*Møller & Rørdam, 1985; Linton & Goulder, 2003*). As to other groups of organisms, for instance zooplankton and phytoplankton, species number does not depend on lake size /3/. Lakes – including the small-sized ones – are thus thought to play an important role in the upkeep of high natural biodiversity (*Williams et al., 2003*).

Another significant difference between small and large lakes is the fact that the water of small-sized lakes tends to be more brown coloured that that of large-sized lakes (Fig. 1.4). This is due to the relatively greater contact with the surrounding terrestrial environment, combined with an often poor water exchange and a smaller water volume. The higher content of humus substances does not only influence water turbidity, which, among other parameters, impacts the growth conditions of submerged macrophytes and phytoplankton (*Havens, 2003a,b*). More humus substances also imply that the nutrient turnover of the food chain to a higher extent is based on the allocthonic input of organic matter than on internal primary production only (*Cole et al., 2002; Karlsson et al., 2003)*. By way of example and in accordance with this, in their investigation of two American lakes Pace *et al.* (2004) found that as much as 50% of the particulate organic carbon and carbon bound in zooplankton was of terrestrial origin.

As to nutrient concentrations there is no major difference between large and small Danish lakes, apart from the circumstance that concentrations seem more variable in the smallest lakes (Fig. 1.4). However, recent monitoring data from Danish lakes selected randomly for sampling indicate that small lakes are generally richer in nutrients than are larger lakes (*Lauridsen et al., 2005*). This means that nutrient concentrations are generally high in small-sized Danish lakes even though these, in contrast to large lakes, are not directly connected with the surrounding catchment via stream inlets. This is indicative of a strong effect of the catchment, which in Denmark most often consists of cultivated fields. Thus, as lake size decreases, the extent of cultivated catchment areas gradually increases. By way of example, on the island of Funen 30% of the catchments within a 100 m distance of lakes consist of cultivated fields for lakes >5 ha compared to 65% for lakes <0.1 ha (/3/; Fig. 1.5).

The high phosphorus content does not have the same effect on the concentration of chlorophyll in large- and small-sized lakes, the chlorophyll *a*:TP ratio generally being lower in the small-sized lakes (Fig. 1.6). There may be several explanations of this, including the generally poor light conditions in small lakes due to high humus content, or that nitrogen is potentially more limiting in small lakes because of the high water inflow. Another reason may be that the abundance of phytoplankton in the small-sized lakes is primarily governed by grazing from zooplankton. However, this seems not to be an explanatory variable as the biomass of zooplankton does not increase significantly with reduced lake size /3/. The reason for an unchanged zooplankton density despite a small or even

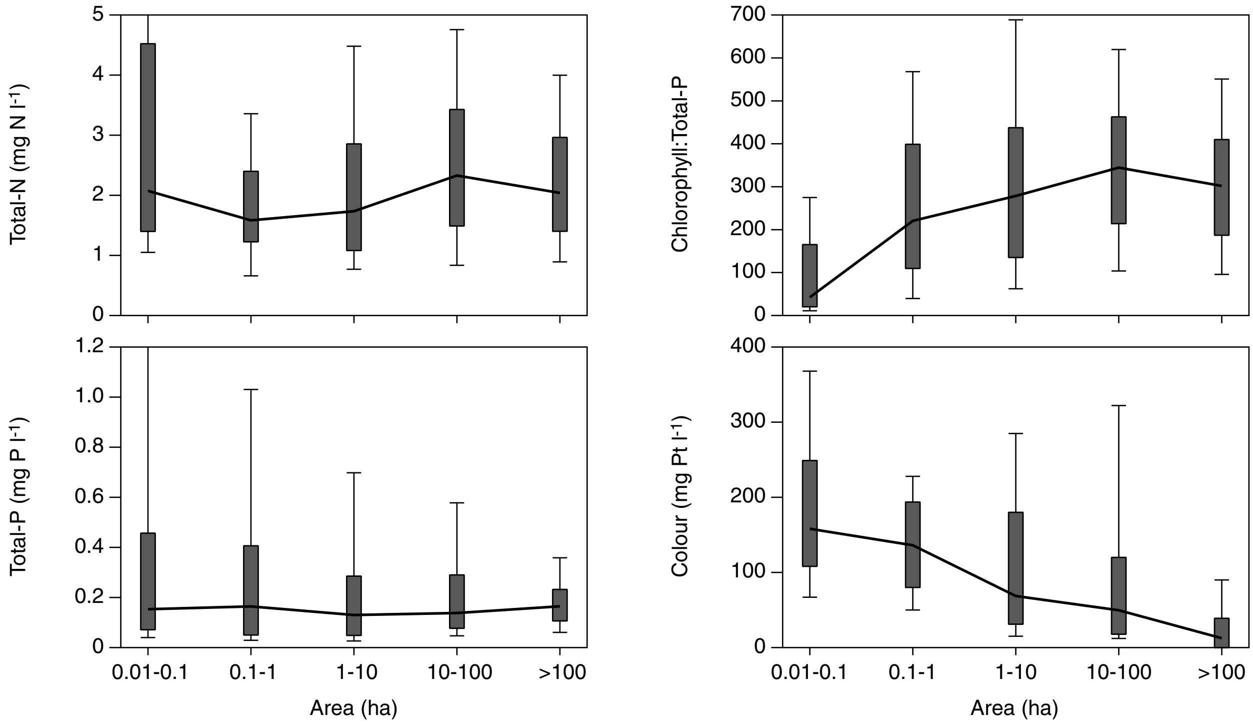


Figure 1.4 Concentrations of nutrients, chlorophyll *a* and colour in Danish lakes of varying size. Each box shows 25 and 75 % quartiles, upper and lower lines indicating 10 and 90 % percentiles. Mean values are connected by lines. Data from 187-777 Danish lakes. From /3/.

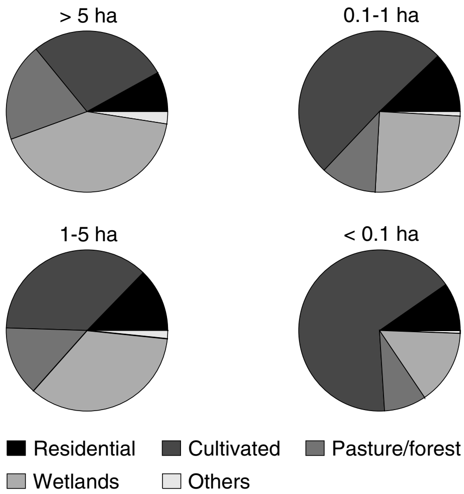


Figure 1.5 Distribution of catchment types at a distance of 100 m for 11,000 Danish lakes of varying size on the island of Funen. From /3/.

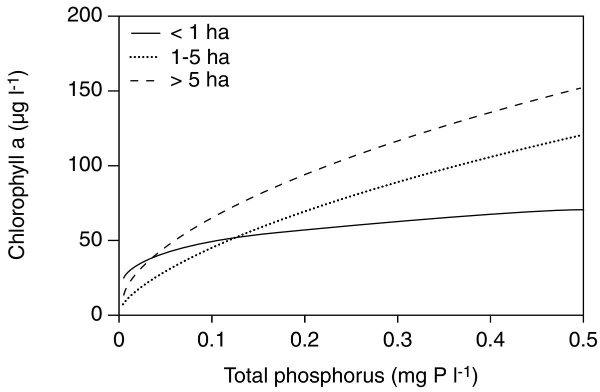


Figure 1.6 Relationships between concentrations of total phosphorus and chlorophyll *a* in three sizes of lakes: < 1 hectare (number of lakes = 41), 1-5 hectares (number of lakes = 363) and > 5 hectares (number of lakes = 236). From /3/.

absent fish stock may possibly be that invertebrate predators, such as *Chaoborus* or *Notonecta (Arnér et al., 1998*), take over the role of fish and thereby have a deleterious effect on the population of large-sized zooplankton. Finally, also abiotic factors, such as pH and temperature, may have a strong influence on the occurrence of large-sized zooplankton (*Steiner, 2004*).

Other lake types

Various other factors influence the environmental state of lakes and their reactions to a changed nutrient input, one of these being the alkalinity. The alkaline soil conditions in the major part of Denmark mean that most Danish lakes are alkaline too. Of the large-sized Danish lakes approx. 90% have an alkalinity above 0.2 meq l^{-1}, and 50% an alkalinity exceeding 2.1 meq l^{-1}. Differences in alkalinity influence among other parameters the species composition of submerged macrophytes; thus, isoetids are typically found in low-alkaline lakes due to their capacity to exploit CO_2 as a carbon source (*Vestergaard & Sand-Jensen, 2000*). Also plankton, invertebrates and fish are affected by pH levels, particularly if low, several snail and fish species disappearing at pH values below 5-6 (*Økland & Økland, 1980*).

Another important structuring element is salinity (*Moss, 1994; Jeppesen et al., 1994, 1997*). Brackish lakes are a very common lake type in Denmark, especially along the western coast of Jutland and along the Limfjord coast. Brackish lakes differ from freshwater lakes in several respects, as increased salinity causes considerable changes in the communities of zooplankton and invertebrate predators (*Jeppesen et al., 1994; Aaser et al., 1995; Søndergaard et al., 2000; Jacobsen et al., 2004*). Most brackish lakes in Denmark are, like the freshwater lakes, highly eutrophic, but salinity may be crucial for the internal nutrient dynamics as the sediment's capacity to bind phosphorus is influenced by the sulphur cycle via sulphate reduction and sulphide formation (see section 3.2). Thus, with increased salinity, decreased proportions of iron-bound phosphorus and TP are often observed in the sediment (*Paludan & Morris, 1999; Coelho et al., 2004*).

Depending on the specificity of the definition of lake types, minimum eleven different types of large-sized lakes are found in Denmark (*Søndergaard et al., 2003b*), the dominant type (ca. 50%) being alkaline-rich, uncoloured, freshwater lakes, and the second-largest group (25%) deep, alkaline-rich, uncoloured, freshwater lakes.

1.3 The environmental state and development of Danish lakes

Danish lakes, large as well as small, are predominantly nutrient-rich, which can mainly be ascribed to various types of anthropogenic impact. However, concurrently with the improved techniques employed at wastewater treatment plants, the nutrient input to many lakes has decreased during the past 20-30 years (*Kronvang et al., 2005*). Since the beginning of the 1990s the mean inlet concentration of phosphorus to the 27 freshwater lakes comprised by the Danish national lake monitoring programme has decreased from 0.18 to 0.09 mg P l^{-1} (/1/; *Jeppesen et al., 1999a; Jensen et al., 2004*), a well-documented example of this being Lake Fure where improved sewage treatment

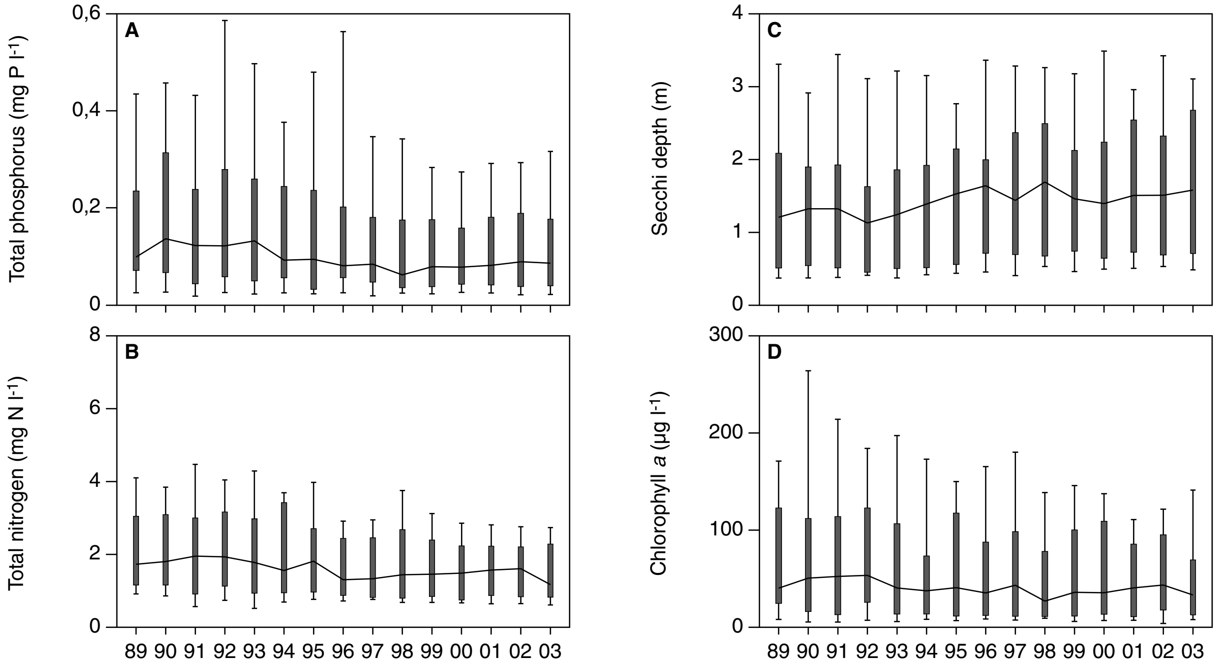


Figure 1.7 The development in Secchi depth, chlorophyll *a* and total phosphorus and total nitrogen concentrations in 27 freshwater monitoring lakes since 1989. From Jensen *et al.* (2004). Each box shows 25 and 75 % quartiles, while upper and lower lines represent 10 and 90 % percentiles. Median values are connected by lines.

and nutrient cut-off in the mid-1970s led to an approx. 90% reduction of the phosphorus load (*Jensen et al., 1997*). Yet, to many lakes the nutrient loading is still too high for stable clearwater conditions to occur (*Søndergaard et al., 2003b*).

Generally, lake water phosphorus concentrations have declined in recent years, but not as much as might have been expected from the relationships normally existing between inlet and in-lake concentrations. Correspondingly, only relatively small improvements of water quality have been recorded. For instance, summer Secchi depth in the Danish monitoring lakes only increased from 1.4 to 1.7 m from the beginning of the 1990s to 2003, while the chlorophyll *a* content decreased from 77 to 50 µg l^{-1} (Fig. 1.7). During the same period TP concentrations declined from 0.17 to 0.11 mg P l^{-1} (*Jensen et al., 2004*). Today, only ca. one third of the Danish lakes meet the environmental objectives set in the regional plans elaborated by the environmental departments of the Danish counties, and this is despite the huge investments made in improved wastewater treatment (*Søndergaard et al., 1999*).

The explanation of the delayed response is the accumulation of large phosphorus pools in the sediment during the high loading period. After a reduction of external loading it will take a while before these pools reach a new chemical equilibrium with the reduced inlet concentration. This means that most Danish lakes are currently undergoing a transitional period where there is no equilibrium between inlet and in-lake nutrient concentrations. The release of phosphorus from the sediment – termed internal phosphorus loading – may contribute to enhancing the lake water concentrations of phosphorus for several decades after a loading reduction (see section 2.4).

A delayed response to decreased external phosphorus loading may also result from biological parameters such as the maintenance of a high predation pressure on the zooplankton by the existing fish stock, which reduces the possibility of top-down control of the phytoplankton (see section 4.3). Finally, also immigration and re-establishment of an extensive coverage of submerged macrophytes, which is pivotal for stabilising the clearwater state, may be delayed due to spreading barriers or herbivory by birds or fish (*Lauridsen et al., 1993; Søndergaard et al., 1996; Marklund et al., 2002; Körner & Dugdale, 2003*; see also section 4.3).

Overall, Danish lakes are nutrient-rich either due to phosphorus release from the sediment and/or a still high external input. Most Danish lakes are consequently turbid with a high biomass of phytoplankton and poor coverage or even absence of submerged macrophytes. The fish stock is dominated by zooplanktivores – particularly roach (*Rutilus rutilus*) and bream (*Abramis brama*). The share of predatory fish, especially perch (*Perca fluviatilis*) and pike (*Esox lucius*), is small, and so is the number of large-sized zooplankton species, most notably *Daphnia*, which again diminishes the likelihood of grazing control of the phytoplankton (*Jeppesen et al., 1999b)*. This means that phytoplankton abundance, besides light and temperature, is primarily regulated by the availability of nutrients.

2 Retention of phosphorus and nitrogen

When a lake is enriched by nutrients such as phosphorus and nitrogen, a certain portion of these will - in a state of equilibrium - be retained, i.e. the nutrient flux out of the lake is smaller than the input flux (*Vollenweider, 1976*). Phosphorus retention occurs via sedimentation of particulate-bound forms or via uptake and incorporation of dissolved phosphate by plants and subsequent sedimentation. There may also be an exchange of dissolved inorganic substances between the sediment-water interface, but the normal transport route is from sediment to water phase, since the nutrient concentrations are higher in the sediment pore water than in the lake water due to mineralisation (see section 3.4). In productive lakes, the sedimentation of phosphorus may rise in connection with the depositing of calcium carbonate (*House et al., 1986; Driscoll et al., 1993; Golterman, 1995; Hartley et al., 1997*). In alkaline lakes, pelagic calcite precipitation has been suggested as being one of the mechanisms stabilising the clear water state (*Kasprzak et al., 2003*).

External input of iron and formation of iron hydroxides may result in adsorption or binding of phosphorus and its sedimentation in an inorganic form (*Hongve, 1997; Prepas & Burke, 1997; McAuliffe et al., 1998; Kleeberg & Schubert, 2000*). This means that input and retention of iron have a substantial influence on phosphorus retention. Iron-bound phosphorus constitutes an essential and dynamic part of the sediment phosphorus pool due to the redox dependency of the phosphorus binding to iron (see section 3.3).

Nitrogen retention in lakes does not only occur as incorporation in sedimenting organic matter, but also, and largely so, via denitrification, where nitrate is exploited for the bacterial turnover of organic matter (Wetzel, 2001). Thereby nitrate converts to ammonium or to free nitrogen (N_2) that may diffuse into the water phase and the atmosphere, and thus is lost from the system. For Danish lakes, calculations show that on average 77% of nitrogen retention can be ascribed to denitrification (Jensen et al., 1990). In eutrophic Lake Søbygård ca. 90% of the nitrogen is estimated to be removed via denitrification, while 10% remains permanently buried in the sediment (Jensen *et al.*, 1992a).

The relative retention of both phosphorus and nitrogen depends strongly on the duration of the hydraulic retention time in the lake. The longer the retention time, the higher the loss percentage of the added phosphorus or nitrogen during its passage through the lake. As for phosphorus, the connection between retention time and phosphorus retention has been modelled by for instance Vollenweider (1976): $P_{lake} = P_{inlet}/(1+ tw^{0.5})$, where P_{lake} is the lake concentration, P_{inlet} the inlet concentration, and tw the water retention time (years) in the lake. A hydraulic retention time of one year thus means that the annual average lake concentration in a state of equilibrium will be approx. 50% of the inlet concentration. As for the lake concentration of nitrogen (N_{lake}), based on data from 16 Danish, shallow, eutrophic lakes, the relationship has been described as a function of the inlet concentration (N_{inlet}) and retention time: $N_{lake} = 0.32^* N_{inlet}{}^*tw$, $r^2 = 0.81$ (*Windolf et al., 1996*). As an average for 69 Danish lakes, calculations show that 43% of the added nitrogen is removed (*Jensen et al., 1990*).

For phosphorus, the fact that models as the above are based on lakes in equilibrium means that they do not adequately describe the situation arising after a loading reduction followed by high internal phosphorus loading from the sediment /15/. As to nitrogen this is less critical, partly due to the smaller reduction of the nitrogen input, and partly because nitrogen does not exhibit the same delayed response as phosphorus (Jeppesen et al., 2005c). The latter can probably be attributed to the circumstance that nitrogen primarily occurs bound in sedimental organic matter, which first has to be decomposed, and not as phosphorus bound and accumulated in inorganic forms.

Moreover, most models normally describe annual averages and not phosphorus or nitrogen concentrations throughout the season. A model describing the seasonal level of nitrogen in Danish lakes was developed by Windolf *et al.* (1996), and recently a Danish model has been established describing seasonal phosphorus concentrations using data on inlet concentrations, the sediment phosphorus pool and temperature (/15/). This phosphorus model is based on detailed data from 16 lakes included in the Danish lake monitoring programme during 1989-1996 and has been tested using data from 1997-2003 (/15/).

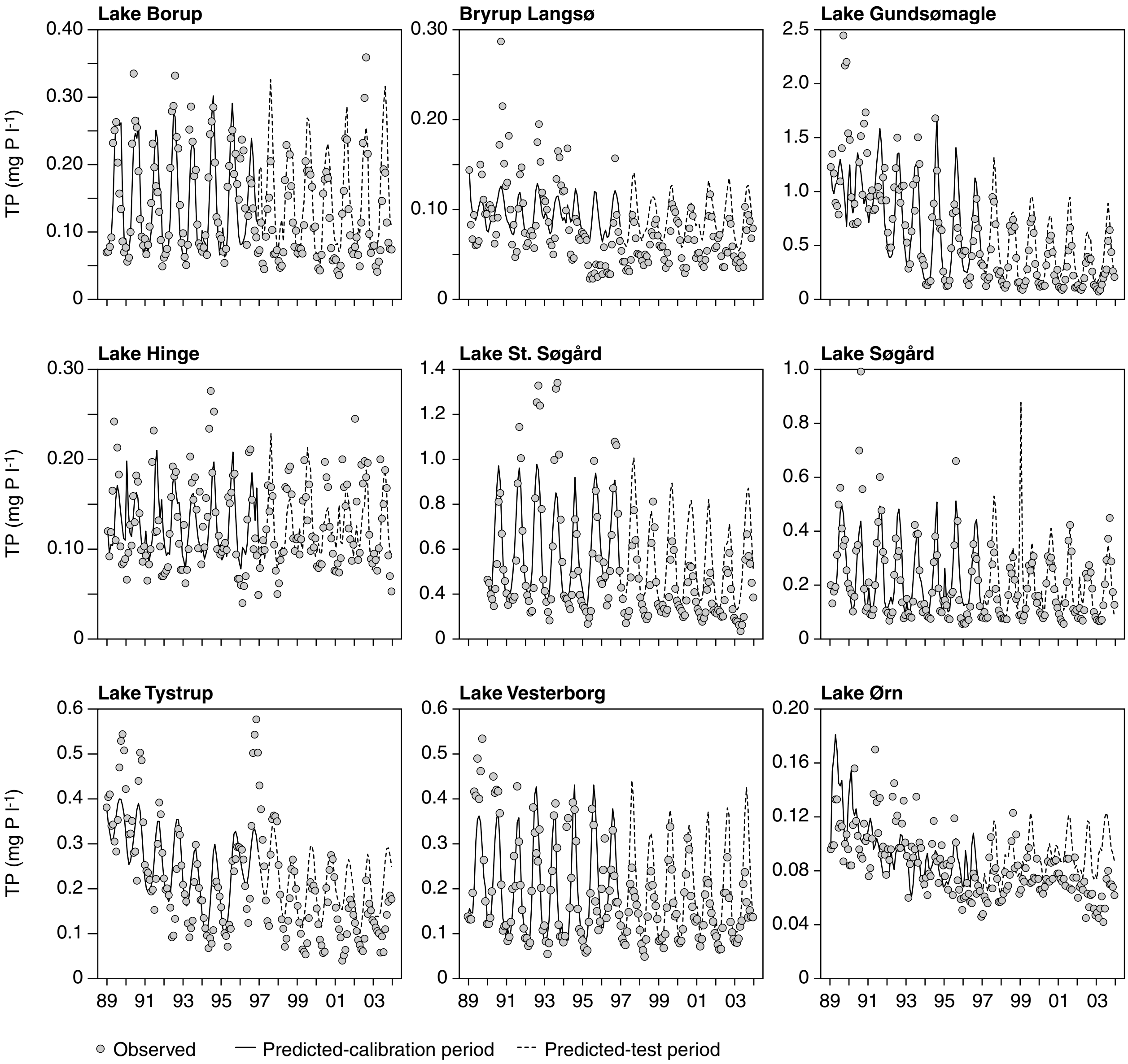


Figure 2.1 Observed and model-calculated concentrations of total phosphorus in 9 Danish monitoring lakes from 1989-2003. Calculated values for the calibration period 1989-1996 and the test period are connected by full and broken lines, respectively. From /15/.

Including only few variables, it provides an adequate description of the seasonal phosphorus concentrations in the lake type on which it is based, i.e. shallow lakes being in the transitional phase after reduced external phosphorus loading and with strong sediment influence (Fig. 2.1). However, in the test the model tended to overestimate lake concentrations, suggesting a reduced sediment impact the longer the time after the loading reduction. Moreover, the model is not capable of estimating lake concentrations when a shift occurs from the clearwater to the turbid state. This is due to the substantial change in the sediment's capacity to retain phosphorus when a lake shifts from the turbid to the clearwater state (see section 2.2).

2.1 Accumulation of nutrients in the sediment

The lake sediment constitutes the ”lowest” part of the lake where organic matter and material added to the lake from the outside or produced within the lake are deposited and accumulated. In most lakes the sediment therefore ”grows” with time, and the lakes become more shallow and more or less overgrown with reeds or other vegetation. In nutrient-rich lakes this sediment growth may be up to several millimetres per year – and considerably more in deep accumulation areas or areas close to stream inlets where the sedimentation of incoming particles may be high. In wind-exposed and shallow areas, termed ”transport areas” (*Håkansson, 1986*), there is no net sedimentation of organic matter because the easily

Table 2.1 Physico-chemical characteristics in Danish surface sediments (0-2 cm depth). N = lake number. From Jensen *et al.* (1997).

	N	Mean	25%	Median	75%
Dry weight (%)	210	12.9	6.1	9.2	13.0
Loss on ignition (%)	211	31.0	20.8	28.0	36.9
Calcium (mg Ca g^{-1} dw)	141	87	12	59	153
Iron (mg Fe g^{-1} dw)	145	26.9	11.0	17.8	33.0
Total phosphorus (mg P g^{-1} dw)	216	2.1	1.0	1.6	2.6
Total nitrogen (mg N g^{-1} dw)	204	14.8	10.0	13.2	18.3

resuspendable matter is constantly stirred and decomposed in the water phase or transported to more tranquil areas where sedimentation occurs. In some lakes these areas include most of the sediment surface. Danish examples are several of the shallow brackish lakes in the nature reserve of Vejlerne and Lake Ferring in Western Jutland (*Søndergaard et al., 2000*). In the latter, despite its highly nutrient-rich and productive lake water, the major part of the sediment consists of sand or hard bottom areas (Fig. 2.2). Only the wind-protected south-western part of the lake is soft and organically rich sediment accumulated. Thus, sediment conditions may vary significantly both within a lake and between lakes (Table 2.1). In shallow and wind-exposed lakes frequent resuspension may also result in an overall smaller accumulation of organic matter in the lake, as decomposition primarily occurs in the water phase. In less wind-exposed lakes, where sedimentation is permanent over most of the lake bottom, the variations seen in the large mid-lake area are typically more modest, and, correspondingly, the sediment content of nutrients is more uniform (*Jensen et al., 2004*).

Traditionally, there has been a tendency to observe and treat the water mass and the sediment as two more or less separate parts of a lake. However, often there is a very close interaction between these, not least in shallow lakes where conditions of mixing and contact between sediment and water are favourable. An example of the very dynamic interactions between the sediment and the water phase is illustrated in nutrient-rich Lake Søbygård by the marked increase in the phosphorus concentrations of the water phase that occurred in periods with poor sedimentation but continuously high phosphate release from the sediment (Fig. 2.3). In peri-

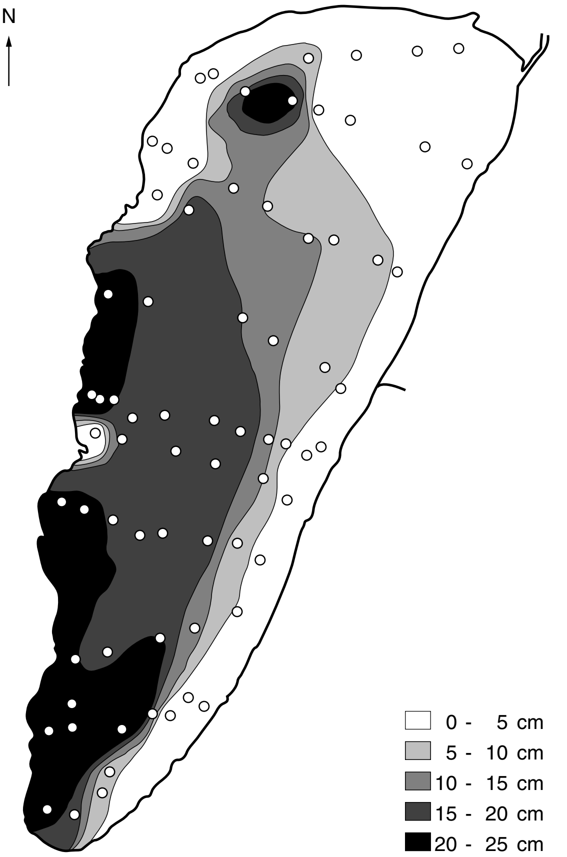


Figure 2.2 Sediment hardness in Lake Ferring measured as the penetration depth of a metal spear using a penetrometer. Measurement stations are indicated by circles. From Søndergaard *et al.* (2000).

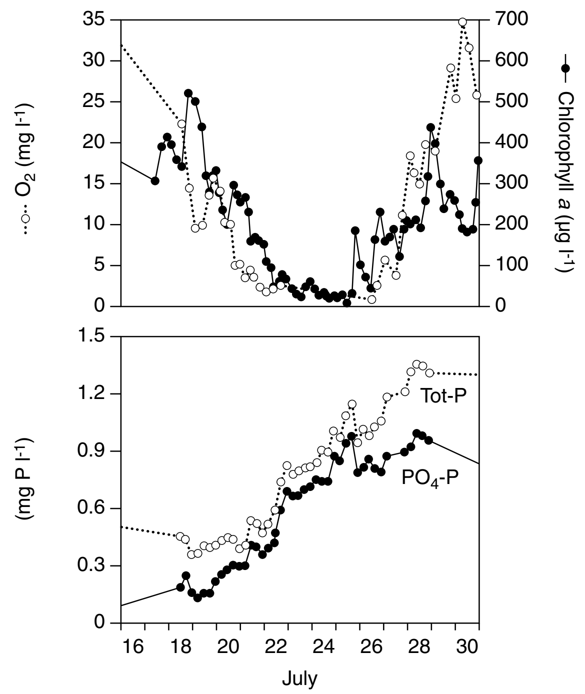


Figure 2.3 Changes in lake water oxygen, chlorophyll *a* and total phosphorus and orthophosphate concentrations in Lake Søbygård during a two-week period of phytoplankton population collapse in July 1985. From Søndergaard *et al.* (1990).

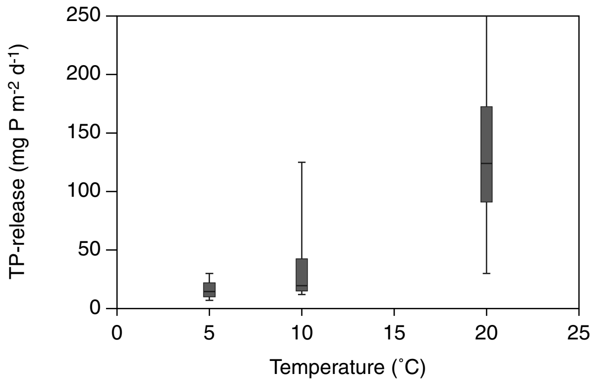


Figure 2.4 Boxplots showing the release of phosphorus from 8 sediment columns from Lake Søbygård incubated at 5, 10 and 20 °C. Each box shows 25 and 75 % quartiles, while upper and lower lines represent 10 and 90 % percentiles. Based on Søndergaard (1989).

ods with a marked decrease in phytoplankton biomass and poor sedimentation, the lake water's TP concentration rose from 0.37 to 1.33 mg P l^{-1} in only 10 days, corresponding to a net release from the sediment of 145 mg P m^{-2} d^{-1} (*Søndergaard et al., 1990*). Laboratory experiments with undisturbed sediment columns confirmed the occurrence of such high release rates (Fig. 2.4; *Søndergaard, 1989*). At typical summer temperatures, release rates under oxic incubation ranged between 100 and 200 mg P m^{-2} d^{-1}. Release particularly occurred from the sediment's loose- and iron-bound forms. Thus, under normal conditions, net changes in the phosphorus concentrations of shallow, nutrient-rich lakes are equal to the difference between two large contrasting fluxes: a downward (sedimentation) and an upward (sediment release) flux (Fig. 2.5).

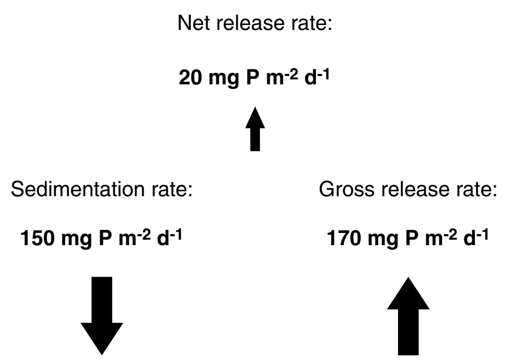


Figure 2.5 Sedimentation, gross and net release rates of phosphorus from Lake Søbygård. Based on Søndergaard *et al.* (1990).

Also nitrogen is accumulated in the sediment, but contrary to phosphorus, which is bound in many organic and chemical compounds, nitrogen is primarily bound in organic compounds. Therefore, a close relationship exists between sediment loss on ignition and the content of TN, whereas the correlation between TP and loss on ignition is much weaker (Fig. 2.6). Both the content of phosphorus and that of nitrogen vary substantially from lake to lake, whereas the levels of iron and calcium are more similar (Table 2.1).

2.2 Phosphorus in the sediment

Phosphorus is found in the sediment in both dissolved and a number of organically and inorganically bound forms (Fig. 2.7). Even though the concentration of dissolved phosphate is usually substantially higher in the pore water than in the water phase, it normally constitutes only a very small fraction of the sediment's TP pool, i.e. less than 1% (*Boström et al., 1982; Søndergaard, 1990*). Nevertheless, pore water phosphate is extremely important for the exchange of phosphate between the sediment and the water phase, which primarily occurs via dissolved forms. Pore water phosphate is, though, continuously in equilibrium with the inorganically bound fractions. Here, chemical and microbial processes play a pivotal role (/6/; *Löfgren & Ryding, 1985; Hupher et al., 1995*). The microbial decomposition of organic matter depends on the presence of electron acceptors, primarily oxygen in the surface sediment, but deeper down when oxygen is depleted these are replaced by nitrate, iron, manganese or sulphate (*Thomsen et al., 2004*). Pore water concentrations of phosphate may thus increase both as a result of the release of organically bound phosphorus during the decomposition of organic matter, and due to depletion of oxidised substances and the declining redox potential. The latter affects the binding of phosphate to inorganic compounds, including the binding of phosphorus to iron minerals.

The binding of phosphorus in sediments is in principle governed by four processes (*Jacobsen, 1978*): 1) transport of dissolved phosphate between the particulate components, 2) adsorption – desorption mechanisms, 3) chemical binding, and 4) uptake, accumulation and turnover in organisms. While chemical bindings are normally perceived as being independent of the surrounding dissolved forms, the physical adsorption on the surface of particles is in constant equilibrium with the dissolved concentrations. Both the adsorption and the chemical binding of phosphorus may comprise a number of different substances and compounds, of which the most important are iron, calcium, aluminium, mangane, clay particles and organic matter. The adsorption and chemical binding are besides concentrations also dependent on both pH and redox conditions, which again are mainly determined by the bacterial metabolism.

Phosphorus fractionation

Throughout time multiple methods have been developed for describing the different particulate forms of phosphorus found in lake sediments (*Hieltjes & Lijklema, 1980; Psenner et al., 1988; Paludan & Jensen, 1995*). Mostly chemical fractionation methods have been used, implying successive addition of different extraction media to sediment samples, each of which is expected to extract certain binding forms of phosphorus. Typically, first water or "soft" chemical reagents such as NH_4Cl are used, followed by stronger basic reagents such as NaOH and acid reagents such as HCl. The final procedure involves total destruction of the sample by loss on ignition to ensure inclusion of all phosphorus forms. Fractionation methods such as these, which are based on the method developed by Hieltjes & Lijklema (1980) and used in many Danish lake, presume loosely bound phosphorus (NH_4Cl-P) to be released first, followed by a pool of phosphate bound to surfaces of iron, aluminium and manganese, and possibly also clay (NaOH-P). Then come calcium- and magnesium-bound forms (HCl-P) and, finally, a residual fraction (Res-P) of heavily degradable organic phosphorus compounds (*Jensen & Andersen, 1992*). More extraction steps may be involved in order to define more specific binding types, for instance by separation of phosphorus bound to iron and to aluminium (*Paludan & Jensen, 1995*).

A joint characteristic of the many fractionation methods is that they cannot unambiguously and precisely determine which forms of phosphorus to extract. The individual phosphorus fractions are thus in principle only defined via the extraction process and the extraction media with which the sediment sample has been treated. Nevertheless, the extraction methods often yield reproducible results that allow a description of the sediment's phosphorus forms and an estimation of the dominant binding compounds in a given sediment (*Pettersson et al., 1988*).

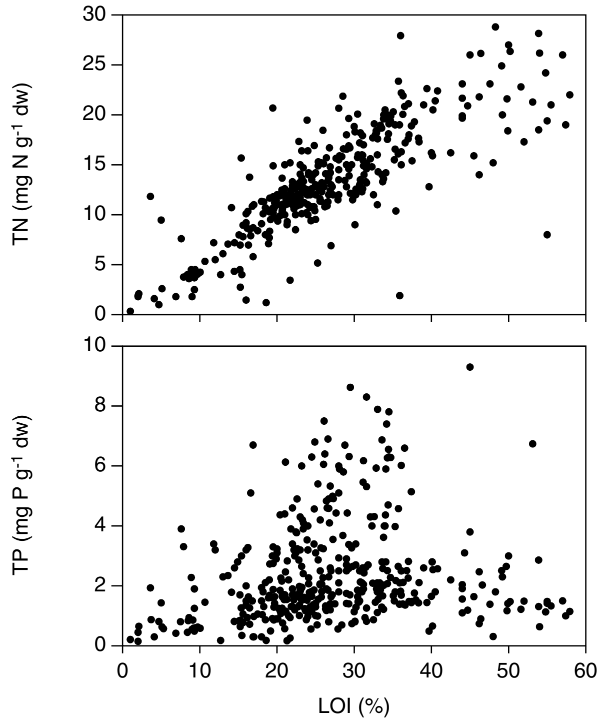
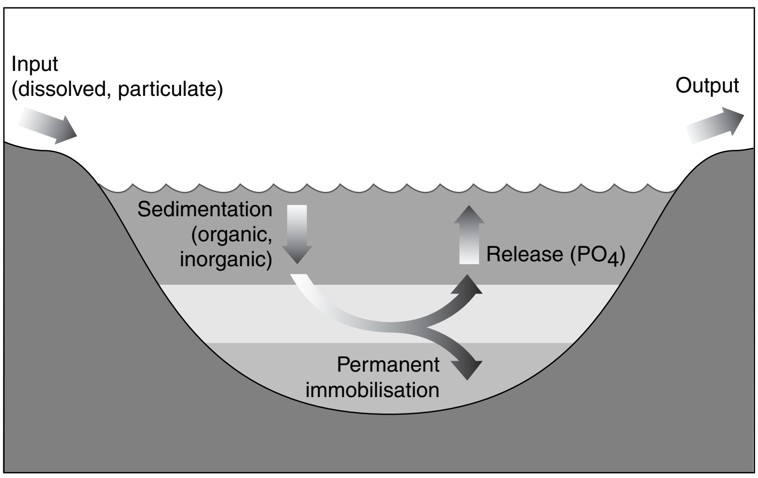


Figure 2.6 The relationship between Danish lake sediment contents of organic matter (loss on ignition/LOI) and total phosphorus (TP) and total nitrogen (TN) concentrations. By linear regression the relationship can be expressed as TN = 2.5 + 0.41*LOI, $p < 0.001$, r^2=0.67, n = 350 and TP = 1.6 + 0.027*LOI, p = 0.03, r^2= 0.03, n = 408.



P-forms in the sediment:

- Dissolved (PO_4, organic P)
- Particulate

Iron:	Fe (III) hydroxides, Fe (OOH), (ads.) Strengite, Fe PO_4 Vivianite, $Fe_3\ (PO_4)_2\ 8\ H_2O$
Alum:	Al $(OH)_3$ (ads.) Variscite, Al PO_4
Calcium:	Hydroxyapatite, $Ca_{10}\ (PO_4)_6\ OH_2$ Monetite, Ca H PO_4 Calcite (ads.)
Clay:	(ads.)
Organic:	"Labile" "Refractory"

Figure 2.7 Illustration of input, sedimentation, accumulation and release of different phosphorus types in shallow lakes. From /4/.

Although the total sediment pool of phosphorus is often very large and therefore changes only slowly for a sequence of years, the proportions of the different phosphorus fractions may not necessarily remain unchanged throughout the season. Loosely adsorbed compounds may vary considerably in the course of the year conditioned by the processes occurring in the water phase. By way of example, in Lake Søbygård the NH_4Cl-P fraction in the surface sediment ranged between 0.2 mg P g^{-1} dw in winter to > 2 mg P g^{-1} dw in summer, corresponding to 2.5-25% of the total phosphorus concentration (*Søndergaard, 1988*). The changes in compounds may owe to the impacts of, among other factors, photosynthetically increased pH, which may again trigger an increased release rate from the sediment. Thereby there may be a self-amplifying coupling between the processes in the water phase and the sediment: high phytoplankton production → high pH → large pool of loosely bound phosphorus → high phosphorus release from the sediment → high phytoplankton abundance, etc.

The different phosphorus pools of the sediments are normally sought described in the attempt to define a potentially mobile phosphorus pool that may be used to predict the role of the sediment as an internal phosphorus source and, with it, its impact on water quality. However, prior experience is not promising. The pools may easily be described, but well-defined relationships between the sediment phosphorus pools and the phosphorus fractions and the impact of these on the interactions between sediment and water remain to be identified (*Welch & Cooke, 1995; Søndergaard et al., 2001; Kisand & Nõges, 2003*). This inadequate coupling between the sediment types of phosphorus and actual measurements suggests that even if the sediment's potential for releasing or adsorbing phosphorus can be described, the actual release rates are mainly determined by other factors (see section 3).

Sediment data from 32 Danish lakes

An investigation of sediment from 32 Danish monitoring lakes shows a clear tendency to somewhat higher phosphorus concentrations in the upper 10-20 cm than in the deeper sediment layers (/11/; Fig. 2.8). The median value is around 2 mg P g^{-1} dw in the surface sediment compared to 1 mg P g^{-1} dw in layers deeper than 20-30 cm. Generally, the relative distribution of the individual phosphorus fractions does not vary significantly with depth. The largest fraction is organically bound phosphorus (Res-P), which on average constitutes around 40-45% of the total phosphorus pool (Fig. 2.8). The remaining part includes NaOH-P (30-35%) and HCl-P (approx. 20%), while NH_4Cl-P most frequently contributes only a few per cent. Remarkably, the investigations show considerable concentrations of NaOH-P even in the deepest sediment layers. As this fraction is presumed to primarily represent iron-bound phosphorus, this indicates that phosphorus can be permanently immobilised in the sediment in an iron-bound form. From a management perspective this suggests that addition of iron to lake sediments may be a useful restoration tool to increase the permanent deposition of phosphorus in the sediment (see also section 4.2). However, higher iron concentrations may also result in enhanced accumulation of phosphorus at high phosphorus loading, to be released in the event of a future loading reduction.

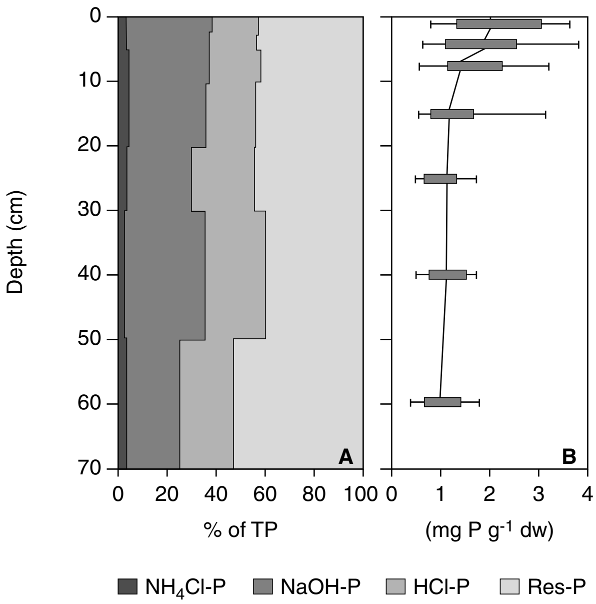


Figure 2.8 Depth profiles with main binding types (A) and average concentrations (B) of phosphorus in 32 Danish lakes. Each box (Fig. B) shows 25 and 75 % quartiles, while upper and lower lines represent 10 and 90 % percentiles. Median values are connected by lines. From /11/.

Higher phosphorus concentrations in the surface sediment are a common feature of almost all lakes, an obvious explanation being the increased external phosphorus loading to many lakes during the past decades. However, also the most nutrient-poor lakes, to which no significant changes in loading have occurred, exhibit markedly higher concentrations in the top sediment. These higher surface concentrations can almost always be attributed to an enhanced pool of organically bound phosphorus, suggesting that a large part of this pool is mobile and eventually will be mineralised and released (Fig. 2.9). The organically bound phosphorus pool in deeper sediment layers only constitutes around

Table 2.2 The relationships between the surface sediment content of total phosphorus (TP) and iron (Fe), external phosphorus loading (P_{ex} measured in g P m^{-2} yr^{-1}), sediment loss on ignition (LOI) and water mean depth (z). From /11/.

	Variable	r^2	p-value
=	$1.72 + 0.28{*}P_{ex}$	0.46	<0.0001
=	$1.50 + 0.031{*}Fe$	0.36	<0.0001
=	$0.92 + 0.23{*}P_{ex} + 0.031{*}Fe$	0.68	<0.0001
=	$-2.46 + 0.26{*}P_{ex} + 0.024{*}Fe + 0.090{*}LOI + 0.58{*}z$	0.91	<0.0001

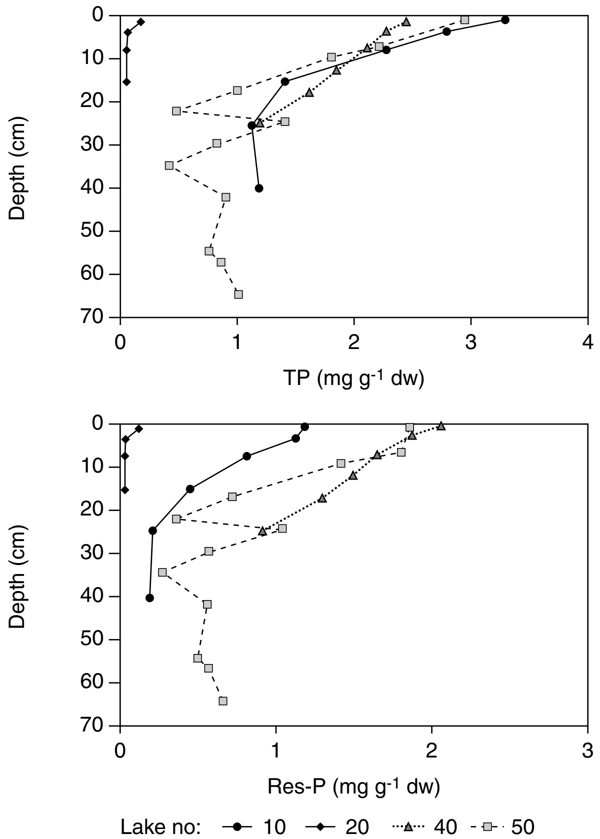


Figure 2.9 Mean concentrations of total phosphorus (TP, upper figure) and organically bound phosphorus (Res-P, lower figure) at various sediment depths in four nutrient-poor Danish lakes (Lake TP = 0.01 – 0.03 mg P l^{-l}). Lake 10: Lake Søby, Lake 20: Lake Holm, Lake 40: Lake Madum, Lake 50: Lake Nors. From /11/.

30-40% of that of the surface layer, which means that only approx. one-third of the organically bound phosphorus accumulated in the surface sediment will later be permanently buried within the sediment as organically bound phosphorus. The capacity of phosphorus to shift between dissolved and particulate forms, and thereby be transported, may also play a role. Thus, examples exist of increasing NaOH-P concentrations towards the surface – also in some of the not externally impacted lakes, which is suggestive of an upconcentration conditioned by increased binding capacity under oxidised conditions.

Besides type, also the concentrations of phosphorus are important when determining the role of the sediment, the decisive part being the significant capacity of iron to bind phosphorus under oxidised conditions. This is illustrated by the close connection between the sediment's iron and phosphorus concentrations. Together with the external phosphorus input and the sediment's iron concentrations, these two variables explain 68% of the variation occurring in the sediment concentrations of total phosphorus (Table 2.2). Moreover, multiple regression yields a statistically significant positive relation to loss on ignition and mean depth; implying that inclusion of all four variables can explain in total 91% of the variation in the surface sediment's content of total phosphorus (Table 2.2, Fig. 2.10). These relations have the user-oriented aspect that external phosphorus loading can, in principle, be estimated on the basis of lake mean depth, surface sediment concentrations of phosphorus and iron, and loss on ignition. Estimation of past external phosphorus loading is, however, only possible under the presumption that phosphorus is not mobile, and this is not necessarily the case (*Carignan & Flett, 1981*). Phosphorus profiles as that of Lake Søbygård, which for decades has shown relatively uniform depths for maximum concentrations of phosphorus, mostly consisting of inorganic forms (see section 2.4), suggest a certain conservatism, though.

Table 2.3 Regression coefficients at linear regression between total phosphorus (TP), iron (Fe), calcium (Ca), loss on ignition (LOI) in surface sediment, external phosphorus input (P_{ex}) and four phosphorus fractions. Number of lakes = 86-95. ns = not significant (P > 0.05). From/11/.

	TP	Fe	Ca	LOI	P_{ex}
NH_4Cl-P	ns	-0.10	ns	0.23	ns
NaOH-P	0.80	0.46	- 0.08	ns	0.25
HCl-P	0.35	0.12	ns	ns	0.68
Res-P	ns	-0.07	ns	0.21	ns

Some phosphorus fractions are also related to external loading and the iron content (Table 2.3). This applies to NaOH-P, which is also closely correlated with the total phosphorus content of the sediment. In contrast, HCl-P is not, as might have been expected, related to calcium (p>0.05). Likewise, other investigations show a negligible relationship between the sediment content of phosphorus and calcium carbonate (*Jensen et al., 1992b; Fytianos & Kotzakioti, 2005*), and even though the sediment phosphorus is partly bound in calcium compounds, there are no indications that a high calcium content entails a good capacity to retain phosphorus. Instead there is a positive relationship between the HCl-P fraction and external loading (Pex, Table 2.2). This is suggestive of enhanced formation of HCl-P in connection with increased calcium deposition at higher primary production. This explanation is rendered more credible by the circumstance that phytoplankton biomass in a multiple regression with Pex is positively related to HCl-P (/11/).

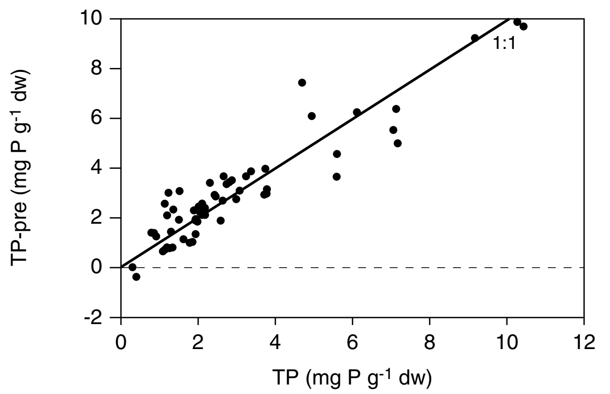


Figure 2.10 Measured and calculated values of total phosphorus concentrations in the surface sediment of 21 lakes (1-4 sampling locations in each lake). Values of total phosphorus (TP) are calculated as: TP = - 2.46 + 0.26 Pex + 0.024*Fe + 0.090*LOI + 0,58*z, $p < 0.0001$, $r^2 = 0.91$, where Pex is the external phosphorus loading (g P m^{-2} yr^{-1}), Fe is the iron concentration (mg Fe g^{-1} dw), LOI is loss on ignition (% of dry weight) and z the lake mean depth (m). From /11/.

2.3 The importance of biological structure for nutrient retention

Danish as well as international investigations show that biological structure has a great significance for the capacity of lakes to retain nutrients (/5/; *Beklioglu et al., 1999; Jeppesen et al., 1998a*). Clearwater conditions, for instance arising from fish manipulation and increased top-down control of phytoplankton via enhanced zooplankton grazing, thus often result in considerably lower concentrations of both phosphorus and nitrogen (*Benndorf & Miersch, 1991; Nicholls et al., 1996*). A Danish example of this is Lake Væng located in Central Jutland, where a 50% reduction of the fish stock and a shift from the turbid to the clearwater state in the mid-1980s yielded significantly lower nitrogen and phosphorus concentrations despite an unchanged external loading (/14/).

Lake Engelsholm

Another example of the significance of biological structure is Lake Engelsholm, situated in the county of Vejle, where markedly lower phosphorus concentrations, increased phosphorus retention and an improved Secchi depth from 1.1 to 2.3 m were recorded following a 66% reduction of the fish stock (/5/; Fig. 2.11). In summer the median total phosphorus concentration decreased from ca. 0.2 to 0.1 mg TP l^{-1}, while winter values remained unchanged. PO_4-P increased during winter and autumn, but stayed below 0.01 mg P l^{-1} during summer, also after the biomanipulation.

Total nitrogen also decreased most markedly during summer in Lake Engelsholm, but was reduced during winter as well. Most of the winter total nitrogen reduction owed to reduced nitrate concentrations, while nitrate during summer was below 0.1 mg N l^{-1} both before and after the intervention. The decreased total nitrogen concentrations during summer largely reflect the reduced phytoplankton biomass. Based on data from 695 Danish lakes, the relationship between chlorophyll and particulate nitrogen (N_part = TN - NO_3 - NH_4) can be described as chlorophyll *a* = 8.9 + 36.2*N_part ($p<0.0001$. $r^2=0.21$), which entails that the observed reduction of 1-1.5 mg TN l^{-1} during summer more or less reflects the simultaneous reduction in chlorophyll *a* of 60-70 µg l^{-1}.

Silicate concentrations responded differently over the season following the shift from the turbid to the clearwater state. When clear water was established, considerably higher silicate concentrations were seen in winter and spring, while lower concentrations were recorded in late summer and autumn. The substantially higher concentrations in spring probably reflect the lower biomass of diatoms, while the reduced increase in late summer probably results from the smaller proportion of organically bound silicate reaching and decomposing in the sediment. A higher benthic production and uptake of silicate at the sediment surface may also subdue the release from the sediment. Alkalinity changes were less marked, but a clear pattern of higher summer and autumn but lower spring concentrations emerged.

The increased retention of phosphorus could be attributed to a reduction of the period with negative retention during summer from 6 to 4 months, with simultaneously declining maximum phosphorus concentrations (Fig. 2.12). In consequence, annual phosphorus retention increased from -2.5 to 3.3 g P m^{-2} yr^{-1}.

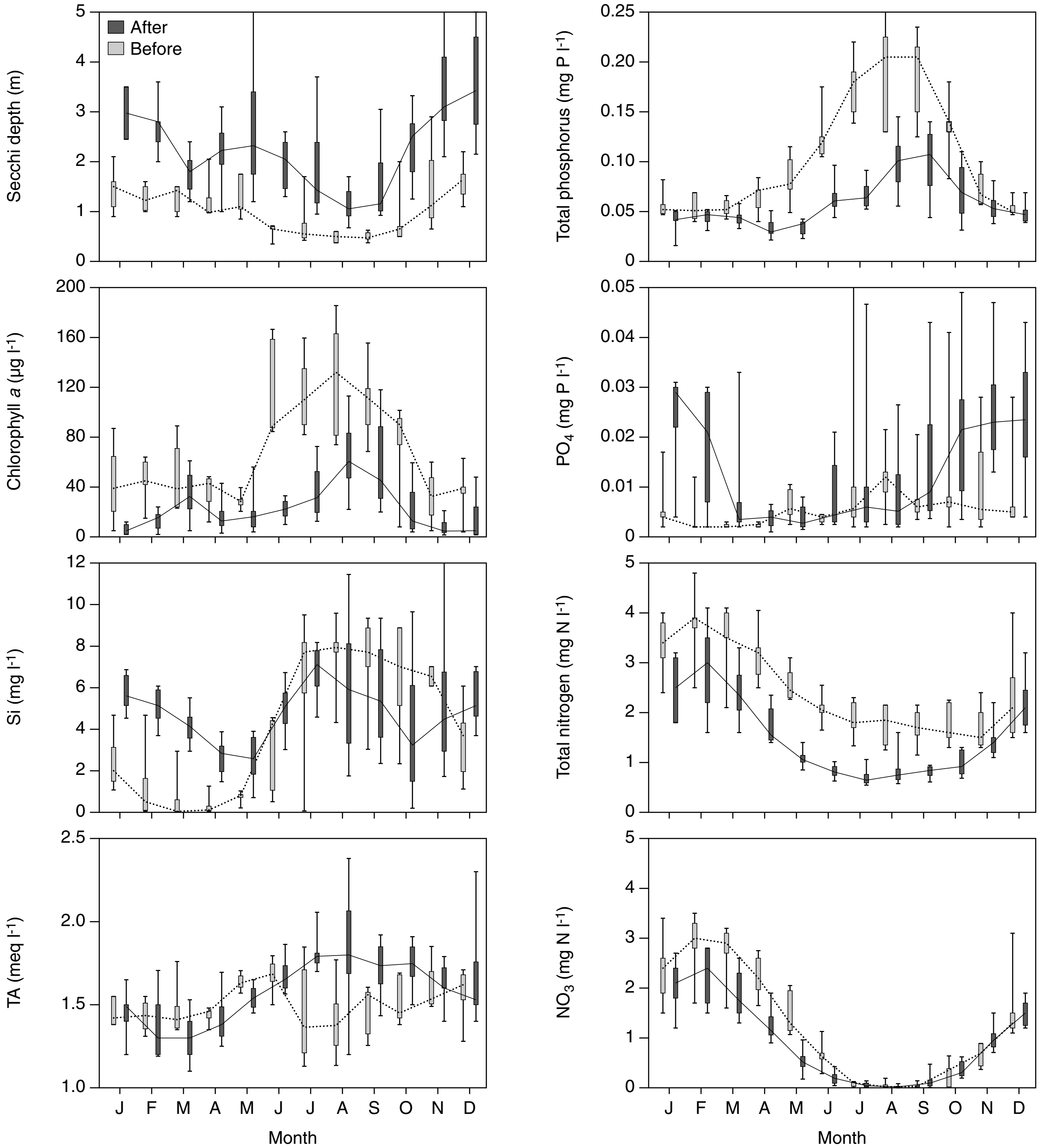


Figure 2.11 Annual variations in Secchi depth, chlorophyll *a* and a number of water chemical variables in Lake Engelsholm before and after fish stock intervention. Before-values are based on measurements from 1989-1993 and after-values on measurements from 1994-1999. Each box shows 25 and 75 % quartiles, while upper and lower lines represent 10 and 90 % percentiles. Median values are connected by lines. Based on /5/ and unpublished data.

Overall, the results from Lake Engelsholm clearly demonstrate that a shift from a turbid to a clearwater state in shallow lakes, owing for example to increased top-down control on phytoplankton, may lead to considerably lower in-lake nutrient concentrations and increased nutrient retention despite an unchanged external loading (/5/, /14/; *Benndorf & Miersch, 1991; Nicholls et al., 1996; Jeppesen et al., 1998b*).

Reasons for increased nutrient retention at shifts from the turbid to the clearwater state

The mechanisms behind the increased nutrient retention occurring at the shift between the clearwater and the turbid state in shallow lakes merit further study to be fully elucidated. However, a key explanatory factor is probably that clearwater conditions may trigger increased growth and production of benthic algae on the sediment surface, as indicated by silicate data in Lake Engelsholm. This leads to increased uptake of nutrients at the sediment surface, preventing these from reaching the water phase. Benthic production at the sediment surface will expectedly also create oxidation deeper down the sediment, which, via the redox-dependent binding of phosphorus to iron compounds, will enhance phosphorus retention (*Hansson, 1989; Van Luijn et al., 1995; Woodruff et al., 1999*). In a comparison of shallow, nutrient-rich and turbid Lake Søbygård with shallow, nutrient-rich but clearwater Lake Stigsholm, Liboriussen & Jeppesen (2003) found that although total algal production was of the same magnitude, epipelic algae were responsible for 77% of the production in the clear lake compared to only 4% in the turbid lake.

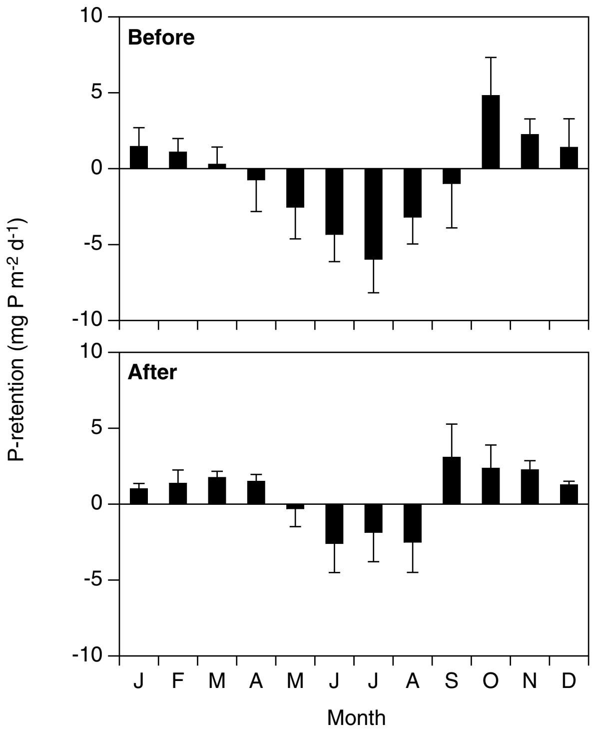


Figure 2.12 Monthly retention of phosphorus in Lake Engelsholm before (1989-1993) and after (1994-1999) the fish stock intervention that triggered a shift from a turbid to a clearwater state. From /5/.

The effect of increased benthic production will be strongest in shallow lakes with extensive shallow areas where a relatively small improvement of Secchi depth may create favourable light conditions over large parts of the lake bottom. In such lake types minor changes in light conditions may have an extensive impact on nutrient retention. An example of how improved Secchi depth markedly expands the area of light-exposed sediment is the changes observed in Lake Væng as a result of the fish stock manipulation (Fig. 2.13). Increased growth of benthic algae due to increased light may also contribute to diminishing the effect of resuspension because the sediment surface becomes more solidly cemented, thus diminishing resuspension (*Y. Vadeboncoeur, unpublished results*).

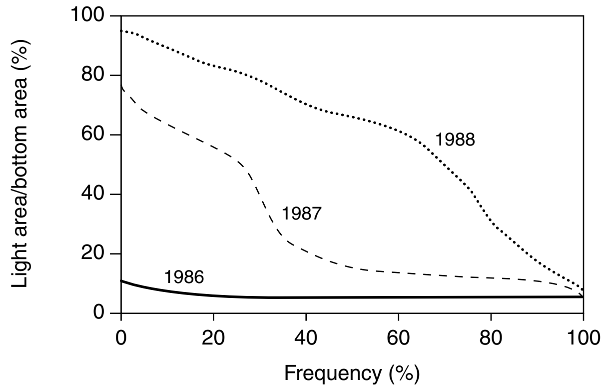


Figure 2.13 Frequency distribution (summer) of the part of the lake area where Secchi depth reached the bottom before (1986), during (1987) and after (1988) the fish stock intervention in Lake Væng. Half of the summer 1986 Secchi depth reached less than 10% of the lake bottom, the corresponding percentage being 70% in 1989. From /14/.

Enhanced coverage of submerged macrophytes in connection with improved light conditions may possibly also contribute to increased retention via among other processes oxidation of the sediment surface (see section 3.5), but there is no evidence that establishment of submerged macrophytes is a prerequisite for increased nutrient retention. In the Lake Engelsholm example there was no major increase in submerged macrophyte abundance, but still phosphorus retention increased markedly following the shift to clearwater conditions. The substantial change in retention of both nitrogen and phosphorus at the shift between the turbid and the clearwater state may well be another reason for the existence of alternative equilibria in lakes (*Scheffer et al., 1993*). In the turbid state the phosphorus pools of the sediment contribute more to the phosphorus pools of the water phase than under clearwater conditions. This may generate self-reinforcing mechanisms contributing to maintaining either the turbid or the clearwater state.

The significant impact on nutrient retention caused by alterations in biological structure entails a risk that biomanipulated lakes may revert to a turbid state if a large sediment pool of mobile phosphorus is again activated. For lake managers it may thus be unwise to undertake fish manipulation with the aim to rapidly obtain clearwater conditions immediately

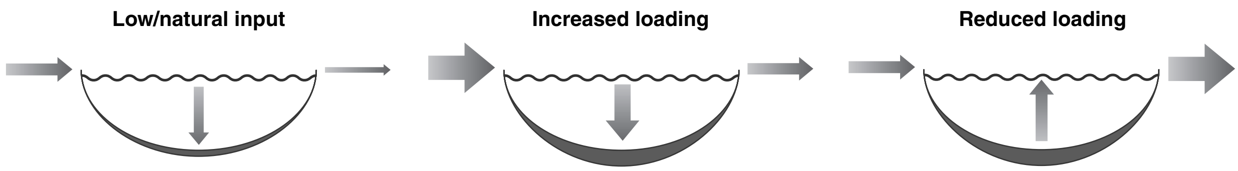


Figure 2.14 Diagram illustrating how input, retention and net release of phosphorus change in lakes following alterations in external phosphorus loading. The size of the arrows symbolises the relative extent of the three transport rates. From Søndergaard *et al.* (2003a).

upon a reduction of the nutrient input. The reason is that if a large pool of potentially mobile phosphorus has been accumulated in the lake sediment, there is a considerable risk that this will become activated, implying that the effects of the manipulation will only be of short duration and that the lake will shift back to the turbid state. In such cases it may be best to postpone the intervention for some years until the size of the mobile pool of sediment phosphorus has diminished.

2.4 Internal phosphorus loading and the transitional phase following nutrient loading reduction

The term "internal phosphorus loading" is used about the situation where the sediment after a reduction of the external phosphorus input exhibits a net release of phosphorus to the water phase (Fig. 2.14), thereby maintaining high in-lake phosphorus concentrations for multiple years despite the reduced loading (/12/; *Marsden, 1989*).

Sediment release of phosphorus occurs in all lakes – also in lakes to which the external loading has not been reduced. The difference is that in the latter, there will be no net release of phosphorus on an annual scale despite periodical events of net phosphorus release, as will be illustrated in the next section by the seasonal mass balance calculations conducted for lakes with different total phosphorus concentrations. Moreover, even in periods without net release of phosphorus, there may still be a large gross release from the sediment, this just being counteracted by a corresponding or higher sedimentation (see section 2.1).

Seasonal retention of phosphorus

Nutrient retention and internal phosphorus loading vary considerably over the season, as clearly shown by the seasonal variations in phosphorus concentrations in shallow Danish lakes with different nutrient levels (Fig. 2.15). In the nutrient-rich lakes phosphorus concentrations are typically 2-3 times higher in summer than in winter (/9/; *Jeppesen et al., 1997a*). Similar observations have been made in lakes abroad (*Welch & Cooke, 1995; Nicholls, 1999*). This difference cannot be ascribed to changed input, but must be caused by seasonal changes in the interactions between the sediment and the water phase.

The difference between summer and winter concentrations is most pronounced in the most nutrient-rich lakes. Correspondingly, phosphorus retention is lower, and in nutrient-rich lakes often negative, during most of the summer (Fig. 2.16). In the most nutrient-rich lakes, net release is found from April to September, while phosphorus is only released in July and August in lakes with concentrations lower than 100 µg P l^{-1}.

There may be several explanations of the seasonal retention and release of phosphorus. Probably the environmental state of a lake is determined by lake-specific conditions, but some general characteristics prevail: most importantly, the sediment holds a significant capacity to retain phosphorus in winter. This applies also to nutrient-rich lakes and lakes exhibiting an annual net release of phosphorus, i.e. lakes suffering from internal phosphorus loading. Such winter retention is probably primarily redox-conditioned. Lower temperatures, less sedimentation and reduced turnover of organic matter in winter result in lower oxygen and nitrate consumption in the top sediment and a thicker surface layer of oxidised sediment (*Jensen & Andersen, 1992*). At the same time nitrate concentrations in the water phase are generally higher in winter due to high runoff from the catchment and lower nitrate loss via denitrification, both of which contribute to enhancing the diffusion of nitrate down the sediment.

The importance of nitrate for phosphorus retention in lake sediments has also been demonstrated by lake restoration experiments using nitrate addition (see section 4.2). Redox-dependent retention has been demonstrated in experiments using sediment from four Danish lakes by Jensen & Andersen (1992) who found wide annual variations (3 to 15 mm) in the thickness of the oxidised sediment layer. In three of the four lakes temperature alone explained

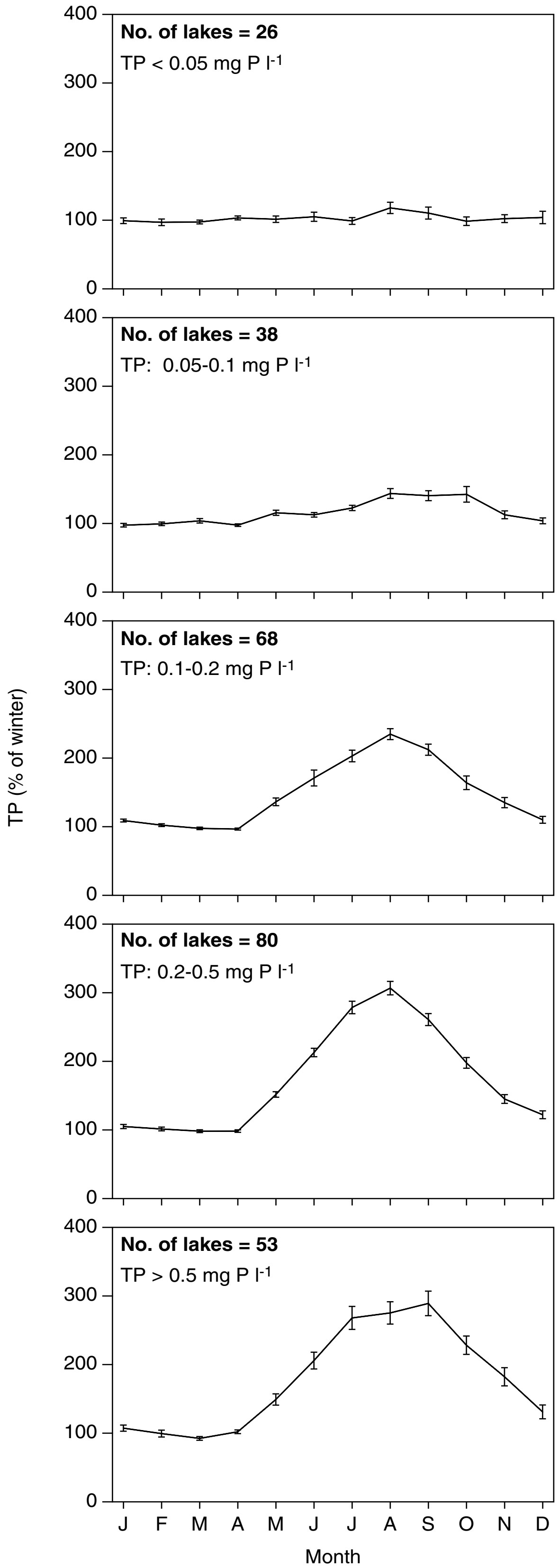


Figure 2.15 Seasonal variation in total phosphorus concentrations of 265 shallow Danish lakes divided into 5 categories according to phosphorus content (summer averages). The seasonal variation is shown as relative values compared to average winter concentrations (= 100%). From /9/.

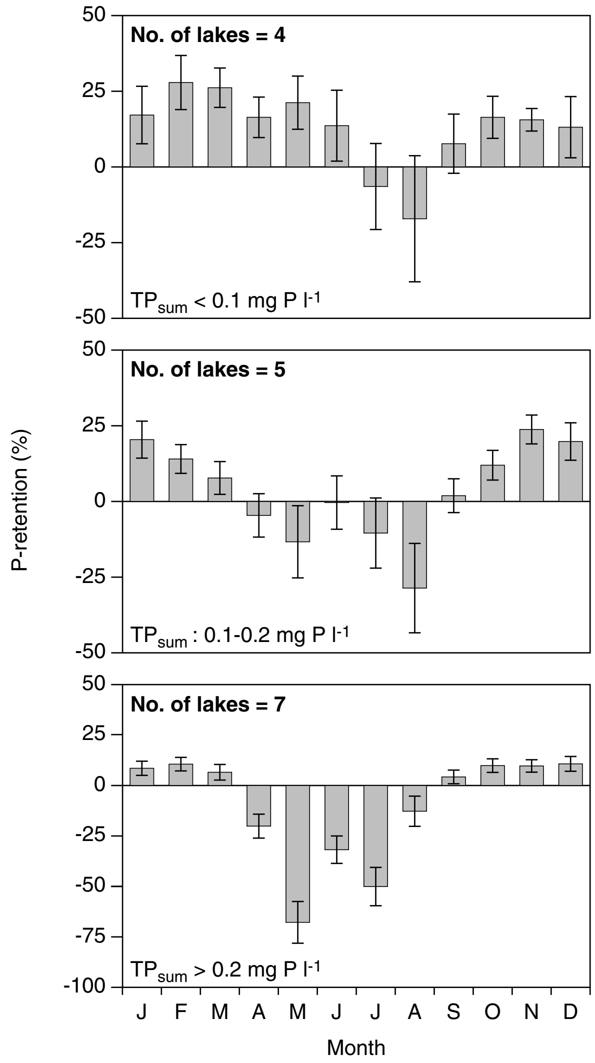


Figure 2.16 Monthly retention of phosphorus in 16 shallow Danish lakes divided into 3 phosphorus categories. The boxes represent eight years of measurements (1989-1996) with indication of standard error. From /9/.

70% of the seasonal variation in the sediment phosphorus release. Moreover, Jensen & Andersen (1992) showed that high nitrate concentrations led to enhanced thickness of the oxidised layer.

In spring with increasing temperatures, enhanced sedimentation and turnover of organic matter, the thickness of the oxidised sediment layer decreases and so does the phosphorus binding capacity (Fig. 2.17). The result is release of phosphorus from the surface layer where it was retained in winter. Later in the season this layer and the potential to release phosphorus will change concurrently with the possible alterations in the water phase. For instance, net release seems to be low or non-existent in early summer, typically June (Fig. 2.16). This agrees well with the clear water observed in many

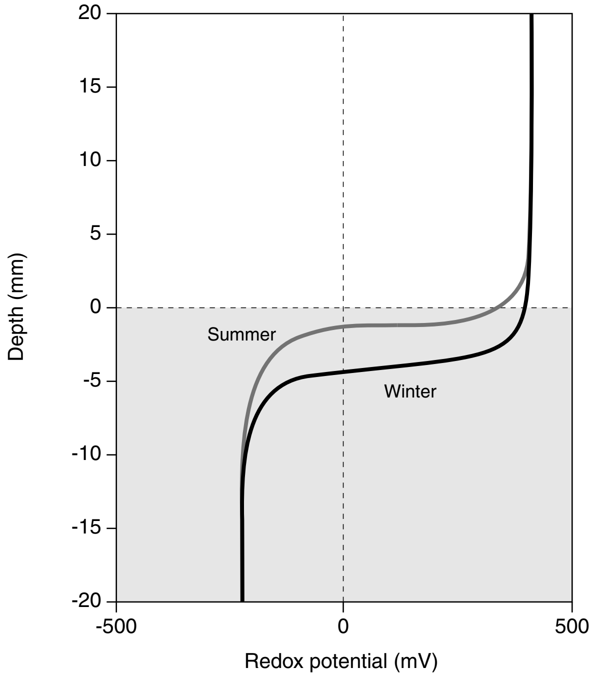


Figure 2.17 General illustration of how the redox potential of the sediment surface changes in a winter and summer setting of eutrophic lakes.

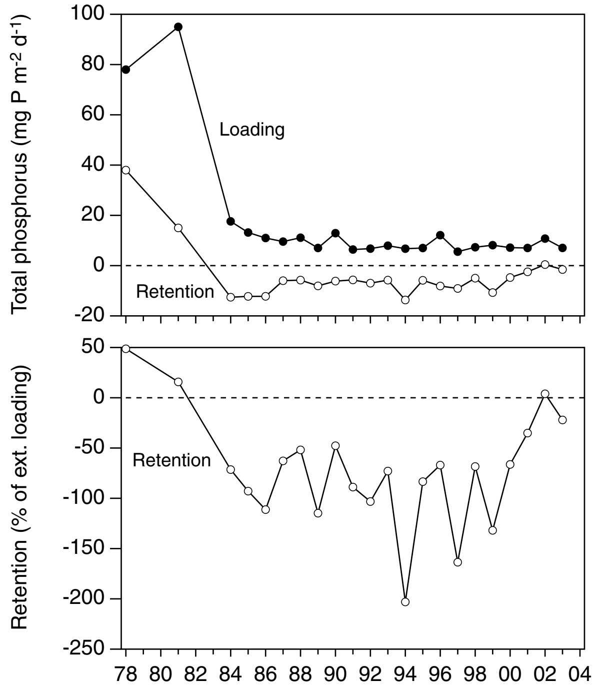


Figure 2.18 Annual input and retention of phosphorus in Lake Søbygård from 1978 to 2004. Below: relative retention as % of external input. Revised after Søndergaard *et al.* (2001a).

lakes at this time of year when the zooplankton is capable of limiting phytoplankton abundance for a short period before the year's fish fry is hatched (*Sommer et al., 1986; Talling et al., 2005*). The enhanced phosphorus retention capacity of the sediment during this clearwater phase may partly be explained by the lower sedimentation and turnover in the surface layer, and in shallow lakes partly by the fact that the reduced turbidity here permits increased benthic production.

Transition period after loading reduction

Concurrently with the release and uptake of phosphorus from the surface layer, phosphorus may also be transported upward from the deeper-lying sediment layers (*Reddy et al., 1996*). Especially in lakes where large quantities of phosphorus have been accumulated, this may contribute to internal loading for a prolonged period of time. In shallow lakes where the surface sediment holds an unused capacity to bind phosphorus, phosphorus transported upward from deeper layers will be retained in the upper oxidised sediment layer; however, if this layer is fully saturated, it may reach the water phase.

An example of this is highly nutrient-rich Lake Søbygård, where decades of extensive external phosphorus loading combined with high iron concentrations resulted in high accumulation of phosphorus in the sediment. For more than 20 years after

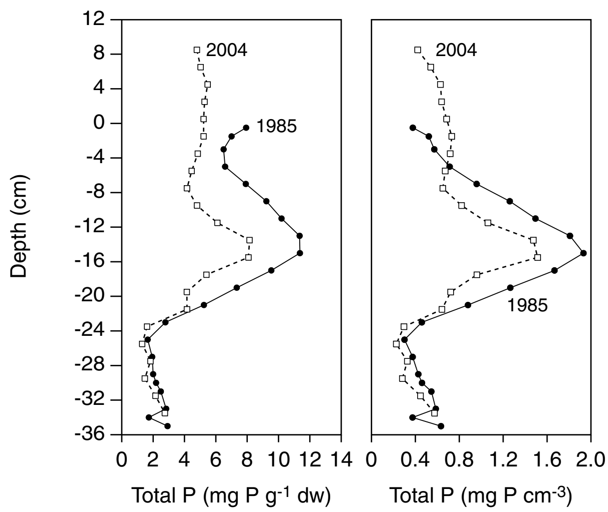


Figure 2.19 Changes in the sediment phosphorus profile from 1985 to 2004 in Lake Søbygård expressed as the phosphorus content per dry weight content (left) and as phosphorus content per volume (right). The 2004 sediment surface is adjusted in accordance with an annual net sediment growth of 0.5 cm. Revised according to /9/.

an 80-90% reduction of the external input in 1982, there has been a net release of phosphorus from this sediment pool, phosphorus retention thus being negative (Fig. 2.18). The release mainly derives from the very nutrient-rich parts of the sediment, but as deep down as to 20-25 cm, as evidenced by 20-year depth profile comparisons (/9/; Fig. 2.19). The release primarily comes from the NaOH-P fraction, and in the upper sediment layers

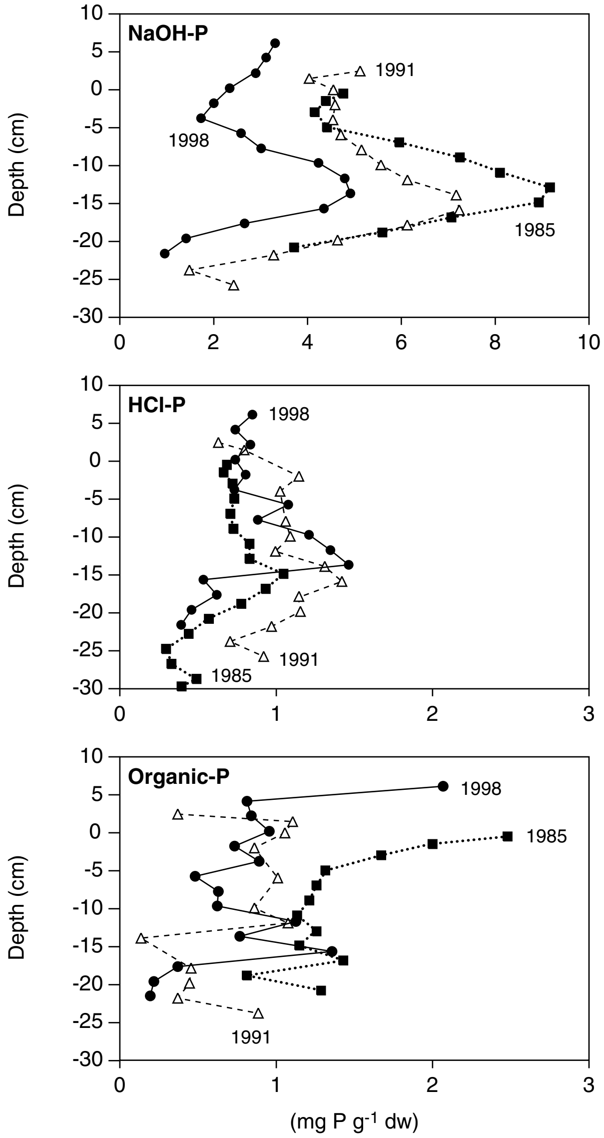


Figure 2.20 Changes in sediment phosphorus fractions from 1985 to 1998 in Lake Søbygård. The 1991 and 1998 sediment surfaces are adjusted in accordance with an annual net sediment growth of 0.5 cm. From /4/.

also from the organically bound phosphorus pool (/4/; Fig. 2.20). In contrast, the HCl-P fraction has changed only negligibly during the period, rather it has shown a tendency to increase with time. Albeit the figures derive from a single mid-lake station, the observed reduction of sediment phosphorus concentrations corresponds well with that determined by mass balance calculations, and the station thus appears to be well representative of the whole lake (/9/).

Traditionally, only approx. 10 cm of the upper sediment are presumed active (*Boström et al., 1982; Wang et al., 2003*). However, the Lake Søbygård example, where much deeper layers are involved in the exchange to the water phase, indicates that this presumption may need to be reconsidered and cannot readily be transferred to all lakes. In fact, not many lakes can be found for which a description of vertical changes following loading reduction can be elaborated. This is because the total phosphorus pool of the sediment in most lakes is so big that the relatively modest changes occurring in net retention often drown in measurement uncertainties and local variations in sediment phosphorus concentrations.

Shallow conditions and frequent occurrence of resuspension are factors hampering a shift to clearwater conditions. In principle, the process may be irreversible, implying that when once a highly organic rich and easily resuspendable sediment layer has been established, the lake will remain in a turbid state. This mechanism supposedly works in many Florida lakes, examples being large and shallow Lake Okeechobee (lake area 1730 km^2, depth only 2.7 m) and Lake Apopka (lake area 124 km^2 and 1.6 m deep) (*Havens et al., 1996; Bachman et al., 1999*), and it has given rise to a debate of whether improved water quality can be obtained solely by changing the input of nutrients (*Lowe et al., 2001*).

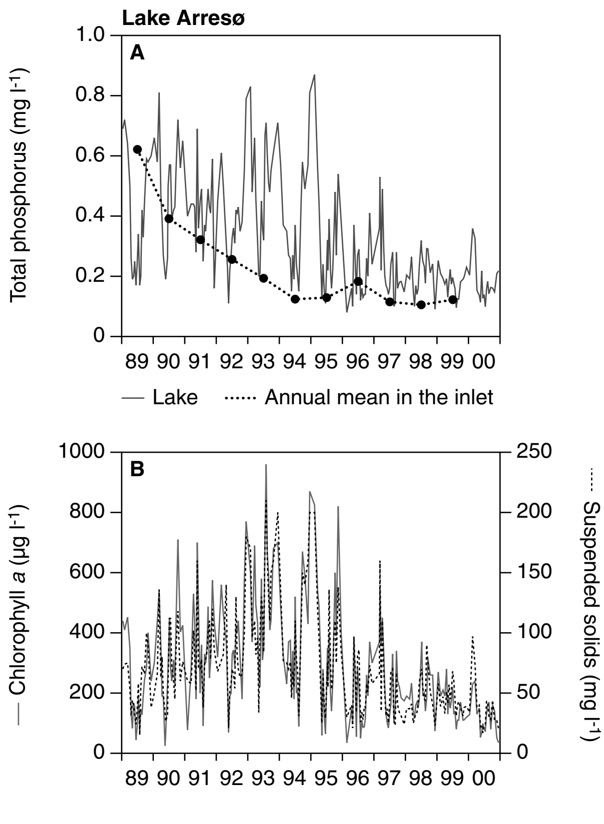


Figure 2.21 The development in total phosphorus concentrations in inlet and lake water, chlorophyll *a* and suspended matter in Lake Arresø from 1989 to 2000. From /16/.

A similar situation applies to Danish lakes, although at a somewhat lower scale. For Denmark's largest lake, Lake Arresø (lake area: 40 km^2), calculations thus showed that resuspension occurred half of the time and that the average flux of suspended solids from the sediment to the water was 300 mg m^{-2} d^{-1} (*Kristensen et al., 1992*). As much as 2 cm of the sediment surface were stirred into the water phase at heavy wind events. However, analyses of 15 shallow Danish lakes do not indicate that low water depth and frequent resuspension in themselves prevent a return to a clearwater state at reduced nutrient input (/15)/. It appeared that the reduction of phytoplankton abundance following reduced external input of phosphorus was accompanied by a proportional or nearly proportional reduction in the levels of detritus and inorganic suspended solids. In correspondence with this, in Lake Arresø the concentrations of chlorophyll *a* and suspended matter have decreased notably since the mid-1990s concurrently with the decreasing lake water concentration of phosphorus (Fig. 2.21).

As mentioned earlier, the sediment of Lake Søbygård has now been a net contributor of phosphorus for 20 years. However, in most lakes the transition period does not last as long, even though the end of the story has not been written yet for many lakes. Based on an international cross-analysis of multiple lakes to which phosphorus loading has been reduced, it was concluded that the average period before internal loading abates lasts 10-15 years (*Jeppesen et al., 2005d*). However, there are large variations, and some lakes respond rapidly and more or less to the flushing time of the lake water (ca. 3 * the water retention time (see S*as* (1989) and *Welch & Cooke* (1999) for examples). Most probably, the large

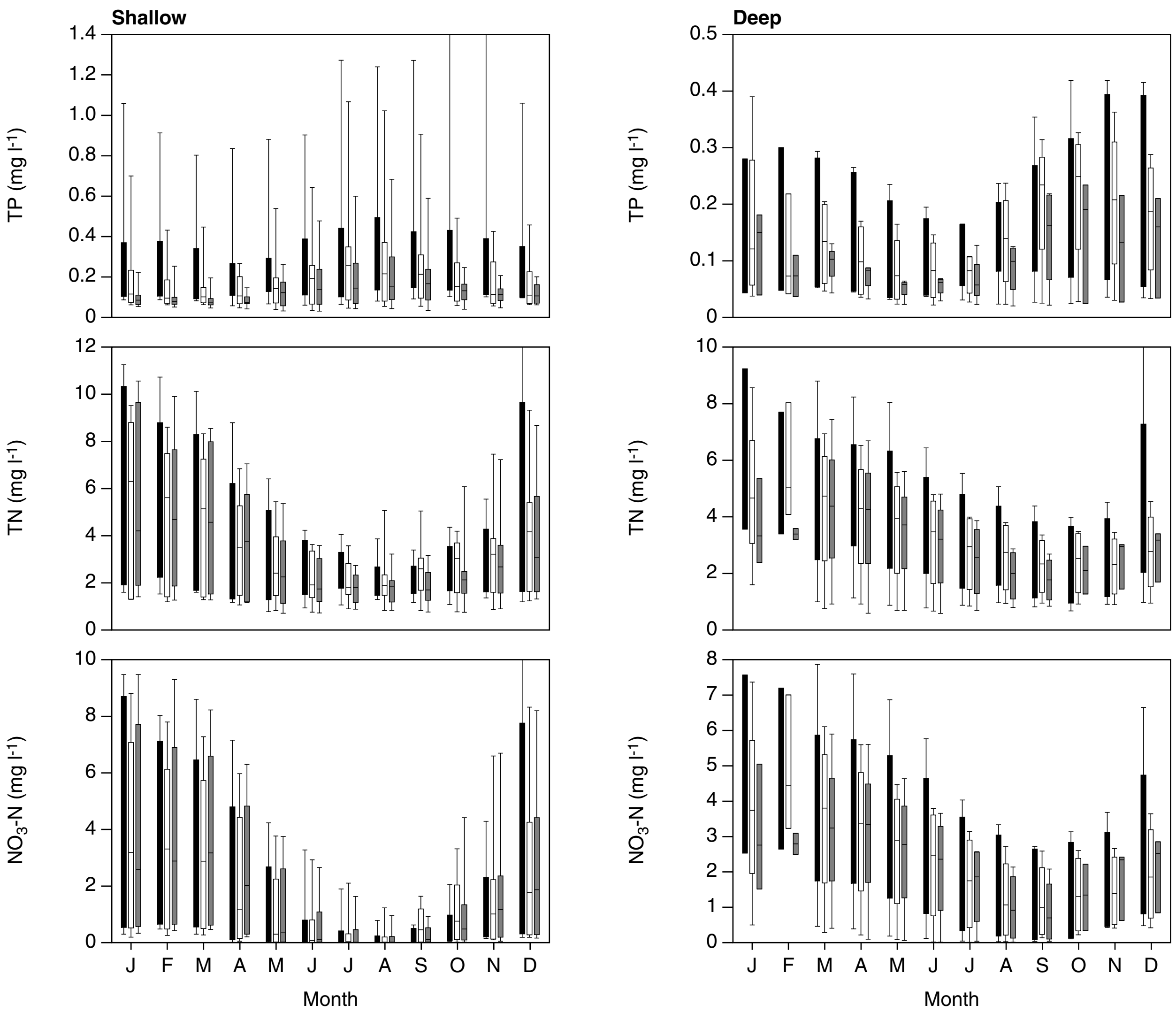


Figure 2.22 Seasonal changes in phosphorus and nitrogen concentrations in 8 shallow and 4 deep Danish lakes to which the external phosphorus loading has been significantly reduced. The boxes represent three periods: 1989-1992 (left, black boxes) characterised by a still relatively high phosphorus loading; 1993-1997 (middle, white boxes) when the phosphorus loading reached its present level; and 1998-2001 (right, grey boxes) with only negligible changes in external loading. Each box shows 25 and 75% quartiles, upper and lower lines indicating 10 and 90% percentiles. From /1/.

Table 2.4 Statistical analyses of seasonal changes (1989-2001) recorded in 4 deep and 8 shallow Danish lakes with reduced total phosphorus (TP) loading. The tests were performed on log-transformed data on medians for each month of the 13-year investigation period using linear regression: +: increase, p< 0.1; ++: increase, p<0.05, +++: increase, p<0.01; -: decrease, p<0.1; --: decrease, p< 0.05; ---: decrease p<0.01; empty cell, p>0.1. The table is modified from /1/.

		Month											
Variable	Lake type	J	F	M	A	M	J	J	A	S	O	N	D
TP	Shallow	---		---	---	---	---			---	---	---	
	Deep					--	--	---					
PO_4	Shallow				---	---	---		--	---	---	--	
	Deep					---		---	---				
TN	Shallow		--	--	--	---	---	---		--			
	Deep								--				
NO_3	Shallow			--				---					
	Deep												
TN:TP	Shallow	++	+	+++	+++	+++	+++	+++	+++	+++	+++	+++	+
	Deep												

variation occurring in the internal phosphorus loading can be ascribed to lake-specific differences, such as loading history and the chemical characteristics of the sediment, including the iron content (*Marsden, 1989*). Also, long periods with high phosphorus loading increase the potential for accumulation of large phosphorus pools, as seen in the Lake Søbygård example.

Seasonal transition following loading reduction

During the transitional phase following reduced external nutrient loading, lake water nutrient concentrations do not necessarily decrease uniformly throughout the season. An analysis of eight shallow and four deep Danish lakes to which phosphorus loading has been significantly reduced thus showed that phosphorus concentrations decreased most markedly during spring and autumn, while the reduction in July-August was negligible (/1/; Fig. 2.22; Table 2.4). In contrast, in deep lakes the largest phosphorus reduction occurred in May-August, emphasising the difference between deep and shallow lakes also in the transitional phase. In Barton Broad (UK) and Lake Müggelsee (Germany), Phillips *et al.* (2005) and Köhler *et al.* (2000, 2005), respectively, observed similar seasonal patterns after reduced external loading.

The differing response of the transitional phase throughout the year may be an effect of increased benthic-pelagic coupling, as mentioned in section 2.3, where reduced external phosphorus input results in lower phytoplankton abundance, improved light conditions at the sediment surface, increased benthic primary production, and reduced phosphorus release from the sediment surface (*Van Luijn et al., 1995; Woodruff et al., 1999*). The weaker effect during summer contrasting the increased phosphorus retention in spring may be interpreted as continuous and non-impacted loading from deeper sediment layers or, alternatively, as increasing temperatures resulting in deteriorated capacity of the oxidised surface layers to retain phosphorus (/9/). Tallberg (1999) has also suggested that the decomposition of diatoms in the top sediment a few weeks after their sedimentation may produce silicate pulses sufficiently high to impact the mobility of phosphorus. However, there are no indications that this process should have quantitative importance in Danish lakes despite that the silicate turnover is evidently influenced by the reduced phosphorus input (/1/). Finally, the reduced pH resulting from decreased primary production, which is particularly pronounced in spring and autumn (/1/), may also have reduced the pH-dependent phosphorus release (*Søndergaard, 1988; Welch & Cooke, 1995*).

Nitrogen concentrations have changed much less conspicuously than those of phosphorus during the recovery phase, which reflects primarily the lower decrease in nitrogen loading. However, despite the poor changes in total nitrogen loading, a clear trend towards lower in-lake total nitrogen concentrations was spotted in both deep and shallow lakes, especially during summer. Inorganic nitrogen in the shallow lakes did not show a similar decline, and the decreasing total nitrogen concentrations were due to a reduction in the particulate fraction as well as to the overall reduced phytoplankton biomass (/1/). Total concentrations do not necessarily represent biologically available forms and predictions of limiting nutrients based on TN:TP ratios may overestimate the importance of phosphorus (*Schelske et al., 1999*). Nevertheless, the increasing ratio suggests that, as recovery proceeds, nitrogen is less likely to become a limiting nutrient in Danish shallow lakes.

3 Mechanisms behind the sediment release and uptake of phosphorus

Various mechanisms determine the release and uptake of phosphorus from the sediment (*Wang et al., 2003*). Contrary to nitrogen, which is primarily found in organic compounds and whose release is mainly induced by the decomposition of organic substances, phosphorus is also bound in a number of inorganic compounds. Consequently, chemical mechanisms have a greater impact on phosphorus than on nitrogen. Below, a survey will be given of the most significant mechanisms behind sediment phosphorus release, and although most of these are closely coupled with no clear distinction between them (e.g. effects of temperature, mineralisation, redox conditions and microbial processes) they will be treated individually.

3.1 Resuspension

The physical resuspension of material from the lake bottom is a factor of particular significance in shallow and wind-exposed lakes, i.e. potentially also in many Danish lakes. In resuspension models this is considered by the fact that the wave height, which influences the sediment surface (benthic shear stress) and creates sediment stirring, is dependent on the distance of the wind-exposed water surface (effective fetch), water depth and wind speed (*Hamilton & Mitchell, 1996*).

The effect of resuspension on the concentrations of suspended matter may also be modelled directly by empirical relations (*Hamilton & Mitchell, 1997; James et al., 2004; Jin & Ji, 2004*). This method was, for instance, employed for Lake Arresø with a mean depth of only 3 m. Here relations were established between wind speed, concentrations of suspended matter and the sedimentation rate (*Kristensen et al., 1992*).

The immediate effect of resuspension is higher concentrations of suspended matter in the water phase during windy periods. This effect is traced by the empirical relationships established between phosphorus concentrations, water depth and Secchi depth. Here, water depth is a significant factor negatively influencing Secchi depth: Secchi depth = $0.33*TP^{-0,53} * Z^{0,20}$, where Z is mean depth (*Jensen et al., 1997*). Also, several examples exist of substantial increases in the concentrations of particulate fractions in Danish shallow lakes in windy periods. An example is Vest Stadil Fjord located north of the town of Ringkøbing, where enhanced wind speeds resulted in a factor 5-10 increase in TP and concentrations of suspended matter within a two-day period (Fig. 3.1; *Søndergaard et al., 2001b*).

Resuspension may also act as a trigger of periodical release of nutrients, which again impacts the development of phytoplankton and water quality (*Reddy et al., 1996; Istvánovics et al., 2004*). The importance of resuspension for the sediment phosphorus release depends, however, strongly on the characteristics of the water into which the sediment is stirred, as the resultant effect is decided by the equilibrium state between particular-bound phosphorus and dissolved phosphate under the given conditions (pH, oxygen, etc.). Thus, at high lake water phosphorus concentrations, there may, in principle, be an uptake from water to sediment if the stirred sediment particles have an unexploited binding potential. An increased binding potential may also exist when iron-rich sediment particles from an oxygen-poor environment are stirred up into the more oxidised environment in the water phase. In the Vest Stadil Fjord example, there was no or only a very modest increase in total phosphorus concentrations after wind action and thereby no traceable long-term effect of resuspension. In other lakes, such as Lake Arresø (/13/) or Lake Taihu in China (*Fan et al., 2001*), resuspension seems to lead to increased release rates. It may also be, as indicated by the resuspension experiments with Lake Arresø sediment (/13/) and concluded from investigations conducted in a Finnish shallow lake (*Horppila & Nurminen, 2001*) and in Icelandic Myvátn (*Einarsson et al., 2004*), that resuspension may periodically cause increased release, while at other times it has no demonstrable longer-term effects (Fig. 3.2).

3.2 Temperature and microbial processes

The strong impact of temperature on the rate of almost all processes and the turnover occurring in the sediment and water phase implies that temperature can be regarded as an integrating factor for a number of mechanisms. The substantial seasonal retention of phosphorus in nutrient-rich lakes, as shown in Fig. 2.14 and other

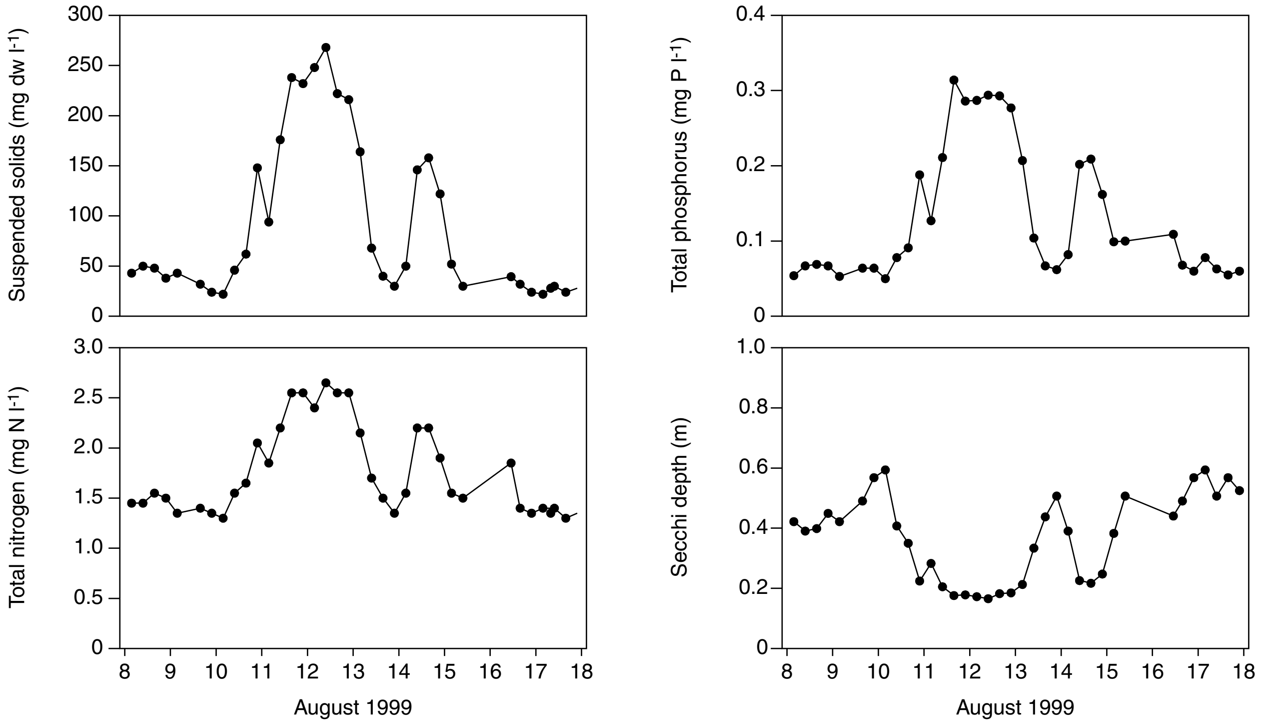


Figure 3.1 Changes in Secchi depth and lake water contents of suspended matter, total phosphorus and total nitrogen in Vest Stadil Fjord during a 10-day period with varying wind speeds in August 1999. The wind speed changed from 0-2 to 5-7 to 2-3 m s^{-1}. Vest Stadil Fjord covers 450 hectares and has a mean depth of 0.8 m. From /4/.

investigations (*Jensen & Andersen. 1992; Boers et al., 1998*), emphasises the overall importance of temperature, although it cannot be excluded that also light may play an important role due to the close linkage between these two factors.

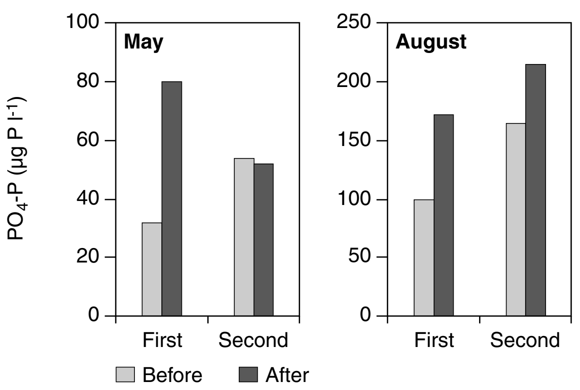


Figure 3.2 Average increase in the concentration of orthophosphate in the water phase above intact sediment columns in experiments simulating stirring of sediment from Lake Arresø. The experiments were conducted on sediment sampled in May-August and each time repeated 24 h's later. The content of suspended matter varied between 20 and 300 mg dry matter per litre. Redrawn after /13/.

Correspondingly, temperature may often be included as an explanatory variable in empirical relations between nutrient input and nutrient concentrations in lakes (*Windolf et al., 1996; Jensen et al., 2005*). Kamp-Nielsen (1975) has described the aerobic and anaerobic phosphorus release from the profundal zone of lakes based on only temperature and phosphate concentrations in the bottom water and the pore water of the sediment.

Increased temperature augments the rate of chemical diffusion and chemical processes, but the most significant impact on the nutrient release is often via the biological processes. Thus, respiration processes often have a Q10 value of approximately 2 (*Atkin & Tjoelker, 2003*) and many processes, such as denitrification, are markedly reduced at low temperatures (*Lewandowski, 1982; Tomaszek & Czerwieniec, 2003*). Enhanced temperatures thus stimulate the mineralisation of organic matter in the sediment and the release of inorganic phosphate (*Boström et al., 1982; Gomez et al., 1998*). For the phosphorus release from four sediments Jensen & Andersen (1992) found an average Q10 between 3.5 and 6.8, it being ca. 4 in Lake Søbygård at temperatures between 10-20 °C, whereas the difference between 5 and 10 °C was smaller (Fig. 2.4). Likewise, Kamp-Nielsen (1975) discovered only a modestly increased phosphorus release rate at temperatures below 10 °C compared to that at higher temperatures. Temperatures also have a direct impact on the amount of available oxygen because the solubility of oxygen in water is temperature-dependent (*Wetzel, 2001*).

Increased temperatures and turnover in spring imply higher bacterial consumption of particularly oxygen and nitrate for oxidation. This diminishes the extent of the oxidised zone in the sediment (*Tessenow, 1972; Jensen & Andersen, 1992*) and consequently also the redox-dependent binding of phosphorus to iron compounds and, in principle, the phosphorus level of the water phase as a whole (*Gonsiorczyk et al., 2001*). In accordance with this, Jensen & Andersen (1992) observed that the temperature effect on phosphorus release was most pronounced in lakes where a large proportion of the phosphorus was bound in iron compounds.

Bacteria play an important role in the turnover of the organic matter sedimenting on the bottom of lakes. The turnover rate is mainly determined by the amount of degradable organic matter and the availability of substances such as oxygen, nitrate, sulphate, iron and mangane used to a varying degree in the oxidation process (*Thomsen et al., 2004*). This means that bacteria are vitally important for the uptake, accumulation and release of phosphorus (*Petterson, 1998; Törnblom & Rydin, 1998*). Some scientists perceive the role of bacteria to be more than that of catalyzers accelerating the oxidation of organic matter and reducing various electron acceptors. In fact, Gächter *et al.* (1988) concluded that a large part of the sediment phosphorus pool was built into the bacteria and that the partially independent release of phosphorus and iron from the sediment in a number of lakes evidences that bacteria play a far more direct role in the phosphorus release process.

If oxygen and nitrate availability is low, but the concentrations of sulphate and easily degradable material are high, sulphate reduction may contribute importantly to the sediment turnover, as seen in marine environments (*Jensen et al., 1995; Holmer & Storkholm, 2001*; /6/). Subsequently, hydrogen sulphide (H_2S) formed by this process may induce the formation of iron sulphide (FeS): $2\ FeO(OH) + 3\ H_2S \rightarrow 2\ FeS + S + 4\ H_2O$. Thereby, the binding of phosphorus to iron compounds may be negatively impacted by a ligand exchange of phosphate with sulphide. This increases the potential release of phosphorus from iron-bound forms (*Ripl, 1986; Phillips et al., 1994; Kleeberg & Schubert, 2000; Perkins &Underwood, 2001*). As with many of the other release mechanisms, this process is less significant in winter when lower sedimentation rates and lower oxygen and nitrate consumption maintain iron in its oxidised form.

The significance of sulphate for the interactions between phosphorus and iron can also be illustrated by marine areas where the sulphate concentrations are much higher than in fresh water (*Hyacinthe & Cappellen, 2004*). In a comparison between freshwater lakes and coastal areas, Blomqvist *et al.* (2004) concluded that the higher availability of phosphorus in marine and estuarine areas (and with it higher nitrogen limitation) was primarily due to enhanced iron sequestration by sulfide. Minimum two iron atoms are needed to bind a phosphate molecule in the oxydative hydrolysis of iron, and as Fe:P is often lower than 2 in marine areas but higher than 2 in freshwater areas, the freshwater areas' capacity to bind phosphorus is significantly higher than that of marine areas under oxidised conditions.

3.3 Redox conditions, pH and alkalinity

Redox potential and oxidised/reduced iron

The sediment's redox potential is the classical description and explanation of how oxidation conditions influence the chemical binding of phosphorus. As described already more than 60 years ago by Einsele (1936) and Mortimer (1941), sediment phosphorus release depends on the binding potential of iron and manganese under varying redox conditions. Under oxidised conditions, iron occurs in a particulate oxidised form (Fe(III)) possessing a high affinity for binding phosphorus. In contrast, if conditions are anoxic, iron converts to a dissolved form (Fe(II)), which implies that also the adsorbed phosphorus becomes dissolved and may be transferred to the water phase.

In non-stratified lakes, water above the sediment is normally well oxidated throughout the season and the redox potential is sufficiently high to maintain iron in an oxidised form. The theoretical redox potential in oxygen-saturated water is 800 mV, and well-oxidised water normally has a redox potential between 400 and 600 mV, which is well above the 200-300 mV level where iron is reduced from Fe(III) to Fe(II). The oxidised water will diffuse down the sediment and, in the presence of sufficient iron, thereby put a lid on the sediment phosphorus pool (*Penn et al., 2000*). Diffusion of oxygen down the sediment depends on the oxygen consumption rate of microbial processes, and the rate of replenishment of oxygen from the water phase. Normally, the diffusion depth of organic-rich sediments will be 1-2 cm at the most, showing, however, seasonal variations dependent on temperature and the turnover rate of organic matter (*Jensen & Andersen, 1992; Maasen et al., 2003; Thomsen et al., 2004*; Fig. 2.17).

The presence of nitrate, which often penetrates somewhat deeper than oxygen, will also create a redox potential sufficiently high to maintain iron in an oxidised form. In this way, nitrate may be important for the redox-dependent retention of phosphorus (*Mcauliffe et al., 1998; Duras & Hejzlar, 2001*). For instance, Kozerski *et al.* (1999) found high phosphorus release rates in Lake Müggelsee, Berlin, at low nitrate input in summer. Nitrate may, though, have the opposite effect on phosphorus release; a high nitrate input stimulates the mineralisation of organic matter and with it the release of phosphorus. Thus, in a number of Danish lakes Jensen & Andersen (1992) found that occurrence of nitrate in winter and early summer reduced the phosphorus release, while high nitrate concentrations in late summer led to increased release rates in some lakes. Most Danish lakes exhibit a seasonal pattern with high nitrate concentrations in winter and low concentrations in summer (/1/; *Jensen et al., 1997*).

The significant capacity of iron to bind phosphorus implies that as long as there are free binding sites to iron in oxidised sediment, retention rates will be positive. In an investigation of 15 Danish lakes Jensen *et al.* (1992b) found that phosphorus release under oxidised conditions was poor at a weight based iron:phosphorus ratio exceeding 15. Correspondingly, Caraco *et al.* (1993) suggested that the iron:phosphorus ratio should surpass 10 for iron to regulate the phosphorus release.

pH and alkalinity

The importance of pH for phosphorus release is mainly related to the fact that the binding of phosphorus to oxidised iron compounds decreases at high pH values, phosphorus being replaced by hydroxylions on iron particle surfaces (*Andersen, 1975; Lijklema, 1976*). Thus, increased phosphorus release at high pH has been observed in connection with resuspension events in summer when pH is high due to high photosynthesis activity (*Koski-Vahala & Hartikainen. 2001*). Photosynthetically increased pH in the water phase may also influence the sediment pH (Fig. 3.3), and this may again affect the binding forms of phosphorus in the sediment and thereby also the potential phosphorus release (*Søndergaard, 1988; Welch & Cooke, 1995; Istvánovics & Pettersson, 1998*).

In alkaline lakes, low pH values may influence the solubility of apatite, for instance via low pH values occurring in connection with the decomposition of organic matter (*Golterman, 2001*). In Lake Michigan the concentration of phosphate in the oxidised sediment layer and the water mass above is assumed controlled by calcium (*Brooks & Edgington, 1994*). In accordance with this, Burley *et al.* (2001) based on investigations in three alkaline Canadian lakes concluded that the sediment phosphorus release was controlled by apatite solubility and the bacterial metabolism. It has, however, been suggested that iron and aluminium regulate the release and uptake of phosphorus also in alkaline lakes as long as their concentrations are sufficiently high (*Olila & Reddy, 1997*).

3.4 Chemical diffusion and bioturbation

The transport of phosphorus within the sediment and from the sediment to the water phase mainly occurs as phosphate released during the decomposition of organic or from inorganic compounds. Higher concentrations in the sediment than in the water phase imply an upward diffusion. Often the difference between the sediment and the water phase increases with increasing phosphorus levels and more eutrophic conditions (*Maasen et al., 2003*). The extent of the purely chemical diffusion flux can be calculated using Fick's first law and depends on the size of the concentration gradient, the diffusion coefficient and the sediment's "porosity" (e.g. *Sinke et al., 1990*).

However, often sediment release rates far higher than those explainable from chemical diffusion alone are measured (*Søndergaard, 1990*). For instance, pore water measurements from Lake Søbygård typically showed phosphate gradients between 0.5-1 mg P l^{-1} cm^{-1} in summer (Fig. 3.3), but at the same time sediment exchange experiments found release rates far higher than those expected from diffusion alone (*Søndergaard, 1990*). This strongly indicates that mechanisms other than diffusion are involved in the release of phosphorus from the sediment. One of these may be increased transport rates due to gas ebulition consisting, for instance, of methane or hydrogen sulphide generated from microbial processes in the deeper layers of the sediment. These bubbles may physically bring up water when rising through the sediment (*Ohle, 1958, 1978; Kamp-Nielsen, 1975*). The presence of invertebrates such as chironomids and bristle worms may also directly, via excretion and pumping of water, or indirectly, via stimulation of the turnover of organic compounds, contribute to the phosphate transport (*Hansen et al., 1998; Devine & Vanni, 2002*). Finally, also fish may contribute to the phosphorus transport from sediment to water in their search for food at the lake bottom (*Brabrand et al., 1990;*

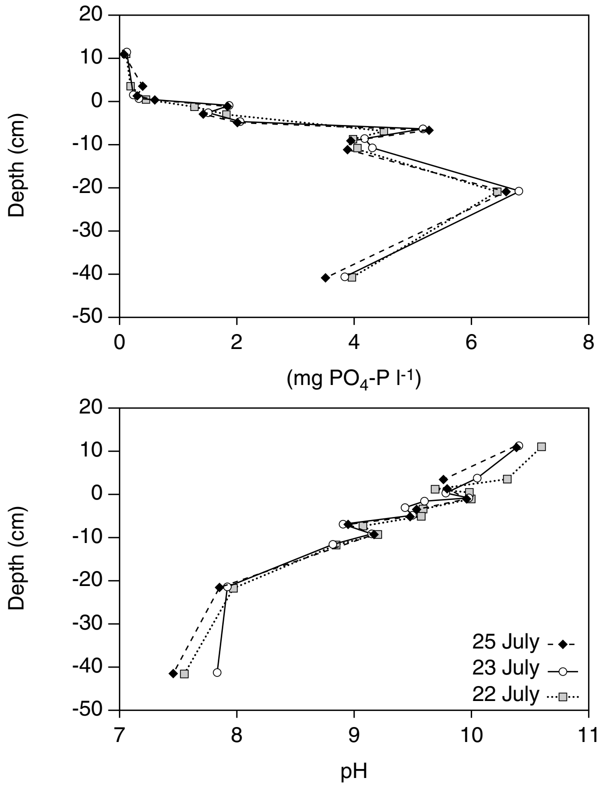


Figure 3.3 Content of orthophosphate and lake water pH value immediately above the sediment and in the sediment pore water on three different days in July 1985 in Lake Søbygård. From Søndergaard (1990).

Andre et al., 2003; Tarvainen et al., 2005). Also bottom-feeding fish increase the risk of resuspension in windy periods and they may thus have great significance for water transparency (*Scheffer et al., 2003)*.

3.5 Submerged macrophytes

The presence of submerged macrophytes impacts the accumulation and exchange of nutrients between sediment and water (*Brenner et al., 1999; Rooney et al., 2003*). This impact may be positive as well as negative. In shallow and relatively nutrient-rich lakes, the density of submerged macrophytes may become so high that water mixing is restrained. This may periodically result in surface sediment oxygen concentrations so low that redox-dependent release of iron-bound phosphorus takes place (*Frodge et al., 1991; Stephen et al., 1997*). High density of submerged macrophytes and high primary production may also lead to increased pH, which may affect the exchange of phosphorus via pH effects on the iron binding of phosphorus (*Søndergaard, 1988; Barko & James, 1997)*. In shallow lakes high macrophyte abundance may also contribute to diminishing the extent of sediment resuspension during windy periods (*Dieter, 1990*), which may again positively or negatively influence the phosphorus exchange (see section 3.1).

In less nutrient-rich lakes where the submerged macrophyte community consists of species with well-developed root systems, the roots enhance the penetration of oxygen down the sediment. This may, in turn, enhance the binding potential of the sediment phosphorus (*Andersen & Olsen, 1994; Christensen et al., 1997*).

Results from Lake Væng and Lake Stigsholm, which exhibit wide between-year fluctuations in macrophyte density (/14/; *Søndergaard et al., 1997a*), suggest that macrophytes do not as such significantly affect nutrient retention. An effect may occur during senescence where increased concentrations are observed (*James et al., 2002*), but otherwise changes in nutrient retention in shallow lakes such as Lake Engelsholm (see section 2.3) seem independent of increased plant coverage. Rather the existence of either clearwater or turbid conditions is the determinant factor (/9/; *Jeppesen et al., 1998a*).

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4 Lake restoration

In recent decades, the vital impact of nutrients on lake water quality and the increased eutrophication have led to extensive investments world-wide to reduce the nutrient input to lakes. The main focus has been directed at phosphorus removal via sewage treatment and diversion. In consequence, inlet phosphorus concentrations have, as mentioned in the introduction, decreased significantly, also in Denmark. The nitrogen input to lakes has been less markedly reduced (*Jeppesen et al., 1999a, 2005c*). Stream nitrogen concentrations have decreased by approx. 30% since 1989 (*Bøgestrand, 2004*).

Unfortunately, the massive investments have often not led to the desired improvement of water quality. This may owe to inadequate nutrient reductions or, as mentioned in section 3, a delayed and only negligible response due to internal phosphorus loading. To counteract this, many physical, chemical and biological restoration techniques have been developed during the past five decades, aimed to improve lake water quality (*Cooke et al., 1993; Jeppesen et al., 2003a*). Also in Denmark various restoration projects have been undertaken, mainly since the mid-1980s (Table 4.1). Biological tools have been most frequently employed (> 50 lakes), physico-chemical methods having been applied in less than 10 lakes (/8/; *Søndergaard et al., 1998a*).

To achieve positive and lasting effects of restoration interventions, the importance of reducing the external nutrient loading has often been emphasised (*Benndorf, 1990; Jeppesen et al., 1990; Steinman et al., 2004*). If the external loading is too high, a lake may soon after restoration revert to a state reflecting the magnitude of the loading input. To obtain lasting effects, the recommended equilibrium concentration of phosphorus for shallow Danish lakes is < 50-100 µg P l^{-1} (*Jeppesen & Sammalkorpi, 2002*).

Successful restoration interventions may also be lake-specific and depend on morphological conditions. Genkai-Kato & Carpenter (2005) concluded that the possibilities of obtaining a regime shift from turbid to clearwater conditions are poorest in lakes of intermediate depth, and restoration of such lakes is therefore difficult. The explanation is that in shallow lakes there is good potential for the occurrence of a high macrophyte coverage, which will help maintain the clearwater state, and that in deep lakes the stratification and separation of the water may limit the impacts of internal phosphorus loading on the photic zone.

4.1 Physical methods

Physical restoration methods usually aim to reduce the availability of phosphorus by physically removing phosphorus using different methods. Physical restoration has only been applied to a few large-sized Danish lakes.

Table 4.1 Survey of lake restoration methods used in Denmark.

Methods	Main principle and purpose	Danish examples
Physical		
Sediment removal	Removal of phosphorus-rich surface sediment to reduce the internal phosphorus release	Lake Brabrand
Chemical		
Oxidation of bottom water	Oxygen addition to the hypolimnion to improve the phosphorus-binding potential	Lake Hald
Nitrate treatment	Nitrate addition to bottom water to increase surface sediment oxidation and turnover of organic matter	Lake Lyng
Aluminium addition	Aluminium addition to the water/sediment to improve the phosphorus-binding potential	Lake Sønderby
Biological		
Removal of non-predatory fish	Removal of zoo- and benthivorous fish species (particularly roach and bream) to increase top-down control of phytoplankton	Lake Væng
Stocking of predatory fish	Stocking of predatory fish (pike and others) to reduce the abundance of non-predatory fish and increase zooplankton grazing	Lake Udbyovre
Transplantation	Transplantation/protection of submerged macrophytes in lakes where natural migration is hampered by bird grazing, etc.	Lake Engelsholm

Diversion of bottom water in deep lakes

The purpose of diverting nutrient-rich bottom water from deep lakes is to increase the transport of phosphorus out of the lake in order to reduce phosphorus availability. Depending on lake morphology, various technical measures are available (*Klapper, 2003*). Results are available from Austrian and Canadian experiments (*Tolotti & Thies, 2002; Macdonald et al., 2004*), but apart from a short-term experiment in Lake Søllerød in the 1970s (*Hovedstadsrådet, 1986; Søndergaard et al., 1998a*) the method has not been used in Danish lake restoration projects.

Sediment removal (Lake Brabrand)

Another method of reducing nutrient concentrations is sediment removal (*Cooke et al., 1993*), which has been performed in many small-sized Danish lakes, large-scale removal having been conducted only in Lake Brabrand near the city of Aarhus. During a 7-year period 500,000 m^3 of nutrient-rich sediment were removed from large parts of the lake at 0-90 cm depth (*Jørgensen, 1998*). Subsequently, average lake depth increased from 0.8 to 1.1 m, which was considered an important side effect in the attempt to avoid weed overgrowth. Concurrently with the sediment removal, also the external phosphorus input from the catchment was significantly reduced, from ca. 100 to 20 t annually.

The parallel interventions render a distinction between the effect of sediment removal from that of loading reduction difficult. However, the impact of sediment removal on the water quality of Lake Brabrand appeared to be only minimal. During the experimental period the average lake water phosphorus concentration declined from almost 1 mg P l^{-1} to 0.3-0.4 mg P l^{-1}, but Secchi depth increased only marginally due to the still high concentrations of phosphorus. When comparing the transitional period for phosphorus between Lake Brabrand and Lake Søbygård, Lake Brabrand proved to react much faster to the reduced external input, signalling a less significant sediment impact (*Jørgensen et al., 1998*). Recently, an inlet lake (Århslev Engsø) to Lake Brabrand has been established to reduce the nutrient input. Preliminary results and calculations indicate that this will lead to a reduction of nitrogen and phosphorus inputs (*Hoffmann et al., 2004*). It is problematic, though, to apply first-year results from new-established lakes as both state and nutrient retention may vary considerably from the values observed in the longer term (*Søndergaard & Jeppesen, 1991*).

4.2 Chemical methods

There are multiple methods of chemical restoration, all of which are designed to reduce phosphorus availability, either by improving the existing binding potential, for instance via oxidation of bottom water and surface sediment (*Jäger 1994; Müller & Stadelmann, 2004*), or by increasing the binding potential via, for example, aluminium or iron addition (*Boers et al., 1994; Welch & Cooke, 1999*). In some instances, for example when oxidizing bottom water, the purpose may be to improve the living conditions for fish and bottom animals (*Müller & Stadelmann, 2004*). In Denmark oxidation projects have been undertaken in five large-sized lakes, aluminium or iron addition in two lakes and nitrate addition in one lake.

Oxidation of bottom water in deep lakes (Lake Hald)

The most ambitious oxidation project in Denmark so far is that undertaken in Lake Hald near the city of Viborg in Central Jutland. In most years since 1985 pure oxygen has been added via diffusers to the bottom water in the two deepest areas of the lake for parts of the summer (/8/; *Rasmussen, 1998*). However, as concurrent efforts have been made to reduce external phosphorus loading by the closure of nearby fish farms, the effects of the oxidation are difficult to discern.

The addition of oxygen to Lake Hald has not led to a significant increase in average oxygen concentrations in the bottom water, whereas nitrate concentrations have risen in consequence of ammonium nitrification (*Rasmussen, 1998*). Internal phosphorus loading, however, decreased considerably before the restoration, from a net release of 1.5-3.7 t P per year to a net retention of 0.3-0.7 t P per year following the intervention.

Oxidation was stopped after 12 years, but was resumed when signs of deterioration appeared (*Rasmussen. pers. comm., www.miljo.viborgamt.dk*). Today, oxidation is still performed during a sequence of months each summer. The prolongation illustrates that this and similar interventions treat the symptoms rather than the underlying and fundamental problems, namely the sediment's large consumption of oxygen and the release of phosphorus from a mobile pool. Only when the easily degradable organic pool accumulated in the sediment – and new matter added – is mineralised or buried deep down the sediment, the sediment oxygen consumption can be expected to decline, leading to increasing phosphorus retention.

The possibility of limiting the sediment phosphorus release via bottom water oxidation may be debated. Based on a 15-year oxidation record of Lake Sempach (Switzerland), Gächter & Müller (2003) concluded that increased hypolimnetic dissolved oxygen concentrations neither led to reduced release of phosphorus from the sediment in summer nor to increased phosphorus retention. They concluded that oxygenation only leads to increased phosphorus retention if the sulphide production is lowered and more ferrous phosphate (e.g. vivianite) and less FeS are deposited in the anoxic sediment.

Nitrate addition to bottom water and sediment (Lake Lyng)

The use of nitrate for restoration purposes was first applied in Swedish lakes according to the "Riplox-method" developed in the 1970s (*Ripl, 1976*). The method involves addition of nitrate to the sediment to increase mineralisation and improve the redox-sensitive binding of phosphorus to iron. Contrasting the bottom water oxidation described above, the objective is not only to oxidise the surface sediment but also to increase the turnover of organic matter via denitrification and, with it, rapidly reduce the sediment oxygen consumption and increase its phosphorus binding potential. The nitrate is added by stirring it into the upper 15-20 cm sediment layer (*Ripl, 1976;* /7/). Subsequently, extra iron may be added if natural concentrations are low. The method has been criticised for not considering situations where phosphorus release is not redox dependent (*Petterson & Boström, 1982*). Also, the method in its original form is relatively harsh towards lake organisms, as several extreme values have been recorded (e.g. pH values as low as 3), but this may be remedied by simultaneous adding of calcium (*Ripl, 1976*). The method has only been sparsely used since its introduction, but experiments conducted by Willenberg *et al.* (1984) and full-scale investigations undertaken by Foy (1986), Donabaum *et al.* (1999) and Wauer *et al.* (2005) demonstrated positive effects and reduced phosphorus release pursuant to nitrate addition. The method has only rarely been applied in deep lakes (*Erlandsen et al., 1988*).

Nitrate has not been used in Denmark to restore lakes, but full-scale experiments to elucidate its applicability have been made in summer-stratified Lake Lyng (lake area: 10 ha, depth: 7.6 m) situated near the city of Silkeborg in Central Jutland (/7/) (Fig. 4.1). During two summers calcium nitrate ($Ca(NO_3)_2$) was added to approx. 10% of the lake area having water depths above 5 m. Each year the lake stratifies during summer, from ca. 1 June to 1 October with a hypolimnion at 3-5 m depth. The first year nitrate was added as dissolved nitrate, while pellets were used the second year. Smaller amounts than those of the original Riplox method were used, i.e. only 8-10 g N m^{-2} yr^{-1} compared to 141 mg N m^{-2} yr^{-1} in Swedish Lillesjöen (*Ripl, 1976*), and the added nitrate was therefore consumed before the autumn circulation. This procedure was used due to the poor water exchange and potential nitrogen limitation for parts of the year in Lake Lyng.

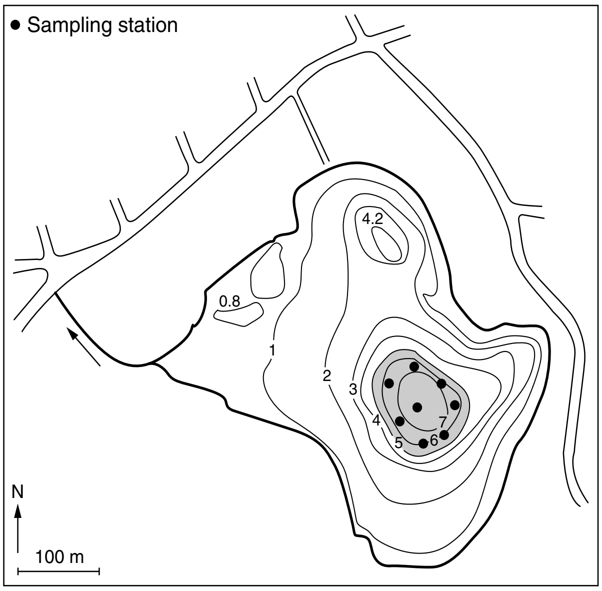

Figure 4.1 Depth map of Lake Lyng indicating the 8 sampling stations. From /7/.

Nitrate addition to Lake Lyng had marked effects on the accumulation of phosphate in the bottom water. In years without addition, concentrations as high as 2.9 mg P l^{-1} were recorded compared to 0.8 and 1.2 mg P l^{-1} during the two treatment years (Fig. 4.2). The effect was greatest when dissolved nitrate was added, probably because the nitrate pellets sank deeper down the soft sediment and therefore did not have the same impact on the surface sediment's capacity to bind phosphorus. Likewise, the iron concentrations were smaller in years with nitrate addition, which confirms the occurrence of the expected coupling between the sediment release of iron and phosphorus. The nitrate addition led to increased accumulation of ammonium in the bottom water, which contrasts the findings of other nitrate addition experiments (*Ripl & Lindmark, 1978; Foy, 1986; Erlandsen et al., 1988; Faafeng et al., 1997*). The enhanced accumulation may be an indication of enhanced mineralisation, but the possibility that the added nitrogen was reduced dissimilatorily to ammonium cannot be ruled out. In reduced environments, this process has been suggested to be more important than denitrification due to its provision of extra electrons (*Priscu & Downes, 1987*). The reduction to ammonium may be as high as 30-50% (*Kasper, 1985; Priscu & Downes, 1987; Downes, 1991*), and the increased accumulation of ammonium in Lake Lyng may, in principle, be ascribed solely to this mechanism (/7/).

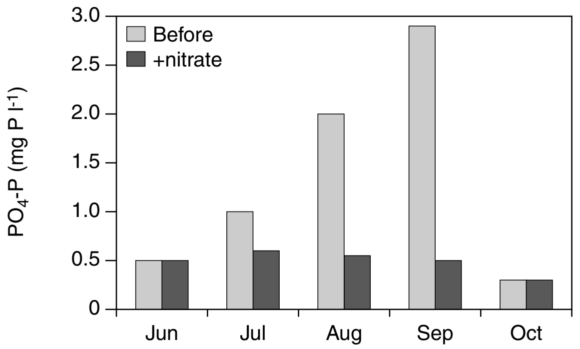

Figure 4.2 Concentration of orthophosphate in the hypolimnion during summer and autumn (6.5 m depth) in Lake Lyng without (before) and in a year (+nitrate) with addition of nitrate to the bottom water. Redrawn after /7/.

The importance of nitrate for the accumulation of phosphate in bottom water is also illustrated by measurements made in 29 m deep Danish Lake Knud showing inter-annual variations in bottom water nitrate concentrations. In years with nitrate concentrations below 0.1 mg N l^{-1} phosphate concentrations of 0.8-1.5 mg P l^{-1} were measured, whereas phosphate concentrations did not exceed 0.01 mg P l^{-1} in years with a nitrate level exceeding 1.3 mg N l^{-1} (*Andersen et al., 1980; Andersen 1994*).

Addition of aluminium and iron

Addition of iron (*Foy, 1985; Yamada et al., 1986; Boers et al., 1994*) and aluminium salts (*Kennedy & Cooke, 1982; Sonnichsen et al., 1997; Hansen et al., 2003*) has been used to enhance the binding potential of phosphorus and thus reduce lake phosphorus concentrations. The phosphorus-binding capacity of both iron and aluminium is good, but in contrast to the binding to iron that to aluminium is not redox-dependent. Aluminium may, however, have toxic effects at low alkalinity and pH values below 5.5 (*Cooke et al., 1993*). Negative effects on *Daphnia* have also been recorded (*Schumaker et al., 1993; Reitzel et al., 2003*). Several descriptions of aluminium addition can be found, but only few treat its long-term effects (*Lewandowski et al., 2003*).

In Denmark, the only full-scale experiment so far was conducted in Lake Sønderby (8 ha) on the island of Funen (*Reitzel et al., 2003*). Mesocosm experiments with aluminium addition showed a marked reduction of phosphorus and increased concentrations of NaOH-P in the sediment, and full-scale dosing was subsequently applied to the lake. Experiments with iron and aluminium addition have also been conducted in the bog Kollelev Mose on the island of Zealand

4.3 Biological methods

Various biological methods exist. Most are directed at the fish stock and aim either to 1) reduce the abundance of non-predatory fish by selective removal of zooplanktivorous species, or to 2) increase the zooplankton potential to graze down the phytoplankton, and thus create clear water, by the stocking of predatory species (*Shapiro & Wright, 1984; Benndorf et al., 1988*). Removal or stocking are the two most frequently used biological measures in Danish lakes.

Removal of non-predatory fish

In Meijer *et al.* (1999), Hansson *et al.* (1998) and Jeppesen & Sammalkorpi (2002) experiences regarding the use of biomanipulation as a restoration tool are summed up. A description of Danish examples can be found in Søndergaard *et al.* (1998a and /8/). Generally, the Danish and the international investigations exhibit widely different results. Long-term experience is limited; in Denmark the restoration of Lake Væng in Central Jutland, involving an approx. 50% reduction of the non-predatory fish stock, is the most well-documented example. Here, a total of 2.5 t fish were removed over a 2-3-year period, corresponding to 16 g m^{-2} (/14/, *Søndergaard et al., 1998a; Jeppesen et al., 1998c)* of which bream constituted 48% and roach 51%. The intervention generated substantial changes in the overall environmental state of the lake (Fig. 4.3): phytoplankton abundance decreased, the average size of zooplankton increased and Secchi depth improved.

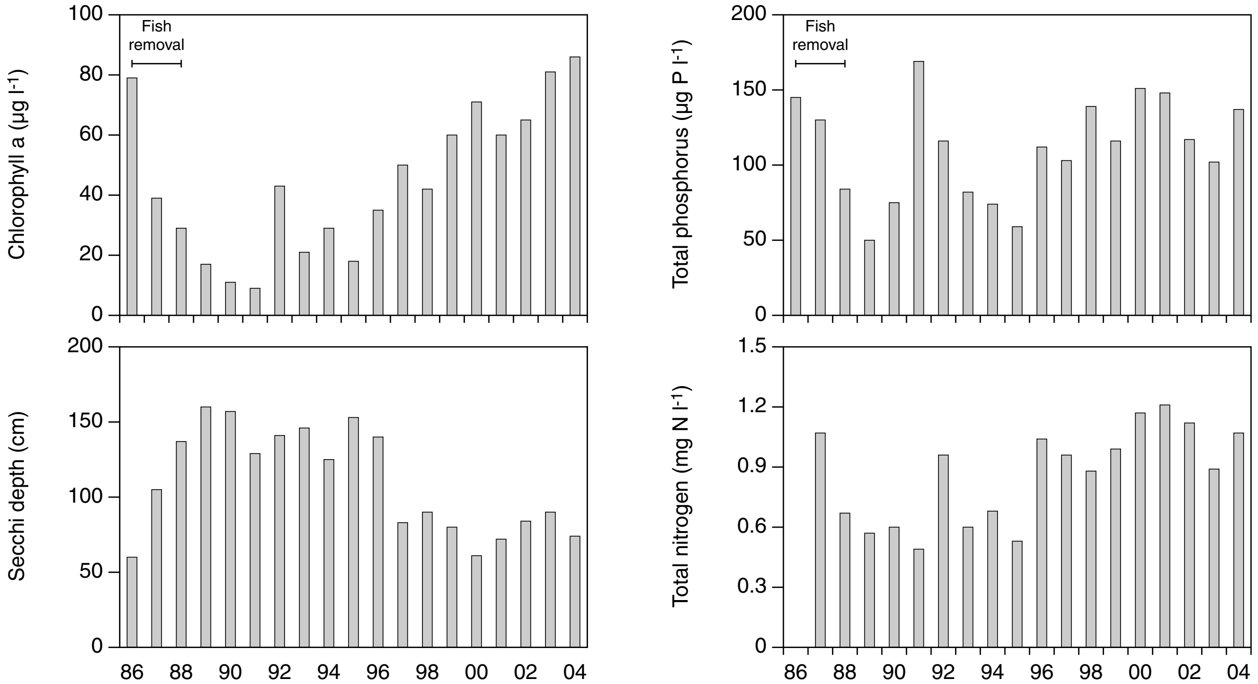

Figure 4.3 Summer average Secchi depth, chlorophyll *a*, total phosphorus and total nitrogen in Lake Væng from 1986 to 2004. Based on Søndergaard *et al.* (1997a, 1998a) and unpublished data.

After a period of 1-2 years submerged macrophytes arrived, covering in a few years the total lake area (Fig. 4.4). Initially, pondweed (*Potamogeton crispus*) was the dominant species but was later replaced by water thyme (*Elodea canadensis*), which became totally dominant covering most of the lake from sediment to surface. Concurrently with the increased expansion of submerged macrophytes, the number of waterfowl increased, not least that of herbivorous mute swan (*Cygnus olor*) and coot (*Fulica atra*). In years with high coverage of submerged macrophytes, as many as 300 mute swans and 800 coots were observed in the only 15 ha large lake. According to estimations, the birds were capable of removing up to 25% of the plant biomass in winter (*Søndergaard et al., 1997a*).

The intervention into Lake Væng had strong positive effects on the water quality for approx. a 10-year period (Fig. 4.3). After the intervention there have been wide fluctuations in nutrient concentrations, which may reflect a varying capacity to retain phosphorus at shifting turbidity. Large-scale variations may, in some years, also be attributed to the substantial changes in submerged macrophyte cover. Results from other lakes show that plant decay and decomposition may impact the internal recycling of phosphorus (*James et al., 2002*). The results from Lake Væng illustrate that fish stock intervention does not necessarily have lasting effects and that successive interventions may be needed to achieve stable conditions. The reasons for the only temporary effects despite the low external nutrient loading remain to be clarified. A likely explanation is the risk that a large pool of mobile phosphorus has been accumulated in the sediment and that this constitutes a potential risk of significant internal phosphorus loading and, with it, turbid water, unless the clearwater state is maintained. This indicates that the role of the sediment needs to be considered when assessing the use of fish stock interventions to obtain long-term positive effects. The establishment of water thyme as the dominant species in Lake Væng may also have contributed to the creation of unstable conditions, as water thyme is a species known to vary extensively in abundance from year to year (*Rørslett et al., 1985*).

Positive effects on water quality of fish stock interventions are not only obtained by lowering the predation pressure on the zooplankton. By reducing the density of benthivorous species, not least bream, also the predation pressure from fish on bottom invertebrates is released. This is of great significance to other species, for instance perch for which bottom invertebrates constitute an important food source. A large stock of bream may thus act as a competitive bottleneck for the invertebrate-feeding species (*Persson & Greenberg, 1990*) and may in the case of perch prevent it from reaching the predatory stage. Several of the Danish examples have shown that the growth rate of perch increases significantly at bream removal (*Søndergaard et al., 1998a*). Finally, removal of benthi-

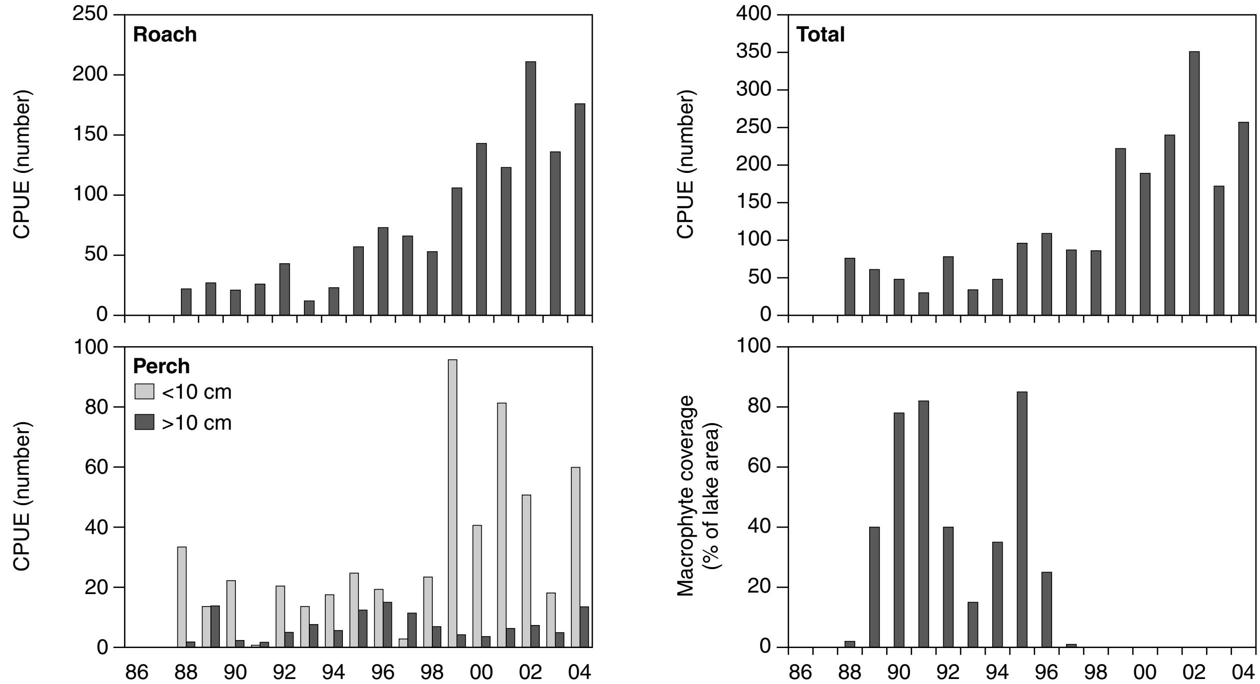

Figure 4.4 Development in fish abundance and composition and coverage of submerged macrophytes in Lake Væng after a 50% removal of the planktivorous fish stock in 1985-1988. Fish abundance is given per net per night (catch per unit effort, CPUE). Based on Søndergaard *et al.* (1997a, 1998a) and unpublished data.

vorous species such as bream and roach may have a positive impact on water transparency, because these species, when searching for food at the sediment surface, stir up particles in the water (*Breukelaar et al., 1994; Tátrai et al., 1997*) or increase the release of nutrients (*Brabrand et al., 1990; Havens, 1991*).

The best effects of non-predatory fish removal have been obtained when a large percentage of the total fish stock has been removed. If the portion removed is too small, there is a risk that the breeding and growth rates of the remaining stock will rise and thereby rapidly reach the same size as before. Therefore, minimum 70-80% of the non-predatory fish stock should be removed, preferably within a relatively short period (*Jeppesen & Sammalkorpi, 2002*). Jeppesen & Sammalkorpi (2002) also conclude that the fish biomass should be reduced to below ca. 100 kg ha^{-1} and that, depending on the existing total phosphorus concentration (TP µg P l^{-1}), 16.9 * TP$^{0.52}$ (kg ha^{-1}) should be removed to obtain positive results.

Stocking of predatory fish

As an alternative or supplement to the removal of zooplanktivorous fish species, predatory fish may be stocked (*Prejs et al., 1994; Berg et al., 1997; Skov & Berg, 1999; Skov et al., 2003a*). In Denmark, especially pike (*Esox lucius*) has been used, and in a few cases perch or rainbow trout (*Oncorhynchus mykiss*) (*Søndergaard et al., 1998b*). The objective of predatory fish stocking is similar to that of removal, i.e. to reduce the impact of fish predation on zooplankton. Normally 2-6 cm fry are used for pike stocking (*Berg et al., 1997; Søndergaard et al., 1997b)* with the purpose of limiting the abundance of roach and bream fry. Recently, pike stocking has been applied in the restoration of the inner lakes of Copenhagen (*Skov & Berg, 2003*).

One of the most comprehensive and earliest experiments with pike fry stocking was made in 10 ha large Lake Lyng at Silkeborg. During a 6-year period between 0 and 3600 pike fry were stocked per hectare to evaluate the effects on the other trophic levels. The impact of the different stocking densities was substantial and seemed to cascade to all trophic levels. In years with no or limited stocking, high abundance of zooplanktivorous species, low zooplankton biomass and high chlorophyll *a* concentrations were observed, the opposite being true in years with high stocking densities (*Søndergaard et al., 1997b*).

One of the problems associated with stocking of zooplanktivorous fish fry, such as pike, is that an effect is only obtained during the year of the stocking and, in principle, only for a brief period around the time of the stocking event. The sustainable stock of pike in a lake depends first and foremost on the extent of the littoral zone (*Grimm & Backx, 1990*), which means that an artificially high density of pike will rapidly decline. If density is high and habitat opportunity poor, mortality due to cannibalism will be high (*Skov et al., 2003b*).

The timing of pike fry stocking is therefore highly critical, and to obtain optimum effects stocking should be made immediately after the hatching of the benthivorous fish fry. As the time of hatching varies widely from year to year, optimisation may be tricky. Generally, restoration interventions using pike have yielded very variable results (*Grønkjær et al., 2004*). Likewise, massive stocking of pike fry seems to be required to generate adequate effects (*Søndergaard et al., 1997b*). A recent analysis of the experiences arising from pike stocking in 8 Danish lakes suggests a generally poor impact of the stockings (*C. Skov, personal communication*).

Transplantation and protection of submerged macrophytes

Submerged macrophytes have a number of positive effects on the water quality of shallow lakes and play a central role for the maintenance of a clearwater state (*Scheffer et al., 1993; Jeppesen et al., 1997b; Jeppesen, 1998*). Therefore, in shallow lake restoration it is of vital importance to ensure an adequate and stable coverage of submerged macrophytes, as illustrated by for instance the Lake Væng experiments.

Via a series of direct and indirect mechanisms submerged macrophytes affect strongly also the phytoplankton communities of shallow lakes (/10/): submerged macrophytes create a structure boosting the abundance of sessile or plant-associated and pelagic zooplankton species and thereby generate a higher grazing pressure (*Timms & Moss, 1984; Stansfield et al., 1997; Lauridsen et al., 1997*); changed nutrient availability in the presence of plants may influence both the abundance and composition of phytoplankton; allelopathic effects of species such as *Chara* or *Stratiotis* are suggested in several investigations (*Brammer, 1979, Wium-Andersen, 1982; Mulderij et al., 2003*); sedimentation increases due to more stagnant water *(Barko & James, 1997*); and shading effects become more important (*Sand-Jensen, 1989*). Thus, normally, considerably lower abundance of phytoplankton is observed in the presence of submerged macrophytes (Table 4.2). Also the average size of some phytoplankton species may be impacted by the presence or absence of submerged macrophytes (Fig. 4.5).

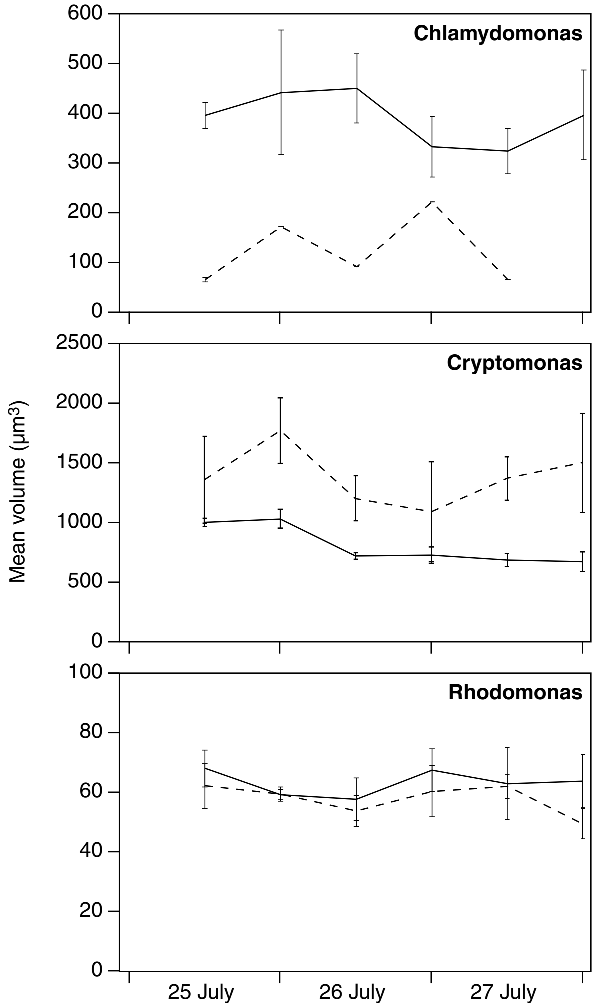

Figure 4.5 Differences in the biovolume of three phytoplankton species (*Chlamydomonas* sp., *Cryptomonas reflexa*, *C. curvata* and *C. marssonii*, as well as *Rhodomonas* sp.) in enclosures holding submerged macrophytes (broken line) and without submerged macrophytes (full line). The experiments were undertaken as triplicates (average values and standard error given) in 20 m^2 enclosures in Lake Stigsholm from 25 to 27 July 1994 with samplings at noon and midnight. From /10/.

Within a restoration context, transplantation of submerged macrophytes has been used to ensure a stable clearwater state (*Donabaum et al., 1999; Lauridsen et al., 2003*). This is particularly relevant in situations where immigration and spreading of

Table 4.2 Chlorophyll *a* concentrations (µg l^{-1}) and phytoplankton biomass (mm^3 l^{-1}) day and night in enclosures with 50% plant coverage and without plants. Averages from three enclosures are shown (±SD in parenthesis) from 1-3 days of sampling. From /10/.

	With submerged macrophytes		Without submerged macrophytes	
	day	night	day	night
Chlorophyll *a*	17 (19)	8.3 (4.0)	64 (10)	37 (7.9)
Biomass	0.80 (0.78)	0.31 (0.21)	8.1 (1.4)	7.6 (2.1)

plants are limited by lack of spreading potential or when herbivorous birds such as coot and mute swan diminish plant abundance and in some instances actively prevent plant establishment and spreading (*Mitchell & Wass, 1996; Søndergaard et al., 1996*). The setting-up of a wire fence around existing or transplanted plants may be a means to improve plant conditions (*Jeppesen et al., 1997a; Lauridsen et al., 2003*).

5 Perspective and future challenges

As illustrated in the previous chapters, the external nutrient input and the internal interactions between water and sediment play a decisive role for the water quality of lakes. Even though the environmental state of Danish lakes has improved within the last 10-20 years, development of tools to reduce external as well as internal loading will remain key research areas in the future in the attempt to obtain a satisfactory water quality. However, also new management and climate issues need to be placed on the agenda.

5.1 Nutrients and water quality

Since the input from most nutrient point sources has been reduced to a minimum, but the objectives set for an equilibrium state are still far from being met in many lakes, focus should now be directed at the more diffusive nutrient sources of which the loading from agriculture and scattered dwellings is the most significant. During the period 1989-95 to 1996-2002, relative phosphorus loading from diffuse sources increased from 47% to 57% (*Jensen et al., 2004*). This implies that if serious attempts should be launched to lower the phosphorus loading to lakes, a reduction of the agricultural input is a prerequisite (Fig. 5.1). The share of nitrogen originating from diffuse sources (agriculture + nature) has remained almost constant since 1989 and constituted 72% of the total lake input in 2003 (*Jensen et al., 2004*).

The relative increase in the diffuse contribution of phosphorus is not necessarily due to decreased point source loading, but may also be caused by increased runoff from agricultural areas. During the 20th century there has been a net input of phosphorus to agricultural soil (Fig. 5.2). The phosphorus surplus peaked around 1980 by 30 kg phosphorus per hectare, but had declined to approx. 15 kg phosphorus per hectare in 2000 (*Nielsen et al., 2005*). A similar pattern has been observed in other countries, for instance in the catchment of Lough Neagh in Northern Ireland (*Foy et al., 1995*) and in Ireland (*Jennings et al., 2003*). This complex of problems will probably remain relevant also in the future, as there is still – also after the implementation of the latest Action Plan on the Aquatic Environment – a net input of phosphorus, and thus accumulation, to the agricultural soil and thereby an increased risk of runoff.

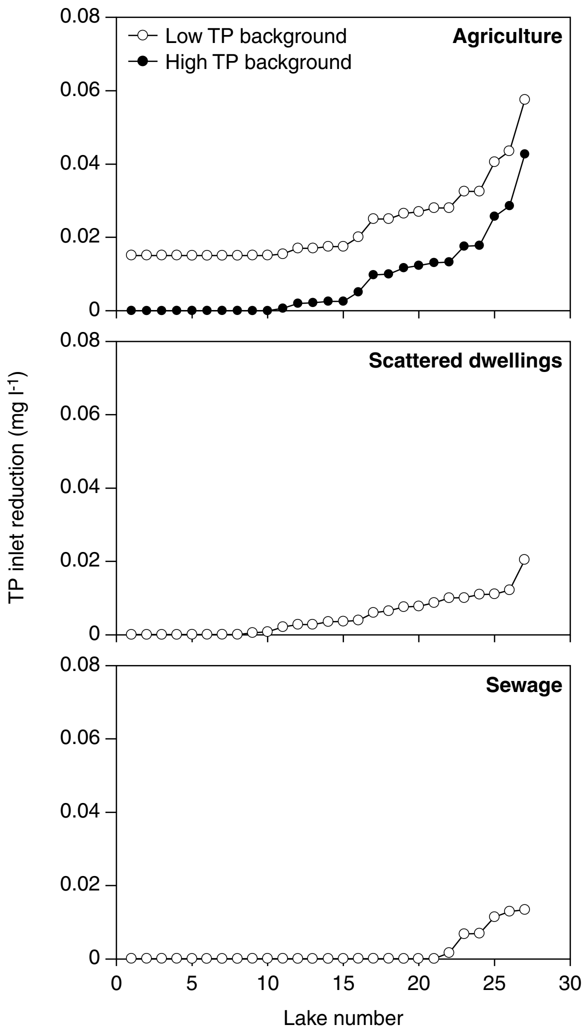

Figure 5.1 Reduction of inlet concentrations of total phosphorus in 27 monitoring lakes to be obtained if the present (2002) loading from, respectively, agriculture, scattered dwellings and sewage is decreased by 50%. For the agricultural input two scenarios are shown, one with a natural input of 55 µg P l^{-l} (high TP background) and one with 25 µg P l^{-l} (low TP background). The lakes are categorised according to increasing reduction in inlet water and are not shown in the same order in the three partial figures. From Søndergaard *et al.* (2003b).

The internal phosphorus loading will gradually decrease as the lakes approach a state of equilibrium with the external loading, and in lakes where internal loading is decisive for water quality it is therefore just a question of time when the improvement occurs. In some lakes, however, this transition period may endure for a long time, as illustrated in section 2.4. Therefore, in the future internal phosphorus release may

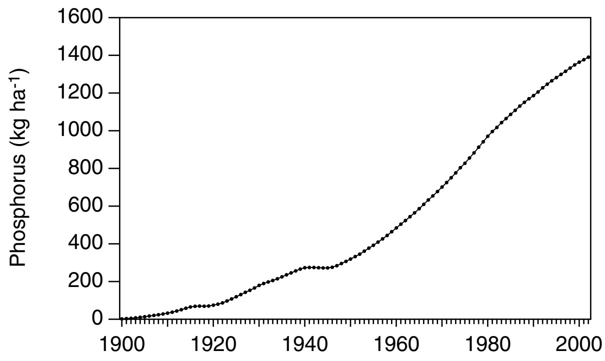

Figure 5.2 Accumulation (net surplus) of phosphorus in Danish agricultural soil during the past 100 years. From Nielsen et al. (2005b**).**

need to be considered a serious threat – not least in lakes where a large pool of phosphorus has been accumulated and in lakes where the mobile phosphorus pool is only slowly reduced.

Also the interactions between the individual biological components and their influence on the chemical conditions have proven to affect the state of lakes in multiple ways. The availability of nutrients may set the scene, but the roles and the environmental state are determined by the different biological components and their interactions. Furthermore, there may, as mentioned before, be strong interactions where also changes in biological conditions influence nutrient conditions. Seen in a larger perspective, retention of both nitrogen and phosphorus can be improved if the number of clearwater lakes is increased. This would reduce the nutrient loading of downstream wetlands, including fjords and estuarine areas

5.2 Climate effects

More and more indications show that the global climate is changing, the reasons for and the degree of the changes still being subject to intensive debate. The future climate changes may influence the environmental state of lakes and possibly counteract the initiatives taken to improve water quality. The effects arising from enhanced temperatures and increased precipitation are predicted to be of particular importance (*Jeppesen et al., 1992*).

Increased precipitation and potentially also more extreme weather events with exceptionally heavy precipitation will lead to higher nutrient runoff from catchments and, with it, increasing eutrophication (*Jeppesen et al., 1992*). The interdependence between nutrient runoff and precipitation is clearly demonstrated by the results of the lake monitoring programme showing that differences in precipitation lead to variations in the input of both phosphorus and nitrogen to the Danish lakes (*Jensen et al., 2004*). Effects of changed runoff were also seen in marine waters where marked improvements were recorded (reduced nitrogen concentrations, decreased algal production and higher oxygen content) in the two dry years of 1996 and 1997 (*Christensen et al., 2004*).

Exactly which direct effects are to be expected from increased temperatures on the biological state of the lakes are subject to discussion. Scheffer *et al.* (2001) suggested that climate warming may contribute to the recolonisation of submerged macrophytes and thus to the maintenance of a clearwater state. In contrast, Jeppesen *et al.* (2003b) predicted negative effects on water quality by climatic changes due to increased abundance of heat tolerant zooplanktivorous and benthivorous fish species such as carp (*Cyprius carpio*). Also Genkai-Kato & Carpenter (2005) foresee negative effects because increasing temperatures via increased internal phosphorus loading can reduce the critical phosphorus loading needed to initiate ecosystem regime shifts.

Increased temperatures may also impact the seasonal retention and release of phosphorus. Based on investigations in Bay of Quinte, Lake Ontario, showing a close interaction between temperature and summer concentrations of phosphorus, Nicholls (1999) concluded that a temperature increase of 3-4 ^{o}C may almost double summer total phosphorus concentrations. The preliminary results of 24 Danish mesocosm experiments simulating a 3-5 ^{o}C temperature increase within the next 100 years suggest that the duration of the winter period with phosphorus retention will decrease, triggering an earlier onset of internal loading (*Liboriussen et al., 2005; Søndergaard et al., unpubl.*). However, increased temperatures may also lead to less pronounced variations in phosphorus concentrations since reduced winter retention implies a reduced potential for later phosphorus release. This may well be the explanation of why significant seasonal variations between summer and winter values of phosphorus do not occur in nutrient-rich lakes located in areas with warmer climates, such as southern Europe and Florida (*Jeppesen et al., in prep.*).

Finally, more extreme events such as mild winters with short-term ice coverage, absence of winter fish kills, and exceptionally cold or windy periods may alone or in conjunction establish conditions unfavourable to submerged macrophytes, and thereby generate a shift from the clear to the turbid state (*Hargeby et al., 2005*). Increased appearance of catastrophic or stochastic events may thus enhance the risk of sudden and drastic switches between the clear and the turbid state (*Scheffer et al., 2001*).

5.3 The Water Framework Directive and future lake management

In a densely populated and intensively agricultivated country such as Denmark, lakes will easily receive more nutrients than only those deriving from natural loading. Nevertheless, the demand of the EU Water Framework Directive (*WFD; European Union, 2000*) is that all lakes must reach "good ecological status" by latest 2015, this being a state that only slightly deviates from the unimpacted state (the socalled reference state). This constitutes a major challenge to the future management of lakes.

The WFD operates with five different ecological classes deviating to a varying degree from the reference state (Fig. 5.3). This has given rise to much debate as the determination of a reference state has proven a difficult task. Various methods may be used: palaeolimnological analyses, identification of the characteristics of unimpacted sites, historical data, modelling, expert judgement – or a combination of these (*Laird & Cumming, 2001; Gassner et al., 2003; Nielsen et al., 2003*). However, defining reference conditions is problematic given the often limited availability of data and high natural variability. Moreover, it is debatable how far back in time we should go to find minimally impacted conditions as lakes often undergo gradual change over time, as demonstrated by palaeolimnological studies (*Bradshaw, 2001; Johansson et al., in press*). Recent studies reveal that it may be extremely difficult to find minimally impacted lakes to act as reference sites (*Bennion et al., 2004*). Finally, it may be discussed whether a single reference condition can, in fact, be defined, as all lakes are continuously impacted by variations in for instance climate, while simultaneously undergoing a succession from, by way of example, deepness to shallowness (*Amsinck et al., 2003; Søndergaard et al., 2004*).

The five ecological classes are to be defined from biological indicators supported by water chemical and hydromorphological elements. The WFD mentions phytoplankton, macrophytes, invertebrates and fish as elements to be used in the classification. The directive is, however, not very specific and provides only general guidance on how to define the proposed ecological classes (*Wallin et al., 2003*). One of the major and more practical challenges for implementation of the directive is therefore how to define and determine the ecological status of a specific waterbody. To pave the way for the implementation, an analysis was conducted comprising a number of water chemical and biological characteristics of 700 Danish lakes (/2/; *Søndergaard et al., 2003c*).

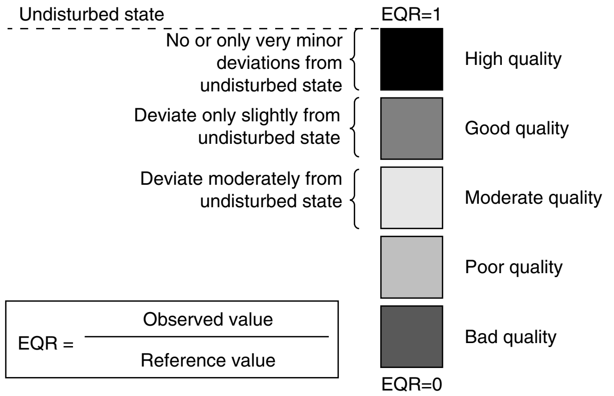

Figure 5.3 Survey of the five ecological classes of the Water Framework Directive. EQR (Ecological Quality Ratio) is a figure between 0 and 1 and is the ratio between the observed value of the ecological state under conditions without anthropogenic impact. From Wallin *et al.* (2003).

In the analysis, total phosphorus was selected as a key variable for lake water quality. This neglects the philosophy of using the reference state to define a present ecological state, but as mentioned in the introduction (section 1.1) TP is the main environmental stressor and the primary determining factor for numerous biological variables and is also used in present-day lake classification (*Vollenweider & Kerekes, 1982; Wetzel, 2001*). In the analysis 22 preselected ecological variables often used in lake monitoring were ordered along a TP gradient in order to trace their potential applicability for ecological classification.

As a preliminary classification we applied a TP-based division into high, good, moderate, bad and poor ecological quality using 0-25, 25-50, 50-100, 100-200 and > 200 $\mu g\ P\ l^{-1}$ boundaries for shallow lakes and 0-12.5, 12.5-25, 25-50, 50-100 and > 100 $\mu g\ P\ l^{-1}$ boundaries for deep lakes. Within each TP category, median values were then used to define preliminary boundaries for the biological indicators. The analysis showed that most of the 22 indicators investigated responded strongly to increasing TP; there were only minor differences between low and high alkalinity lakes and modest variations between deep and shallow lakes (Fig. 5.4). Yet, the variability

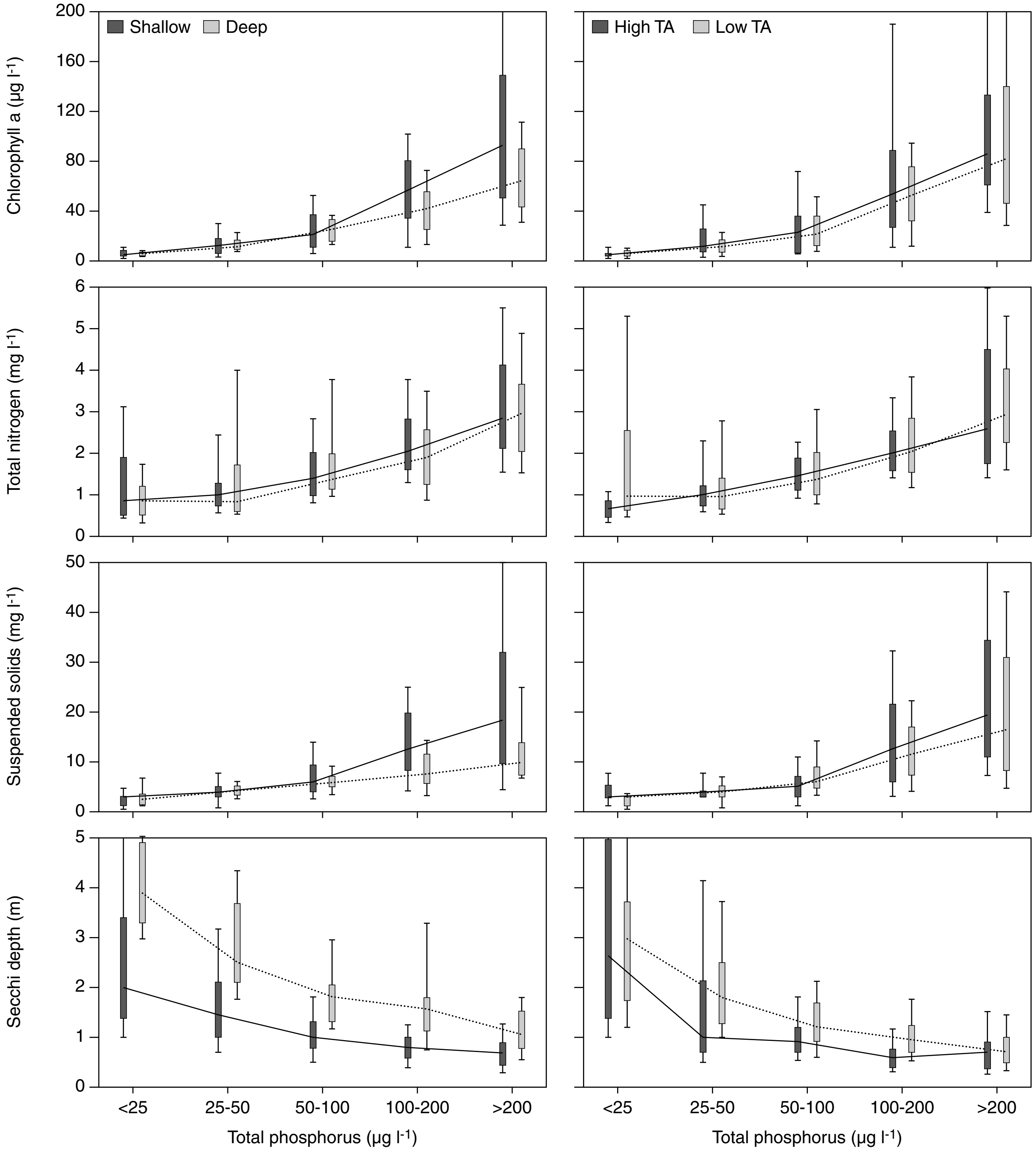

Figure 5.4 Boxplots of chlorophyll *a*, total nitrogen, suspended matter and Secchi depth for five phosphorus categories divided into shallow (mean depth < 3 m) and deep lakes (mean depth ≥ 3 m) and in lakes with low (≤ 0.2 meq l^{-l}) and high (> 0.2 meq l^{-l}) alkalinity. Each box shows 25 and 75% quartiles, and upper and lower lines give 10 and 90% percentiles. Mean values are connected by lines. Data from 451-631 Danish lakes are included. From /2/.

of indicators within a given TP range was strong, and for most indicators there was a considerable overlap between adjacent TP categories (Fig. 5.5). Cyanophyte biomass, submerged macrophyte coverage, fish numbers and chlorophyll *a* were among the "best" indicators, but their ability to separate different TP classes varied with the TP level.

The analysis also showed that when using multiple indicators the risk that one or more of these will point towards different ecological classes is high due to the high variability between all indicators within a specific TP class. Consequently, the "one out–all out" principle, implying that lacking fulfilment of only one criterion demands complete rejection, as suggested in the WFD context, seems inapplicable here. Alternatively, a certain compliance level or a "mean value" of the indicators can be used to define ecological classes. A precise ecological quality ratio (Ecological Quality Ratio, EQR) using values between

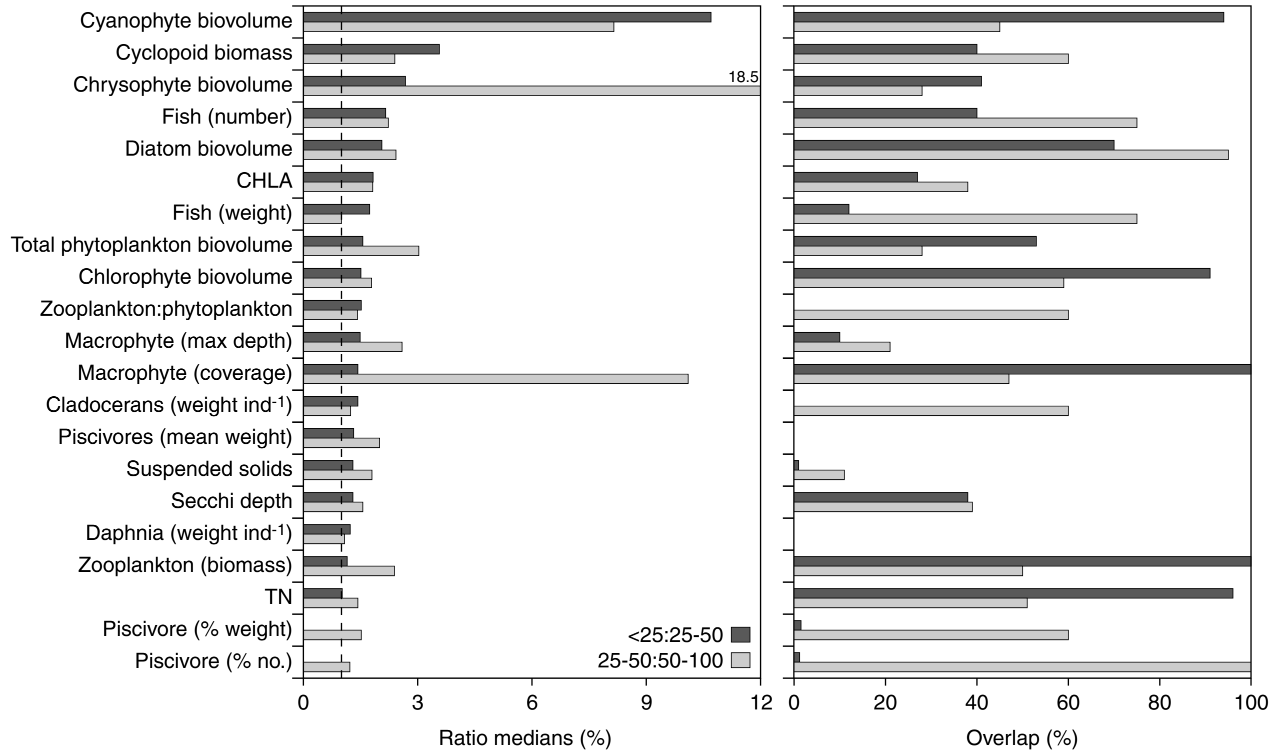

Figure 5.5 Left part: The ratio between the median values for the 0-25:25-50 µg P l^{-1} and the 25-50:50-100 µg P l^{-1} phosphorus classes for 21 potential ecological indicators. The broken line shows a ratio of 1, i.e. a situation where there is no difference between the median values for the two phosphorus categories. Right part: the overlap between adjacent phosphorus classes (0-25 and 25-50 and 25-50 and 50-100 µg P l^{-1}) for 19 indicators. The degree of overlap is based on the 25-75% quartiles and expressed as % overlap with the following phosphorus class. From /2/.

tween 0 and 1 can be calculated based on the extent to which the total number of indicators meet the boundary conditions. The latter was demonstrated for three Danish lakes showing that the calculated EQR traced the changes in phosphorus, chlorophyll *a* and Secchi depth relatively well (Fig. 5.6). However, overall the analysis demonstrates that the implementation of the WFD faces several challenges, such as gradual rather than stepwise changes for all indicators, large variability of indicators within lake classes and problems regarding the use of the "one out – all out principle" for lake classification.

The objectives of the WFD will probably lead to significant changes in Danish lake management, as the system currently in operation varies considerably from county to county (Fig. 5.7; *Søndergaard et al., 2003c*). By way of example, the present maximum concentration of phosphorus to meet the given objectives for the 27 freshwater lakes included in the Danish lake monitoring programme varies between 20 and almost 200 µg P l^{-1}. In contrast, our suggested boundary between good and moderate ecological state elaborated on the basis of the WFD requirements is 25 or 50 µg P l^{-1}, depending on water depth (/2/). The lacking uniformity of county objectives renders it difficult to compare fulfilment of objectives between Danish lakes and areas.

5.4 Future need for knowledge

Knowledge about the functioning of the interactions between biological structure and nutrient loading in lakes has increased considerably during the past 10-20 years, but is, however, still incomplete seen from both a basic scientific and user-oriented aspect.

One of the areas requiring further study is the role of nitrogen. There is a growing awareness that not only phosphorus but also nitrogen plays an important role if clearwater conditions are to be obtained and maintained in our lakes. The mechanisms behind and the threshold levels for phosphorus and nitrogen triggering the two alternative states of equilibria remain to be established. Thus, in future lake management both substances must be considered as there may well be situ-

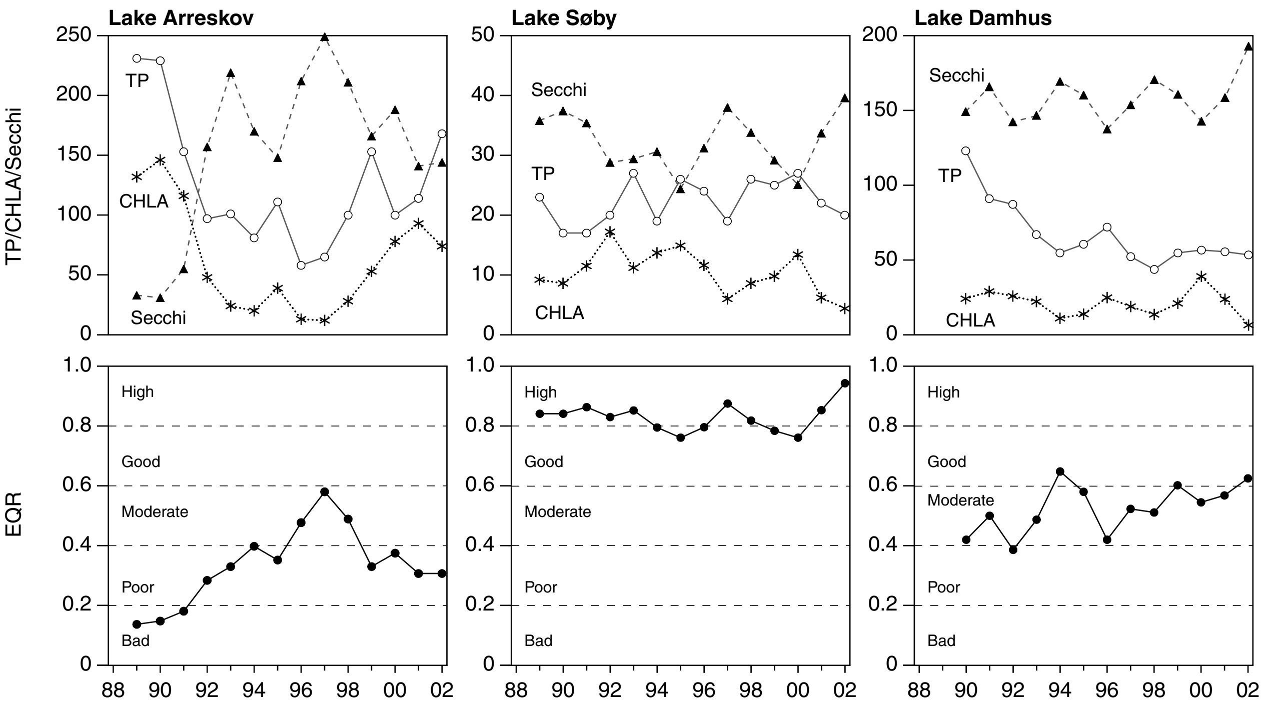

Figure 5.6 Examples of calculation of EQR (Ecological Quality Ratio) from three Danish lakes from 1989 to 2002 based on the use of 22 ecological indicators (five from phytoplankton, zooplankton, fish and water chemical parameters and two from submerged macrophytes). All indicators were equally weighed in the calculations. The upper panels show summer means of total phosphorus (µg l^{-l}), chlorophyll *a* (µg l^{-l}) and Secchi depth (in cm for Lake Arreskov and Lake Damhus and in dm for Lake Søby).

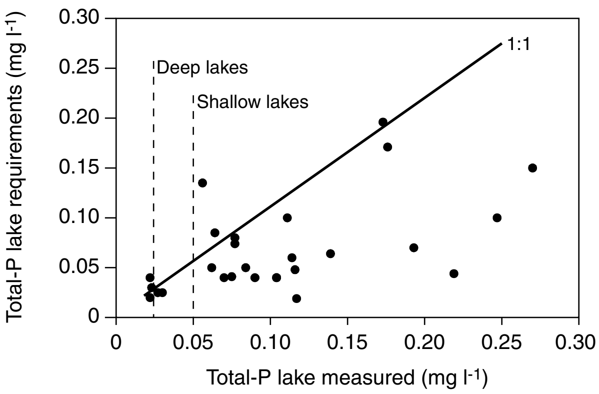

Figure 5.7 Required phosphorus concentrations relative to measured total phosphorus concentrations in 27 monitoring lakes. The required values are calculated based on the objectives set by the counties for each individual lake. The measured values are mean values from 2000-2002. Broken lines are the suggested limit between good and moderate ecological state for shallow and deep lakes, respectively. From Søndergaard *et al.* (2003b).

ations where a reduction of the nitrogen input may serve as a supplementary tool to achieve a satisfactory water quality. In connection with this, also the roles of the sediment and the benthic-pelagic coupling merit further investigation. Which mechanisms generate the strong variations observed in nutrient retention and may permanent retention be optimised? The predicted climatic changes and their impacts on the aquatic environment are yet uncertain, but enhanced temperatures and precipitation may require the implementation of stricter nutrient reduction levels than hitherto anticipated.

There is also a general need for considering lakes as elements of the catchments and downstream water areas. This applies both when attempting to increase biodiversity via the establishment of more closely connected wetlands, and when reducing the phosphorus and nitrogen input to both freshwater and marine areas. When establishing new or re-establishing former lakes it is essential to ensure clearwater conditions to optimise nutrient retention, but how to achieve such optimisation remains an open question. Further studies will help resolve whether it is actually possible to create and maintain aquatic areas that both live up to the demand for high biological value and the need for good retention capacity (*Hansson et al., 2005*).

So far, lake research has mainly focused on large lakes, investigations in small lakes and ponds being less extensive and often focused on a few selected biological variables. This may not be optimal given that small lakes are completely dominant in number in the Danish landscape and contribute substantially to the biological diversity with their scenic landscape values and high spreading potential. However, the ecological understanding of how this biotype functions and reacts relative to its catchment is incomplete, and although NOVANA will certainly expand our knowledge, an integrated view of the structure and functioning of our small aquatic areas is needed.

Lake restoration will also in the future be used as a measure to obtain improved water quality. This also applies relative to the WFD whose objective of "good" ecological state may otherwise be difficult to achieve. However, lake restoration is still surrounded by a certain portion of "trial and error" despite that some overall guidelines exist. Long-term effects have only been sparsely elucidated, and how each individual lake will react is difficult to predict (*Genkai-Kato & Carpenter, 2005*). New solutions keep popping up, and consideration of alternative restoration measures may be relevant, including the question of whether the solutions should be permanent or applied repeatedly by lake managers as a tool to obtain stable clearwater conditions.

In conclusion, the challenges within lake research and user-oriented aspects remain daunting. The interaction between research, monitoring and advising has proven very fruitful, benefiting all three areas. It is a strategy that ought to be maintained and possibly even expanded in some cases in the future. Hopefully, this dissertation and the associated articles have helped emphasise this.

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7 List of associated papers

NERI **National Environmental Research Institute**

DMU Danmarks Miljøundersøgelser

National Environmental Research Institute, NERI, is a part of University of Aarhus.

NERI's tasks are primarily to conduct research, collect data, and give advice on problems related to the environment and nature.

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This dissertation including 16 selected English papers and book contributions was accepted by the Faculty of Science, University of Aarhus, for the doctor's degree in natural sciences. The dissertation describes the dynamic interactions between nutrients, the sediment and the biological conditions in lakes. First, Danish lake types are characterised and their development described relative to changes in nutrient loading. After this a more detailed description is given of lake retention of nutrients, including the binding of phosphorus in the sediment and the exchange of phosphorus between the sediment and the water phase. Next follows a section on the multiple mechanisms behind the release and uptake of phosphorus in the sediment. As a user-oriented aspect, lake restoration and the results obtained from the various chemical and biological methods applied in Denmark so far are treated. Finally, reflections are made on future management and research issues for Danish lakes, including the future climate and the implementation of the EU Water Framework Directive.

Doctor's dissertation (DSc) Nutrient dynamics in lakes

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