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Impacts of chemical precipitation of phosphorus with polyaluminum chloride in two eutrophic lakes in southwest Finland

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ABSTRACT
In an attempt to improve water quality in 2 eutrophic shallow Finnish lakes, Kirkkojärvi and Littoistenjärvi, phosphorus precipitation with polyaluminum chloride was performed in June 2002 and May 2017, respectively. Here we compare the effects of the chemical treatment between the lakes to enhance our understanding of the mechanisms involved and to improve the predictability of similar management actions in the future. All plankton was killed in the treatment, but phytoplankton recovered in 4 weeks and crustacean zooplankton in 2 months. Because removal fishing had not been successful, the chemical dosage in Kirkkojärvi was intentionally set so high that the treatment killed all fish. In Littoistenjärvi, pH was adjusted so that most fish survived. In Kirkkojärvi, the summer phosphorus (TP) and chlorophyll (Chl-*a*) concentrations 3 years after treatment dropped by 85% and 88% compared to those recorded 3 years before treatment. Cyanobacterial biomass declined by 88%, with only occasional blooms appearing in 3 of 22 years. The average TP and Chl-*a* of the post-treatment period 2006–2020 indicated substantial improvement in the ecological state from "bad" to "moderate" rating of the EU Water Framework Directive (WFD). In Littoistenjärvi, the corresponding declines due to the Al treatment were 72% in TP and 87% in Chl-*a* concentration, and 92% in cyanobacterial biomass. Longevity of treatment effects was estimated using the upper boundaries of the WFD quality classes as the target values. Water quality changes followed the internal loading of TP, affected by temperature and pH.

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chlorophyll *a*; eutrophication; lake management; shallow lakes; total phosphorus

Introduction

Eutrophication of lakes leading to toxic cyanobacterial blooms is a general problem for human water use worldwide (Schindler 2006, Heisler et al. 2008, Paerl and Otten 2013, Huisman et al. 2018, Ho et al. 2019). Reducing excessive external nutrient loading is the principal remedy to improve water quality (Jeppesen et al. 2005, 2007, Schindler 2006, Hamilton et al. 2016), but when internal loading (release of nutrients from the sediment back to water) becomes the major source of nutrients, particularly the case in many shallow lakes, the restoration of acceptable water quality can take decades (Marsden 1989, Sas 1990, Jeppesen et al. 2007, Søndergaard et al. 2013, Chorus et al. 2020, Steinman and Spears 2020). The shifts between turbid and clear water often show a hysteresis effect, so that when the nutrient load is decreasing, the critical nutrient level for shifting from the turbid to the clear-water state is lower than the level leading to a shift to the turbid state under increasing nutrient levels (Scheffer 1998).

The ecosystem of turbid, phytoplankton-dominated shallow lakes is resistant to change and requires a strong external intervention to return to a clear-water state (Ibelings et al. 2007, Lürling et al. 2020, Sarvala et al. 2020). Potential methods to manage internal phosphorus (P) loading in small and shallow waterbodies include sediment excavation or dredging, both of which are expensive, have strong environmental impacts, and often provide only temporary improvements in water quality (Lürling et al. 2020). Hypolimnetic withdrawal can work only in stratified lakes, where aeration and oxygenation are most useful. Hypolimnetic oxygen enrichment is one of the most common lake restoration methods, but it often yields minor effects (Tammeorg et al. 2017). Biomanipulation, usually as removal of planktivorous and benthivorous fish, can improve water quality but must be repeated for maintenance (Jeppesen et al. 2017).

Most of these methods have been inefficient. In recent years, increasing numbers of eutrophic shallow lakes in

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urban environments have been restored by chemical precipitation of P (Cooke et al. 2005, Zamparas and Zacharias 2014, Jensen et al. 2015, Mackay et al. 2015, Araújo et al. 2016, Huser et al. 2016a, 2016b). Optimally, chemical treatment can lead to immediate water quality improvement, but not always, and the duration of the positive results is highly uncertain. Such restoration attempts continue to be experimental (Mackay et al. 2015), and therefore, to advance our understanding and to improve predictions of ecosystem responses, well-documented case studies are still needed.

By the 2000s the water quality in 2 small and shallow lakes in southwest Finland, Kirkkojärvi (Rymättylä, to distinguish it from several other lakes with the same name) and Littoistenjärvi, had deteriorated so badly that the lakes were no longer suitable for recreational or other human uses. Both lakes were subject to a number of restoration measures, but these were insufficient to improve water quality. As a final option to improve the ecological state in both lakes, P in water and surface sediment was precipitated with polyaluminum chloride. Kirkkojärvi was treated in May and June 2002 and Littoistenjärvi in May 2017 (Sarvala et al. 2020).

Here we examine the results of the chemical restoration during the 20 years subsequent to the treatment of Kirkkojärvi and 6 years subsequent the treatment of Littoistenjärvi, thus extending the follow-up period of Littoistenjärvi from 2 years in Sarvala et al. (2020) to 6 years. We also assess the longevity of the treatment to estimate when a renewed treatment might be appropriate.

Study lakes

In Kirkkojärvi, eutrophication was initially due to historical sewage loading as well as to diffuse loading from agriculture. Arable fields comprise almost one-third of the Kirkkojärvi catchment but less than one-tenth of the Littoistenjärvi catchment (Table 1; Sarvala et al. 2020). In the late 1990s and early 2000s, Kirkkojärvi became hypertrophic, and internal loading was the dominant source of nutrients in summer (Sarvala et al. 2000). In the EU Water Framework Directive (WFD) national classification, Kirkkojärvi has been ascribed to nutrient-rich lakes and Littoistenjärvi to shallow nonhumic lakes. Here, however, Littoistenjärvi is regarded as a nutrient-rich lake as well, and the official process for a change in lake type has been initiated. The Littoistenjärvi ecosystem was in similar condition in the early 1900s (Wahlberg 1913) and in 1983 (Sarvala et al. 2020), and paleolimnological data also support the view that Littoistenjärvi would be naturally eutrophic (Glückert et al. 1992). In the late 1980s and the 1990s, cyclic mass occurrences of the invasive submerged plant *Elodea canadensis* dominated the system (Sarvala et al. 2020). At the end of each 5–6-year cycle, the mass death of the plants rendered the lake as a turbid, phytoplankton-dominated nutrient-rich system for 1 summer. In the following 3–4 years, the lake was an almost oligotrophic clear-water system dominated by submerged plants. Towards the end of the cycle, macrophyte abundance led to extremely high pH (up to 11) and ensuing release of P in water. Anoxia in winter was prevented by aeration, except for winter 1998–1999 when the aerators were broken. After the third, and so far last, population collapse of *E. canadensis* in 1999, the plants remained scarce. Between 2003 and 2006 the lake shifted to a turbid, phytoplankton-dominated hypertrophic system and remained in this state. Scarcity of macrophytes in the 2000s was associated with strongly increased internal loading while external loading was moderate or reduced (Sarvala et al. 2020).

Table 1. Basic characteristics of the study lakes.

	Littoistenjärvi	Kirkkojärvi (Rymättylä)
Coordinates	60.45°N, 22.39°E	60.37°N, 21.93°E
Area (ha)	150	44
Mean depth (m)	2.2	2.9
Maximum depth (m)	3.0	4.8
Drainage area (ha)	389	314
Drainage area:lake area	2.59	7.14
Osgood index[§]	1.80	4.37
Arable fields (% of watershed)	7.5	30.6
Water residence time (a)	1.8	0.9
External TP loading (g m^{-2} a^{-1})	0.036[+]	0.38*
Internal TP loading (g m^{-2} a^{-1})	0.13[++]	3.31* (1.10**)

[§]Osgood index = mean depth (m) divided by the square root of lake area (km^2). [+] Before the chemical treatment (Sarvala et al. 2020). [++] Net internal load calculated as the difference in total phosphorus (TP) between the maximum observed in 1 July to 15 September and the minimum in March–May (Sarvala et al. 2020). * From phosphorus budget before chemical treatment (personal communication, M. Tarvainen, Uusimaa Centre for Economic Development, Transport, and the Environment). ** Net internal loading before treatment.

Methods

Chemical treatment

By the early 2000s, the ecological state in both lakes was deemed “bad” in the quality classification of the WFD (Aroviita et al. 2019). All feasible restoration methods were attempted, including aeration (Littoistenjärvi only) and the reduction of external loads and fish populations (Sarvala et al. 2020). These interventions were not sufficient to improve water quality, however, and chemical restoration was applied as a last option.

In Finland, chemical restoration requires an environmental permit granted by the Regional State Administrative Agency. Conditions for the granting permit are strict, stipulating that the operations not cause harm to health or significant environmental pollution or a risk of such pollution. The permit may contain special orders and instructions concerning the treatment itself (e.g., timing, target pH) and post-treatment monitoring. The permit for Littoistenjärvi required that, to save the fish, the pH should not be <6.0. By contrast, the permit for Kirkkojärvi allowed lower pH to result in death of the fish.

Polyaluminum chloride (PAX-XL100, Kemira Oyj), the restoration chemical, was distributed to the whole lake by driving along parallel transects with special boats equipped with 5 m perforated side-pipes that dripped the liquid into the water (Sarvala et al. 2020). The dosage was decided solely on final pH and water clarity in precipitation experiments in the laboratory. In Littoistenjärvi, the dose was 44 mg L^{-1}, corresponding to 4.1 mg Al L^{-1}, and water became clear within hours. In Kirkkojärvi, the first dose on 19 May 2002 of 75 mg L^{-1} (6.8 mg Al L^{-1}) was too cautious, and the application had to be repeated to achieve clear water. On 4 Jun 2002, the dose was 83 mg L^{-1} (7.5 mg Al L^{-1}). In Kirkkojärvi, an additional 35 tonnes of granulated ferric sulphate (Ferrix 3, Kemira Oyj) was distributed in May 2005 to bind P in the surface sediment. After this treatment, water became brownish for a few weeks until finally clearing. In particular, the chemical treatment of Littoistenjärvi has served as a model for similar operations elsewhere in Finland.

Water quality monitoring

Monitoring followed a similar overall plan in both lakes. In Kirkkojärvi, the first water quality samples were taken in 1973, and annual monitoring started in 1988. A more intensive program was launched for 1996–2006, when samples with a Limnos sampler were taken at 2-week intervals through the summer (May–Oct) from the surface, midwater, and just above the bottom at a 2.5 m deep site. More restricted monitoring continued after 2006. Winter samples from under the ice were taken during several years.

In Littoistenjärvi, water quality monitoring started in 1963 and in 1992 developed into an intensive ecosystem monitoring program that is still running (Sarvala et al. 2020). Since 1992, samples have been collected with a Limnos sampler weekly or twice a month from April–May to September–November, on 10–15 dates annually, from 1 and 2 m depths at the deepest point of the lake. Samples from different depths were analyzed individually. In many years, vertical profiles were also taken 1–3 times in winter from under the ice.

Temperature and Secchi disk transparency were recorded in the field. The water quality parameters determined in the laboratory included pH, total and dissolved inorganic P (TP, DIP), total nitrogen, nitrite/nitrate-nitrogen, and ammonium = nitrogen (TN, NO_2/NO_3-N, NH_4-N, respectively; inorganic nutrients from Nuclepore-filtered samples), chlorophyll *a* (Chl-*a*), and dissolved oxygen (O_2, titrimetric laboratory method, during ice-covered period only). The permission for the chemical treatment of Littoistenjärvi required measuring additional variables such as total aluminum (Al, unfiltered) for 2 years. Earlier data of Al with longer intervals were available. Intensive follow-up of pH in different parts of the lake for 3 weeks after the treatment was also required. In Kirkkojärvi, Al concentrations were determined 7 times in June–October 2002.

In earlier years, samples were analyzed in the SW Finland Environment Centre laboratory and more recently at the Southwest Finland Water and Environment Research, Ltd. Analytical methods followed standard laboratory procedures accepted by the Finnish Accreditation Service (FINAS). No discontinuities were observed in the data that might have been related to the change of the analytical laboratory. Water quality results are available from the Finnish Environment Institute open data service.

Internal loading of P was estimated from seasonal changes in TP concentration in lake water as in Sarvala et al. (2020). In both lakes, the TP concentration of water increased manifold during the summer. Particularly, the early and midsummer in this area are dry, so the increase can be mostly ascribed to internal loading. Accordingly, the difference between the lowest TP concentration during March–May and the highest value between 1 July and 15 September was taken as the net internal loading of P.

Ecological monitoring: submerged plants, phytoplankton, zooplankton, and fish

In Littoistenjärvi, from 1986 the cover and biomass of submerged plants were monitored by Scuba divers along 10 littoral transects and 10 sites in the central open area deeper than 1.5 m (Sarvala et al. 2020), located according to a stratified random design. In Kirkkojärvi, a survey of submerged plants was performed after the treatment in 2004. Scuba divers recorded plant cover and abundance on eight 50 m long transects perpendicular to the shoreline.

Phytoplankton and crustacean zooplankton were sampled on the same dates as water quality in Kirkkojärvi in 1996–2006 and in Littoistenjärvi in 1983 and 1989 and every year from 1992 onward (methodological details in Sarvala et al. 2020). Composite samples of phytoplankton were counted with an inverted microscope, applying the extensive quantitative counting procedure of the Finnish Environment Institute, consistent with the standard (CEN 2006). Zooplankton samples were concentrated with a 25 or 50 µm mesh net and combined in the laboratory to form 1 composite sample for each date. Using an inverted microscope, zooplankton subsamples were counted until 50–200 individuals of each dominant crustacean species were measured. Their lengths were converted to carbon biomass using carbon-to-length regressions as in Sarvala et al. (1998).

From 1993 to 2020, the fish stocks in Littoistenjärvi were monitored with test fishing once a year using Nordic gillnets (CEN 2005: height 1.5 m, length 30 m, each containing 12 mesh sizes from 5 to 55 mm, knot to knot; 20 gillnet nights; Sarvala et al. 2020). Catch per unit effort (CPUE) was recorded as number and biomass of fish. In Kirkkojärvi, test fishing with the Nordic gillnets was performed in 1996–2007. Because of high abundance of fish in Kirkkojärvi, the fishing effort was only 2–6 gillnet nights each year.

Statistical analyses

To evaluate the success of the chemical treatment, changes in TP, TN, and Chl-*a* were examined over a period of 11 years before and 20 years after the treatment in Kirkkojärvi and over 25 years before and 6 years after the treatment in Littoistenjärvi. Changes in phytoplankton, crustacean zooplankton, and fish were likewise examined over 6 years before and 6 years after the treatment in Kirkkojärvi and over a period of 25–34 years before and 6 years after the treatment in Littoistenjärvi. Submerged macrophyte occurrence was checked once after the treatment in Kirkkojärvi and for a period of 22 years before and 6 years after the treatment in Littoistenjärvi. The values observed for TP, TN, and Chl-*a* were compared to the boundary values for the ecological quality classes defined for the WFD (Aroviita et al. 2019).

Statistical calculations were performed in Microsoft Excel 2016 and in the R environment 4.2.2, 2022. Changes of TP and Chl-*a* before and after the chemical treatment were examined with linear regression and covariance analyses (Sokal and Rohlf 1981). Normality of residuals was checked with Shapiro-Wilk's normality test and the homogeneity of variances with Bartlett's test. In Kirkkojärvi, the pre-treatment period extended from the beginning of regular monitoring in 1989 to 2001 and the post-treatment period from 2002 to 2020. In Littoistenjärvi, the pre-treatment period covered 1992 to 2016, and the post-treatment period covered 2017 to 2022 (to 2023 for the spring data).

Results

Internal loading and water quality parameters

Before the chemical treatment, the TP, TN, and Chl-*a* concentrations showed characteristic seasonal variation in both lakes, increasing manifold during summer to peak values, usually in August (TP illustrated in Fig. 1). Decline started in autumn, and under the ice-cover the concentrations returned to the lower winter level. Superimposed over this seasonality, the average June–September concentrations of TP and Chl-*a* were gradually increasing, indicating ensuing eutrophication (Fig. 2). In Kirkkojärvi, eutrophication was already occurring when regular monitoring started in 1989 (Fig. 2) while in Littoistenjärvi the deterioration of water quality started around 1992 (Fig. 3).

The chemical treatment resulted in an abrupt decline of the TP levels in both lakes, in Littoistenjärvi from (mean [SE]) 100 (3) µg L^{-1} in 2014–2016 to 28 (2) µg L^{-1} in 2017–2019 (72% decrease), and in Kirkkojärvi from 314 (35) µg L^{-1} in 1999–2001 to 48 (3) µg L^{-1} in 2002–2004 (85% decrease; Fig. 2 and 3). Consequently, the Chl-*a* concentrations in Littoistenjärvi declined from 65 (7) µg L^{-1} in 2014–2016 to 8 (1) µg L^{-1} in 2017–2019 (87% decrease), and in Kirkkojärvi from 148 (21) µg L^{-1} in 1999–2001 to 18 (2) µg L^{-1} in 2002–2004 (88% decrease). Simultaneously TN declined by 67% in Littoistenjärvi and 72% in Kirkkojärvi. Overall, the concentrations of TN closely followed those of TP in both lakes (regressions of Jun–Sep TN on TP, Kirkkojärvi: adjusted $R^2 = 0.79$, $n = 26$, $p < 0.001$; Littoistenjärvi: adjusted $R^2 = 0.80$, $n = 39$, $p < 0.001$).

These changes were associated with corresponding declines of the net internal P loading. In Littoistenjärvi, internal loading declined by 77%, from 184 mg m^{-2} a^{-1} in 2006–2016 to 60 mg m^{-2} a^{-1} in 2017–2022 (Fig. 4). In Kirkkojärvi, the net internal TP loading decreased from 1101 mg m^{-2} a^{-1} in 1996–2001 to 218 mg m^{-2} a^{-1} in 2002–2009, a drop of 80%. In both lakes, the average Chl-*a* level in June–September was predictable from the net internal loading of P (Fig. 5).

In Littoistenjärvi, spring (Mar–May) TP levels had begun to decrease several years before the chemical treatment. The 40% decline of TP from 2011 to 2017 (spring before treatment) was highly significant (Fig. 6: adjusted $R^2 = 0.81$, $n = 7$, $p < 0.01$), a change that coincided with the intensification of aeration during the ice-covered period and reduction of external loading. The late summer TP and Chl-*a* levels were predictable from the spring TP values (Fig. 7). A similar relationship was not found in

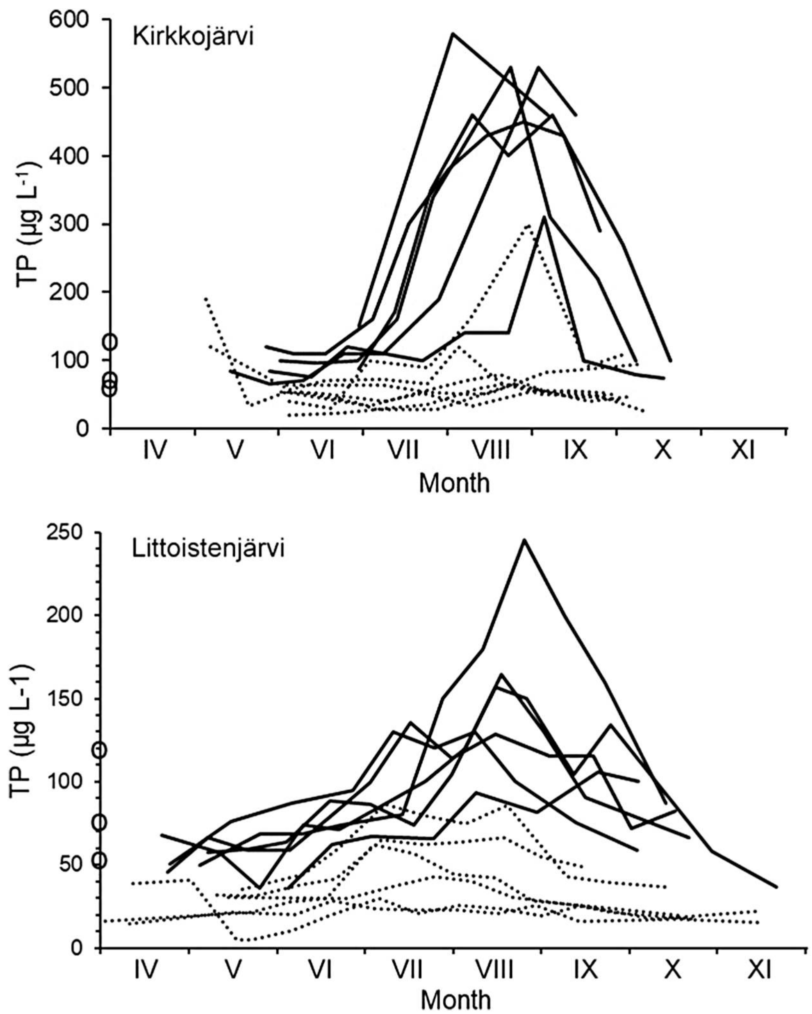

Figure 1. Seasonal development of TP level in 1996–2009 in Kirkkojärvi (top; solid lines before treatment in 1996–2001, dotted lines after treatment in 2002–2009) and in Littoistenjärvi (bottom; solid lines before treatment in 2006–2016 every second year, and dotted lines after treatment in 2017–2022). Circles along the y-axis indicate approximately the upper boundary values of TP of the good, moderate, and poor quality classes for nutrient-rich lakes according to the WFD ecological state classification (TP = 55, 75, and 120 µg L^{-1}, respectively).

Kirkkojärvi, possibly masked by the high between-year variability.

The external and internal P loadings before the chemical treatment were much higher in Kirkkojärvi than in Littoistenjärvi. The internal P load calculated from the P budget for Kirkkojärvi was almost 9 times higher than the estimated external load (Table 1). The net internal P loading calculated here from the increase of TP in lake water during summer was much lower, but still higher than the external loading.

After treatment, along with the increasing internal loading, TP, TN, and Chl-*a* started to increase again. During the first 4 years the levels remained low. In Kirkkojärvi (Fig. 2), the regression slopes of TP and Chl-*a* against year were lower for the post-treatment period (pre- and post-treatment slopes for TP were 15.8 and 3.3 µg L^{-1} a^{-1}; those for Chl-*a* 10.1 and 2.5 µg L^{-1} a^{-1}, respectively), indicating a slower rate of re-eutrophication. The application of regression analyses on the Kirkkojärvi data, however, was questionable because of nonnormal distribution of the annual TP and Chl-*a* values. Clearly elevated values of TP appeared in 2011 and 2020 and of Chl-*a* in 2006, 2011, and 2020, whereas levels in other years remained almost unchanged (Fig. 2). Ignoring the outliers, however, showed little change in TP or Chl-*a* in Kirkkojärvi since the treatment. Although TP and Chl-*a* concentrations after the treatment remained relatively high, the average TP and Chl-*a* of 89.4 and 50.9 µg L^{-1}, respectively, for the whole post-treatment period 2002–2020 (including the outliers) indicated substantial improvement over the initial state (averages for the pre-treatment period 223 and 97.2 µg L^{-1}, respectively).

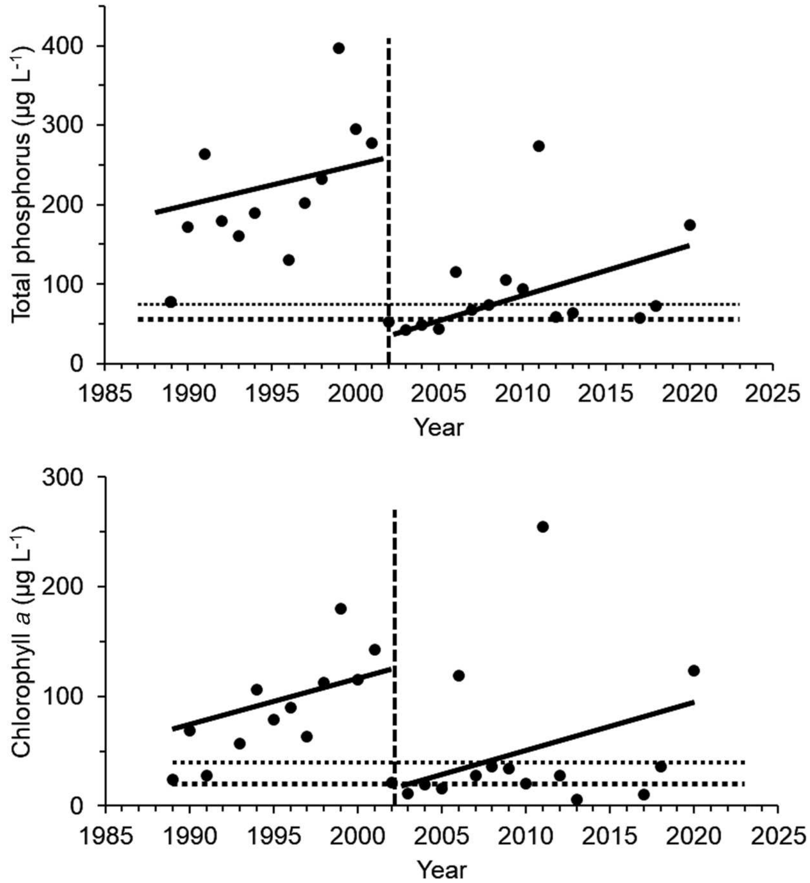

Figure 2. Development of total phosphorus (top) and chlorophyll *a* concentrations (bottom) before and after the chemical treatment in 1989–2020 in Kirkkojärvi. Vertical broken lines indicate the year of the chemical treatment. Horizontal dotted lines show the upper limits of the "good" and "moderate" quality classes for nutrient-rich lakes according to the WFD. Solid oblique lines represent the common slopes for the pre- and post-treatment periods from the covariance analyses, and their crossings with the horizontal lines predict when the quality class would change.

In Littoistenjärvi (Fig. 3), the pre- and post-treatment slopes for TP were 4.0 and 8.1 and those for Chl-*a* were 2.5 and 5.5, respectively, implying faster re-eutrophication after the treatment than in Kirkkojärvi. In Littoistenjärvi, short-term increases of TP and Chl-*a* up to 43–62 µg L^{-1} and 23–53 µg L^{-1}, respectively, appeared in 2019 and 2020 in late June and early July, but declines followed in July. In 2021, high levels of up to 86 and 78 µg L^{-1}, respectively, prevailed from midsummer to September. In 2022, TP and Chl-*a* again remained at a lower level (peak values 66 and 28 µg L^{-1}, respectively). Despite these temporary increases, only in 2021 did the summer average of TP and Chl-*a* exceed the upper limit for "good" ecological state of WFD. Moreover, preliminary data for 2023 indicated substantial improvement of water quality in Litttoistenjärvi.

Most importantly from the water quality management point of view, a drastic decline of the summer cyanobacteria biomass was observed in both lakes. In Littoistenjärvi, cyanobacteria declined by 92% of the average before the treatment in 2001–2016 (Fig. 8) and in Kirkkojärvi by 88% from 1996–2001 to 2002–2006.

Longevity of the chemical treatment

In principle, extrapolation of the linear regressions of the TP and Chl-*a* calculated for the post-treatment periods would make it possible to predict how long the water quality variables would remain below accepted target values (Fig. 2 and 3). In Littoistenjärvi, however, the post-treatment monitoring period was short and the variance of the observed water quality values high, making predictions based on the post-treatment regressions uncertain (Table 2). However, the rates of change of TP and Chl-*a* after treatment did not differ significantly from those before treatment, allowing predictions using the common slope for the pre- and post-treatment periods obtained from the analysis of covariance (Table 2). In Kirkkojärvi, although the difference between the pre- and post-treatment slopes for Chl-*a* was not significant, TP slopes were significantly

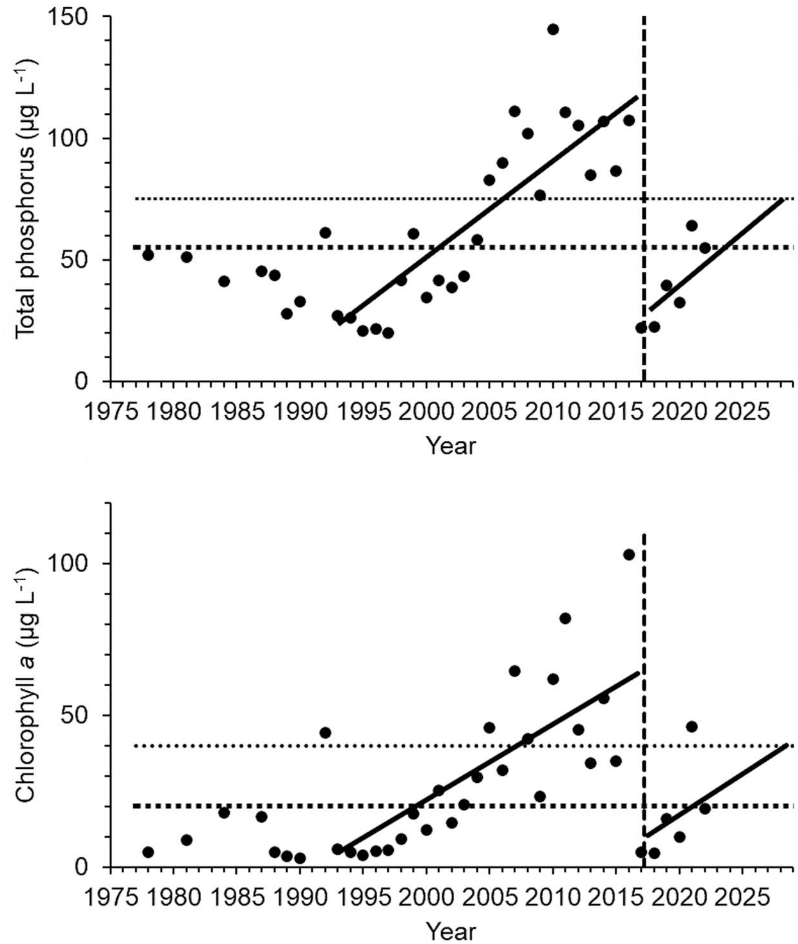

Figure 3. Development of total phosphorus (top) and chlorophyll *a* concentrations (bottom) before and after the chemical treatment in 1978–2022 in Littoistenjärvi. Vertical broken lines indicate the year of the chemical treatment. Horizontal dotted lines show the upper limits of the "good" and "moderate" quality classes for nutrient-rich lakes according to the WFD. Solid oblique lines represent the common slopes for the pre- and post-treatment periods from the covariance analyses, and their crossings with the horizontal lines predict when the quality class would change.

different, making the use of the common slope unreliable. Moreover, in Kirkkojärvi, the distributions of both variables showed irregular development. Because of this nonnormality, confirmed by Shapiro-Wilks tests, the regression approach was only tentatively applied to Kirkkojärvi (Fig. 2). A more realistic view of the post-treatment behaviour of Kirkkojärvi was obtained by comparing the actual monitoring data to the target values.

The lake type-specific boundary limits between the quality classes of the WFD were used as natural reference values. In the nutrient-rich lake type, the upper boundaries for good and moderate water quality are, respectively, 55 and 75 µg L^{-1} for TP and 20 and 40 µg L^{-1} for Chl-*a* (Aroviita et al. 2019). In Kirkkojärvi, the observed Chl-*a* values remained below the moderate boundary in 12 of the 15 monitoring years after the treatment, and even below the good boundary in 5 years. Likewise, the observed TP values were below the moderate boundary in 10 of 15 years and below the good boundary in 5 years. Thus, even 10–18 years after the chemical treatment, Kirkkojärvi mostly maintained moderate water quality, although unpredictable algal blooms have developed 3 times in 20 years.

In Littoistenjärvi, applying the common slope for the pre- and post-treatment periods from the covariance analysis, the good and moderate boundary values for TP would be exceeded in 2024 and 2029 and those for Chl-*a* in 2021 and 2029 (Fig. 3). The actual monitoring results suggest better success, however. In 5 of 6 post-treatment monitoring years, the observed TP and Chl-*a* were both below the good boundary, and in the seventh summer (2023), for which the records were not yet complete, Littoistenjärvi had good water quality, with TP and Chl-*a* varying between 24–33 and 3.5–6.5 µg L^{-1}, respectively. The eutrophication rate in Littoistenjärvi immediately after the treatment was higher than in Kirkkojärvi, but because of the worse

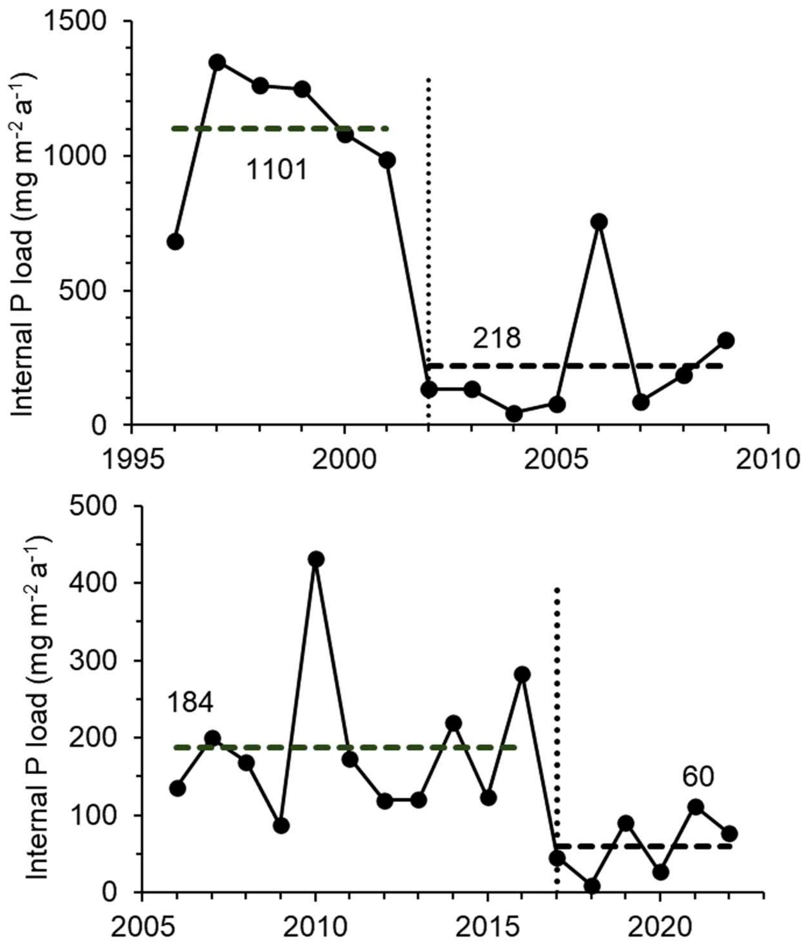

Figure 4. Net internal P loading in Kirkkojärvi (top; 1996–2009) and Littoistenjärvi (bottom; 2006–2022). Average loadings before and after the chemical treatment indicated by broken lines. Vertical dotted lines show year of chemical treatment.

starting condition of Kirkkojärvi, the water quality remained better in Littoistenjärvi. If moderate water quality is the target, the longevity of the chemical treatment would be 13 years in Littoistenjärvi.

Submerged macrophytes

The submerged vegetation of Kirkkojärvi was not regularly monitored, but before the chemical treatment was scarce. The median Secchi disk transparency before treatment was 0.5 m, indicating sufficient light for plants only in areas shallower than 1 m, which comprised 12% of the surface area of Kirkkojärvi. Immediately after treatment, the transparency was 1.5 ± 0.5 m, but decreased to 1 ± 0.5 m 5–10 years after the treatment, and the biomass and biodiversity of the submerged vegetation were increasing. Submerged macrophytes were most abundant at 0.8–1.3 m depth but occurred to 2 m depth, and the most abundant species was *Potamogeton obtusifolius*. The areas shallower than 2 m comprised 26% of the total area of the lake, leaving most of the area below the photic layer. The bathymetry of the lake thus prevents submerged plants from becoming dominant components in the Kirkkojärvi ecosystem.

In Littoistenjärvi, the submerged macrophytes were also scarce before the treatment but were expected to increase after the chemical treatment when water became clear. But *E. canadensis* and another invasive alien species, *Ceratophyllum demersum*, increased little in the first 6 years after treatment, and the native *Myriophyllum alterniflorum* had the highest biomass. A third invasive species, *Potamogeton crispus*, appeared but remained scarce. In the seventh summer (2023), however, *E. canadensis* was again growing faster, as indicated by a pH exceeding 9.1 in early August when the level expected from the phytoplankton chlorophyll was ~7.5, a situation resembling that in the 1990s when *E. canadensis* was dominant.

Phytoplankton

Phytoplankton was reduced substantially during the chemical treatment (Fig. 8), and 2 weeks after treatment the total phytoplankton biomass was still low (0.05 mg L^{-1} in Kirkkojärvi, and 0.09 mg L^{-1} in Littoistenjärvi). The

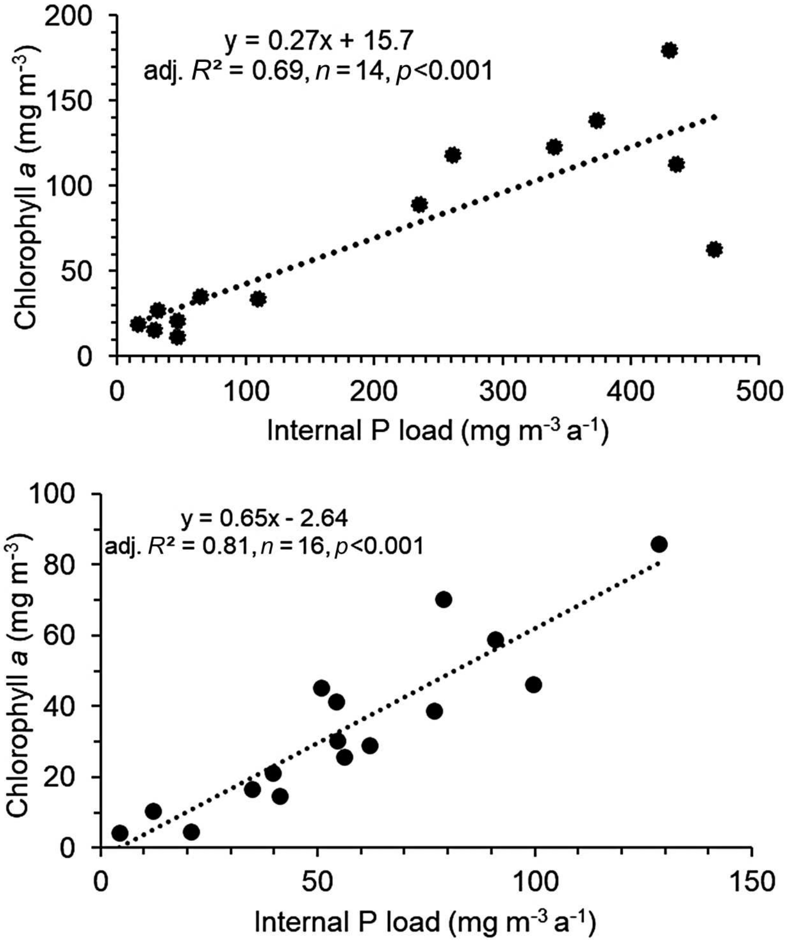

Figure 5. Average June–September chlorophyll *a* relative to the internal P load in Kirkkojärvi (top, 1996–2009) and Littoistenjärvi (bottom, 2006–2022).

recovery was fast, however, and normal levels of phytoplankton biomass were attained 4 weeks after the treatment. During the first 5 years after the treatment, consistent with the reduced TP and Chl-*a* levels, the average summer biomass of phytoplankton in Littoistenjärvi remained low (1.4–2.6 mg L^{-1}), and the proportion of cyanobacteria was variable (1–37%; for this lake type, WFD quality classes of phytoplankton have not been defined). In 2019–2021 in Littoistenjärvi, the cyanobacteria developed minor peaks during hot weather spells before midsummer but disappeared in a few weeks. Only during the exceptionally hot

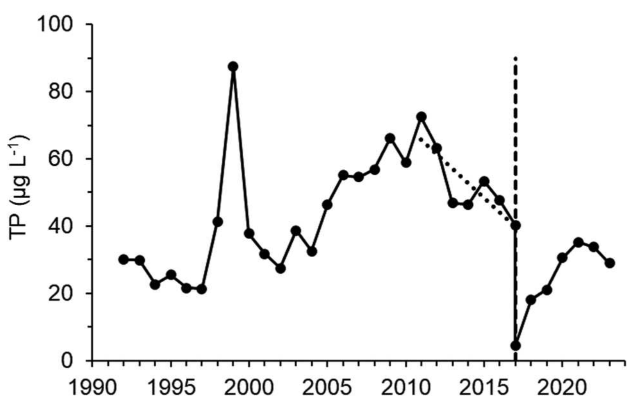

Figure 6. Spring (Mar–May) concentrations of total phosphorus (TP) in Littoistenjärvi in 1992–2023. The high peak in 1999 was caused by anoxia under the ice cover. The oblique line shows the period of declining TP during the intensified winter aeration in 2012–2017.

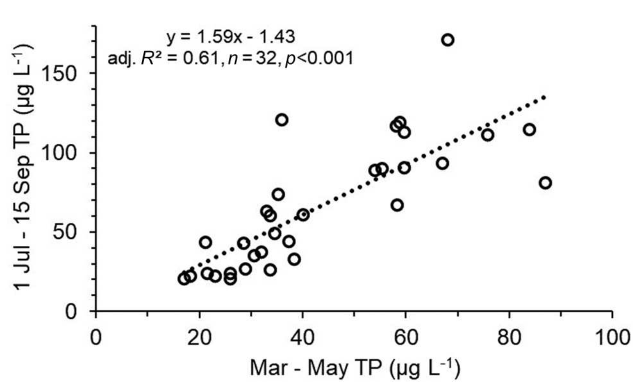

Figure 7. July to mid-September TP concentrations in Littoistenjärvi in 1992–2023 relative to the spring TP level (Mar–May).

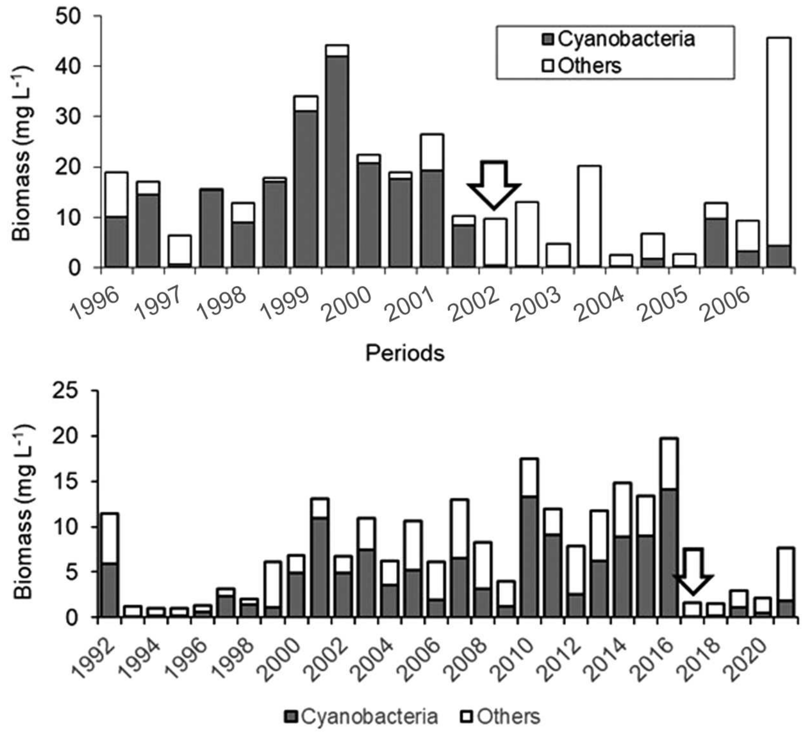

Figure 8. Biomass of cyanobacteria and other phytoplankton in Kirkkojärvi (top) and Littoistenjärvi (bottom). Values are averages for 1 June to 30 September, except that early and late summer averages were kept separate for Kirkkojärvi. Block arrows indicate the date of aluminum treatment.

summer of 2021, phytoplankton biomass and Chl-*a* remained high until September, but cyanobacteria were not dominant. In both lakes, the highest post-treatment phytoplankton biomass consisted mainly of Dinophyceae. Overall, the post-treatment phytoplankton composition seemed unstable, with dominant species varying from year to year.

Zooplankton

Crustacean zooplankton (Fig. 9) also disappeared during the chemical treatment but recovered in ~2 months. In Kirkkojärvi, zooplankton biomass and composition were variable even before the treatment and were little affected. Zooplankton biomass in Littoistenjärvi was positively, although weakly, related to the phytoplankton biomass (adjusted $R^2 = 0.15$, $n = 27$, $p < 0.05$) and increased with the eutrophication through the 1990s to peak values in 2006–2010, followed by a steep decline during the treatment.

Planktonic crustaceans are short-lived organisms, and their biomass tracks the variations in the abundance of phytoplankton food. Therefore, zooplankton biomass remained low even after the recovery from the chemical treatment. By contrast, planktivorous and benthivorous fishes live for several years, and when the chemical treatment was adjusted to save the fish, the predation pressure exerted by fish on zooplankton steeply increased after the treatment. In Littoistenjärvi this higher predation was seen as a drastic increase in the biomass ratio between fish and big zooplankton herbivores (Fig. 10). The potential for grazing control of phytoplankton by the zooplankton was thus weakened by the successful chemical treatment. In Kirkkojärvi similar imbalance did not develop because almost all fishes were killed in the chemical treatment.

Table 2. Analysis of covariance of the pre- and post-treatment total phosphorus (TP) and chlorophyll *a* (Chl-*a*) values (Jun–Sep averages) in the lakes Kirkkojärvi and Littoistenjärvi, with year as covariate, and pre- and post-treatment periods as another explanatory factor. The slopes in the regression equation y = a + b(year) did not differ significantly between periods, except for TP in Kirkkojärvi.

	b	Adjusted R^2	df	F	p	Effect of period
Kirkkojärvi TP	6.19	0.68	3, 22	19.01	<0.001	$p = 0.03$
Kirkkojärvi Chl-*a*	4.22	0.22	3, 22	3.36	<0.05	ns
Littoistenjärvi TP	4.07	0.56	3, 34	14.43	<0.001	ns
Littoistenjärvi Chl-*a*	2.58	0.49	3, 34	12.67	<0.001	ns

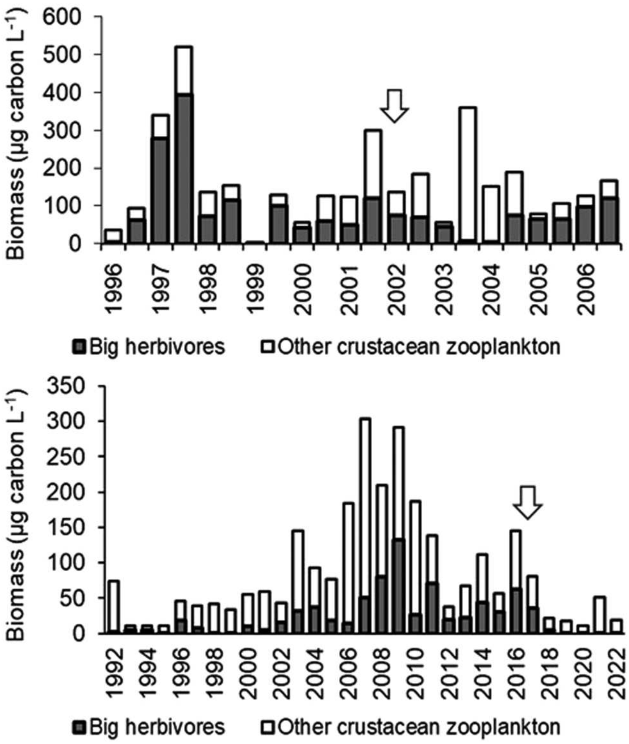

Figure 9. June–September averages of the biomass of big herbivorous and other crustacean zooplankton in lakes Kirkkojärvi (top) and Littoistenjärvi (bottom). Big herbivorous zooplankton comprised *Daphnia* species, *Holopedium gibberum*, *Limnosida frontosa*, and *Sida crystallina* as well as copepodid and adult *Eudiaptomus* spp. calanoids. Block arrows indicate the year of aluminum treatment.

Fish

Experimental fishing with the standard Nordic gillnets before the treatment revealed high abundances of planktivorous and benthivorous fish in both lakes: average CPUE was 7 kg net^{-1} per night for Kirkkojärvi and 3 kg net^{-1} per night for Littoistenjärvi. In both lakes, biomanipulation by removal fishing had been attempted before the chemical treatment, but with poor success. Therefore, to rid Kirkkojärvi of the unwanted fish, the chemical dose was intentionally set so high that fish would die. The lowest pH measured during the treatment was pH 4.0, and as anticipated, a complete fish kill followed; 15 tonnes of dead fish were collected from the shores, half of which were >4 kg bream (*Abramis brama*), corresponding to 357 kg ha^{-1}. Surprisingly, the fish assemblage recovered by the following summer (at least partly because of migration of adult fishes from the Baltic Sea). Later the fish biomass remained at ~50% of the level preceding the treatment (Fig. 11). The dominant species were perch (*Perca fluviatilis*), roach (*Rutilus rutilus*), and stocked pike (*Esox lucius*).

In Littoistenjärvi, the aim was to save the fish, and the chemical dose was adjusted so that the pH would not decrease below pH 6.0. Because of windy weather, however, mixing was uneven, and pH values as low as 5.1–5.8 were recorded 3 days after the treatment. Return to pH >6.0 took 1 week, during which ~5 tonnes of big bream died and were removed. Otherwise, the fish were little affected (Fig. 11).

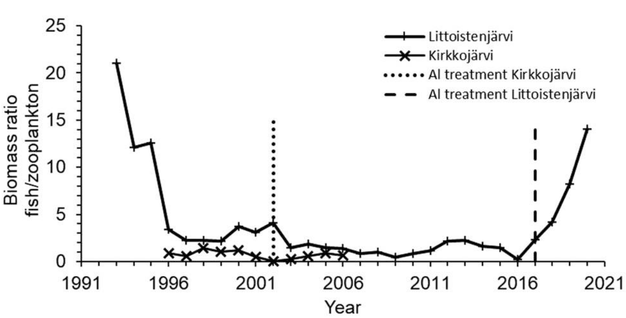

Figure 10. Fish/zooplankton biomass ratio during 1993–2020 in Littoistenjärvi and 1996–2006 in Kirkkojärvi. Fish biomass given as kg gillnet^{-1} night^{-1}, zooplankton biomass converted from µg carbon L^{-1} to g wet mass m^{-3}, assuming the carbon content of zooplankton is 6% of wet mass. Block arrows indicate the date of aluminum treatment.

Factors affecting internal loading

In Littoistenjärvi, the seasonal increases of TP were predictable from water temperature and pH (for temperature, $R^2 = 0.18$, $n = 58$, $p < 0.001$; for pH, $R^2 = 0.42$, $n = 58$, $p < 0.001$), although from field data their effects were difficult to separate because they were significantly correlated ($R^2 = 0.17$, $n = 39$, $p < 0.01$). In a multiple regression both factors were significant, but pH was more important (adjusted $R^2 = 0.46$, $n = 58$, $p < 0.001$; for temperature $t = 2.44$, $p < 0.05$; for pH $t = 5.60$, $p < 0.001$). In a multiple regression of the Kirkkojärvi data, the regression of TP on pH was likewise highly significant while the effect of temperature was not. In a single-factor regression, pH had marginal correlation with temperature ($p < 0.05$, $n = 95$).

A multiple regression of the long-term summer means (1 May–15 Sep) from Littoistenjärvi confirmed that both submerged macrophytes and cyanobacteria were significant factors affecting pH (adjusted $R^2 = 0.51$, $n = 29$, $p < 0.001$; for submerged macrophytes $t = 4.56$, $p = 0.001$; for cyanobacteria $t = 4.18$, $p < 0.001$). In both lakes, increasing Chl-*a* led to higher pH, and because in most years the pH increase in Littoistenjärvi seemed to precede the TP increase, the increase of TP in early summer after the chemical treatment could be a consequence of increasing phytoplankton. This process could then initiate a vicious circle, where more TP means more cyanobacteria, resulting in higher pH and increasing release of P.

In this chain of events, zooplankton grazers play an important role, as became evident in the first summer following the chemical treatment of Littoistenjärvi. Phytoplankton recovered in 4 weeks while the recovery of zooplankton took 4 weeks more. In the absence of efficient grazers during these 4 weeks, phytoplankton abundance rapidly increased. But when crustacean zooplankton had recovered by early July, phytoplankton decreased, and for the rest of the summer the zooplankton and phytoplankton biomasses were inversely related, although a positive relationship would be expected because zooplankton abundance is dependent on the abundance of phytoplankton food.

Although sampling intervals were long relative to the time scales of plankton life, monitoring data for 2019–2021 suggest Chl-*a* peaked earlier than TP. Zooplankton data from both Kirkkojärvi and Littoistenjärvi confirmed that efficient zooplankton grazers were lacking, allowing uncontrolled growth of phytoplankton. In most years in Littoistenjärvi, the summer zooplankton/phytoplankton biomass ratio was low (Fig. 12), indicating that zooplankton was not able to control phytoplankton. By contrast, during the treatment summer this ratio was moderately high (Fig. 12). The long-term development of the Chl-*a*/TP ratio in Littoistenjärvi (data not shown) also illustrated the changes in grazing control, with increasing values up to 2016 indicating weakening grazing control during eutrophication, lower values and restored control in 2017 and 2018 after treatment, and weaker control again since 2019.

Discussion

The precipitation of P with polyaluminum chloride successfully curbed the internal loading in 2 shallow eutrophic lakes. In both lakes, the chemical restoration resulted in a 70–90% decline of TP and Chl-*a*, and the rampant blooms of cyanobacteria disappeared. In both study lakes, the doses were within the range found earlier to be most successful in shallow lakes (Huser et al. 2016b, Araújo et al. 2018). As became evident during the first treatment of Kirkkojärvi, the dosage needs to be defined according to the goals of the treatment. The higher dose required in Kirkkojärvi was understandable because the external loading and in-lake TP concentrations were clearly higher than in Littoistenjärvi. Careful estimation of the Al dose is needed because too low doses weaken the efficiency of the treatment, and too high doses lead to low pH and the appearance of toxic Al ions (Zamparas and Zacharias 2014, Agstam-Norlin et al. 2021). In Kirkkojärvi and Littoistenjärvi, dosing was based on the pH and alkalinity of water, but more accurate dosing based on the amount of mobile P in the surface layers of sediment is currently recommended (Reitzel et al. 2005, Kuster et al. 2020). The Al dose applied in Littoistenjärvi was actually close to that estimated from sediment P fractions (Sarvala et al. 2020).

The potential toxicity of Al is often an issue when planning chemical restoration. Al is the most common metal in the Earth's crust and occurs in natural waters as many different inorganic and organic compounds. Al is a nonessential element for organisms, and its

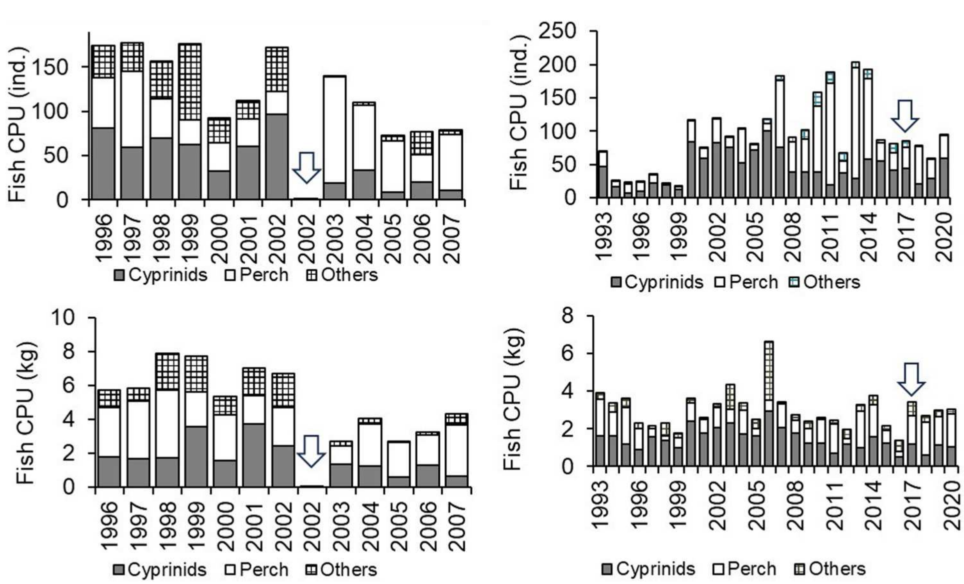

Figure 11. Catch per unit effort (CPU, [gillnet night]$^{-1}$) of cyprinids, perch, and other fish species as numbers (top) and biomass (bottom) in Nordic nets in 1996–2007 in Kirkkojärvi (left) and 1993–2020 in Littoistenjärvi (right). Block arrows indicate the date of aluminum treatment.

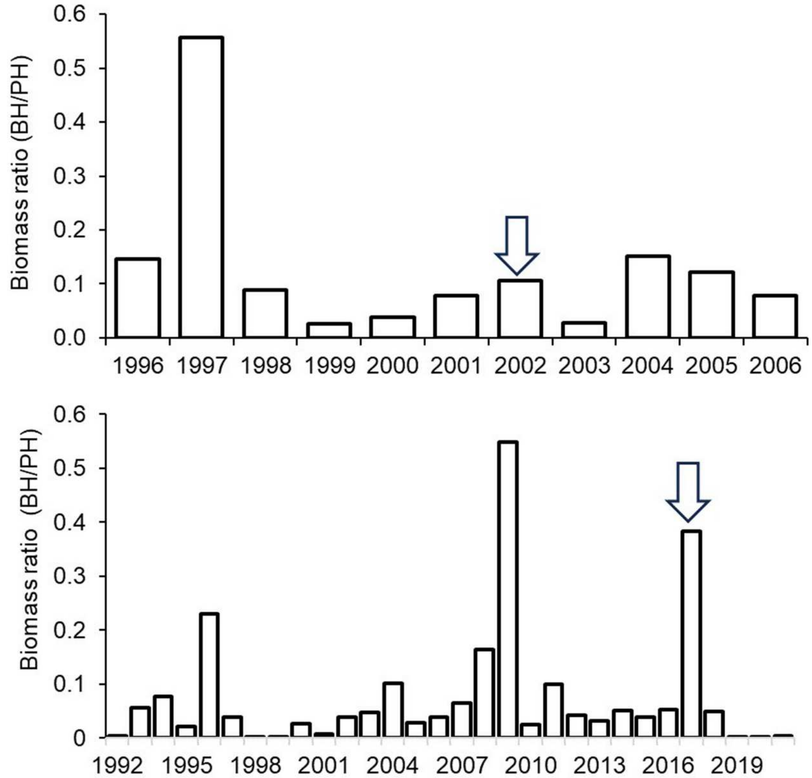

Figure 12. Average summer season biomass ratios between big zooplankton herbivores and total phytoplankton (BH/PH) during 1996–2006 in Kirkkojärvi (top) and 2000–2021 in Littoistenjärvi (bottom). Carbon biomass of zooplankton converted to wet mass as in Fig. 10. Block arrows indicate the date of aluminum treatment.

ionic forms are particularly toxic. The bioavailability and toxicity of the different Al forms depend particularly on pH. At neutral pH, Al is not toxic, but at pH <6.0, a larger part occurs as the highly toxic Al^{3+} ion. Al may accumulate on the respiratory surfaces, clogging fish gills, causing death or impaired growth, and it may also disturb the fertilization and hatching of fish eggs. In addition to pH, Al toxicity is affected by several other water quality factors such as water hardness and concentration of dissolved organic carbon, both reducing the toxicity. Because of the complexity of Al chemistry, no single safe upper limit can be defined for Al concentration. Models developed by the US Environmental Protection Agency (US EPA 2018), however, indicate that at pH 6, when $CaCO_3$ hardness is 40 mg L^{-1} and dissolved organic carbon concentration is 10 mg L^{-1}, the safe upper limit for chronic exposure would be 440 µg L^{-1}. In Littoistenjärvi, the total Al concentrations in water increased for a few days 5–6-fold to 630 µg L^{-1}. In <2 weeks, however, the concentrations fell below the levels observed in 1984–1990 before the chemical treatment (100 µg L^{-1}; Sarvala et al. 2020). Averages for 2018 and 2019 were even lower: 64 and 38 µg L^{-1}. In Kirkkojärvi, the highest Al concentration remained lower than in Littoistenjärvi, yet fish were killed in Kirkkojärvi while most survived in Littoistenjärvi because of the higher toxicity of Al at the low pH of Kirkkojärvi.

Although not directly affected by the chemical treatment, N declined almost as much as P, suggesting the N level was influenced by the N fixation of cyanobacteria, in turn regulated by P concentration.

The restoration success in Littoistenjärvi in the first years after treatment was probably affected by weather conditions both positively and negatively. High summer temperatures result in stronger year-classes of the spring-spawning perch and roach (Böhling et al. 1991). The treatment summer 2017 was cool, and combined with the Al treatment in spring probably led to weak year-classes of perch and roach, and consequently to relatively low predation pressure on zooplankton from young-of-the-year fish. As a result, zooplankton herbivores could control the abundance of phytoplankton in the first post-treatment summer. By contrast, the warm summers in 2018, 2019, and 2021 were favorable for the reproduction of perch and roach, resulting in high predation pressure on big zooplankton from 2019 onward. Accordingly, in the following summers, big zooplankton herbivores remained scarce (Fig. 9) and had little effect on phytoplankton.

Because of the hypertrophic starting conditions, even if the restoration was considered successful, both lakes fulfilled the requirements of a good ecological status for only the first 4 years, according to the WFD. Similar short 4-year successes have been reported elsewhere (Agstam-Norlin et al. 2021). A 50% reduction of TP or Chl-*a* values from the initial level is often regarded as the success criterion for restoration (Welch and Cooke 1999, Huser et al. 2016a). Agstam-Norlin et al. (2021) used a similar regression approach to determine the longevity of Al applications, but instead of the 50% improvement of the initial conditions, their reference value was obtained by subtracting 1 standard error from the 4-year pre-treatment average. Reference values calculated in this way would extend the longevity of chemical treatment in Littoistenjärvi to 2027 for TP and to 2029 for Chl-*a* (reference values TP = 80.8 μg L^{-1}, Chl-*a* = 33.4 μg L^{-1}).

The reference values of Huser et al. (2016a) and Agstam-Norlin et al. (2021) both measure restoration success relative to the initial conditions and, if unacceptable, the reference value may remain rather high. An absolute measure of success would be more desirable, and the boundary values for the WFD quality classes are a natural alternative (Fig. 2 and 3). These values vary depending on lake type. Here both lakes are treated as nutrient-rich. Predictions of treatment longevity with the WFD boundary values suggest that Littoistenjärvi would remain in good state up to 2024 for TP or 2021 for Chl-*a* (i.e., 5–8 years after treatment). In Kirkkojärvi the occasional years with poor water quality confounded the use of regressions, but the observed post-treatment values remained within the moderate or even good quality range for most years, indicating high longevity. The major aim of lake management is to achieve a good ecological state, but another target is to stop the deterioration of water quality, which was apparently attained in Kirkkojärvi; Littoistenjärvi was closer to achieving a good water quality state in the long-term. Overall, in shallow lakes with heavy internal loading of P for many years, even decades, expecting that a single chemical treatment would result in a long-lasting good ecological state may not be realistic. Instead, a moderate ecological state seems an attainable goal, although even its maintenance may require repeated treatments.

The chemical treatment resulted in drastic decline of TP, Chl-*a*, phytoplankton biomass, and zooplankton. In Littoistenjärvi, however, starting the third year after treatment, TP, Chl-*a*, and phytoplankton biomass developed an early summer peak lasting for weeks. In June–July 2019 and 2020, the elevated nutrient and Chl-*a* levels coincided with high water temperatures (maximum 21.3 and 23.7 °C, respectively). Similarly, summer 2021, when high TP and Chl-*a* values prevailed until September, was particularly hot; June–July mean temperature was 22.6 °C, or 3.7 °C higher than the average for 1993–2022. The ongoing warming of climate may thus challenge the success of chemical restoration of lakes.

The small effect of the chemical treatment on the zooplankton in both lakes seems to be explained by the abundance of planktivorous fish both before and after treatment, keeping zooplankton biomass low and individuals small. Therefore, crustacean zooplankton usually lacked the capacity to control phytoplankton abundance, which was almost solely dependent on nutrient availability. Trophic interactions have an important role in determining water quality (Sarvala et al. 2000, Urrutia-Cordero et al. 2015, Ger et al. 2016), and unless the fish are eliminated using a high dose of aluminum chloride, the chemical precipitation of P should be accompanied by fish removal by some other means, as suggested by Araújo et al. (2016) and Huser et al. (2021). Our experiences show that a combination of treatments may be vital for successful restoration.

Although the post-treatment period in Littoistenjärvi was too short to reveal the true rates of change, the post-treatment re-eutrophication seems to have been slower in Kirkkojärvi than in Littoistenjärvi. This difference may be real because the timing of the reduction of external load differed between the lakes. In Kirkkojärvi, load reductions happened mostly after the chemical restoration through changes in agricultural practices. In addition, the small lake Riittiönjärvi draining into Kirkkojärvi was dredged and treated with aluminum chloride (JS 2007, unpubl. report). The iron addition in 2005 seems to have had little effect on water quality. In Littoistenjärvi, the major reduction of external load was accomplished before the chemical treatment, when runoff from roughly 25% of the drainage area was diverted to the River Aurajoki (Sarvala et al. 2020).

Our monitoring data give some insights into possible causes of the increasing internal loading of P, and thus TP, levels in water. Factors involved included abiotic variables such as temperature and pH and biotic variables such as phytoplankton blooms and submerged macrophytes, exerting mostly indirect influence. Higher temperatures enhanced the internal loading directly by accelerating all processes in the sediment, but also in many indirect ways. A major factor regulating P fluxes in our study lakes seems to be pH of water, which is largely determined by the metabolism of submerged plants and phytoplankton. Plant assimilation increases pH, which during the mass occurrences of *E. canadensis* in the 1990s could rise to pH 11 (Sarvala et al. 2020). When pH exceeds ~8.5, P starts to be released from the sediment or suspended solids in water (Reitzel et al. 2013, Zamparas and Zacharias 2014).

Conclusions

Consistent with the results of Agstam-Norlin et al. (2021), our experiences from Kirkkojärvi and Littoistenjärvi suggest that chemical precipitation of P with aluminum chloride is suitable for the restoration of waterbodies that suffer from heavy internal P loading. An important prerequisite for long-term success is that the external loading is also reduced. The appropriate dosage depends on the goals of the treatment. The Al dose can be adjusted to retain pH >6.0 to save the fish, although this weakens the role of top-down control of water quality by herbivorous zooplankton. With a higher dosage, lowering the pH to ~5, the fish can be killed and zooplankton grazers can keep phytoplankton in check. Whichever option is chosen, long-term monitoring provides the basis to estimate the proper dosage and offers the possibility to extrapolate the longevity of the treatment. Post-treatment monitoring is essential to determine whether the original goals were fulfilled and whether further treatments are required.

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Disclosure statement

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Data availability statement

Water chemistry, phytoplankton and fish data are available through Finnish Environment Institute's open data service. Other data are available from the authors on request.

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