Water Research 200 (2021) 117267

Contents lists available at ScienceDirect

Water Research

journal homepage: www.elsevier.com/locate/watres

# A 25-year retrospective analysis of factors influencing success of aluminum treatment for lake restoration

O. Agstam-Norlin[a,*], E.E. Lannergård[a], E. Rydin[b], M.N. Futter[a], B.J. Huser[a]

[a] Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences, P.O Box 7050, 750 07 Uppsala, Sweden
[b] Stockholm University, Baltic Sea Centre, 10691 Stockholm, Sweden

ARTICLE INFO

*Article history:*
Received 22 January 2021
Revised 12 May 2021
Accepted 15 May 2021

*Keywords:*
Lake restoration
Aluminum treatment
Phosphorus
Internal loading
Sediment
Legacy phosphorus

ABSTRACT

For more than 50 years, aluminum (Al)-salts have been used with varying degrees of success to inactivate excess mobile phosphorus (P) in lake sediments and restore lake water quality. Here, we analyzed the factors influencing effectiveness and longevity of Al-treatments performed in six Swedish lakes over the past 25 years. Trends in post-treatment measurements of total phosphorus (TP), Chlorophyll a (Chl_a), Secchi disk depth (SD) and internal P loading rates (Li) were analyzed and compared to pre-treatment conditions. All measured water quality parameters improved significantly during at least the first 4 years post-treatment and determination of direct effects of Al-treatment on sediment P release (Li) was possible for three lakes. Improvements in TP (-29 to -80%), Chl_a (-50 to -78%), SD (7 to 121%) and Li (-68 to -94%) were observed. Treatment longevity, determined via decreases in surface water TP after treatment, varied from 7 to >47 years. Lake type, Al dose, and relative watershed area were related to longevity. In addition, greater binding efficiency between Al and P was positively related to treatment longevity, which has not previously been shown. Our findings also demonstrate that adequate, long-term monitoring programs, including proper determination of external loads, are crucial to document the effect of Al-treatment on sediment P release and lake water quality.



## 1. Introduction

Excess internal loading of phosphorus (P) in lakes, a result of historical sediment P accumulation from external sources, is often the most important driver of in-lake P cycling and thus eutrophication (e.g. Pilgrim et al., 2007, Sondergaard et al., 2003). External P sources, e.g., leaching from agricultural soils and municipal/industrial wastewater are the primary source of excess nutrients in lakes (Conley et al., 2009). Even when external nutrient sources are controlled, recovery can be delayed by decades or longer due to the release of legacy (or mobile) P from lake sediments (Sas 1990). For more than 50 years, aluminum (Al)-salts have been used to permanently inactivate excess mobile P in lake sediments and restore lake water quality (Huser et al., 2016b; Welch and Cooke 1999). The longevity of Al-treatment, however, has been highly variable, with beneficial effects lasting from just a few months to more than 40 years (Egemose et al., 2011; Garrison and Knauer 1984; Huser et al., 2011; Huser et al., 2016b; James et al., 1991). Reported binding effectiveness (measured as Al:Al-P or Al:$P_{Al}$ ratios) has varied by an order of magnitude, i.e., between 2.1 and 21.1 (Agstam-Norlin et al., 2020; Huser et al., 2011; Huser 2012; Jensen et al., 2015; Lewandowski et al., 2003; Reitzel et al., 2005; Rydin et al., 2000; Rydin and Welch 1999; Schütz et al., 2017).

Treatment longevity and effectiveness are influenced by multiple factors including external loading, lake morphology, mixing regime, bioturbation, Al dose, and application method. When external P loading remains elevated after Al-treatment, ongoing P inputs can overwhelm Al binding capacity, resulting in new mobile sediment P accumulation over the treated layer and elevated internal P loading (Huser et al., 2016c). The ratio of lake to watershed areas (WA:LA) is an important predictor of treatment longevity, as nutrient levels in lakes with large watersheds relative to lake area are more controlled by external sources than lakes with relatively small watersheds (Huser et al., 2016b; Sas 1990). The rate of water renewal (i.e. residence time) can strongly affect treatment longevity, as a long residence time can result in P being retained in the lake and sediment instead of moving to downstream water bodies (Sas 1990).

Morphology can decrease overall binding efficiency in lakes with steep bed slopes through translocation and focusing of the added Al-mineral ($Al(OH)_3$) in deep areas. This is because freshly

* Corresponding author.
*E-mail address:* oskar.agstam@slu.se (O. Agstam-Norlin).

https://doi.org/10.1016/j.watres.2021.117267

formed, amorphous Al-minerals have a similar density to the surrounding, organic-rich sediment and a large fraction can rapidly (within 6 months) move down-slope to deeper areas (Huser 2017). This can result in excess accumulation of Al (Egemose et al., 2012; Huser 2012), and in the absence of available P, the Al-mineral starts to crystalize resulting in a lower surface area and a loss of P binding sites (Berkowitz et al., 2006; deVicente et al., 2008; Huser 2012; Huser 2017).

The Al dose used is another critical factor influencing longevity and effectiveness. In the past, Al doses were often inadequate to bind all potentially releasable sediment P. This was usually a result of old dosing calculation methods where dose was based on lake water alkalinity or hypolimnetic P instead of targeting the total pool of available mobile P in lake sediment (Cooke et al., 2005). Greater Al doses tend to bind more P in the sediment, which can increase longevity (Huser et al., 2016b; Sas 1990). However, adding too much Al, or using excessive amounts of Al in one application, will generally decrease binding efficiency due to crystallization of the Al-mineral before P binding occurs (deVicente et al., 2008), with potentially negative effects on longevity (Agstam-Norlin et al., 2020; Huser 2012).

Mixing regime is also important. Shallow, polymictic lakes tend to have shorter treatment longevity (Huser et al., 2016b; Welch and Cooke 1999), but the underlying mechanisms behind this are not entirely understood. Shallow lakes generally have lower water volume per area of sediment, and thus alkalinity needed to support higher doses is often lower and as a consequence, lower Al-doses have generally been added which per se decreases longevity (Huser et al., 2016b). Dense populations of macrophytes often found in shallow polymictic lakes may increase internal loading by translocation of sediment P to the water column (through plant decomposition) or by stabilizing the water column and limiting oxygenation, thus increasing the potential for internal loading (Welch and Kelly 1990). In addition, benthic feeding fish often have much greater negative impacts on nutrients and water quality in small shallow lakes relative to deeper, stratified systems (Bajer and Sorensen 2014).

Bioturbation caused by benthic feeding fish, e.g., carp (*Cyprinus carpio)* can deepen the active sediment layer and mass of P available for release to the water column (Huser et al., 2016a), especially in shallow polymictic lakes (Parkos et al., 2003; Weber and Brown 2009). On the other hand, enhanced sediment mixing by benthic feeding fish may result in higher Al:$P_{Al}$ binding efficiency by increasing the chance of Al to encounter P before crystallization of the mineral occurs (Huser et al., 2016a).

Different Al-application procedures have been used historically, with the most commonly used method being application of Al in solution to the water column and allowing the newly formed mineral to settle to the sediment surface (Welch and Cooke 1999). However, there is a newly developed technique where Al is injected into surficial sediment (10–15 cm deep, (Schütz et al., 2017)). The injection method minimizes Al transport and focusing because the Al-floc is trapped in the vertical sediment profile, which can lead to improved binding efficiency (Agstam-Norlin et al., 2020). Solid forms of Al (pellets) have also been used historically (Cooke et al., 2005), but a shift to liquid application occurred after the 1970s. However, binding effectiveness using pellets may be greater relative to liquid application in some cases (Agstam-Norlin et al., 2020). Treatment using Al pellets has been tested recently in countries lacking equipment needed for liquid application (Kuster et al., 2020).

Here, we determined the longevity of Al treatment and assessed the factors influencing effectiveness and longevity of Al-treatment using available monitoring data for six Swedish lakes of varying size, morphology, Al dose applied, and application technique (Table 1). Nutrient-related water quality variables including historical trends for surface water chemistry/quality were analyzed. Internal P release rates were calculated pre- and post-treatment when adequate data were available.

## 2. Methods

### 2.1. Study sites

Additional information about all study sites, including locations, is presented in Agstam-Norlin et al. (2020).

#### 2.1.1. Dimictic lakes

Six lakes situated within a 30 km radius of Stockholm city were included in the study. Flaten is a 63 ha lake with a 403 ha catchment mostly covered by forest and recreational areas (70%). Al-application was performed in 2000 with combined sediment injection and hypolimnetic water application using a total of 24 tons of Al (as pre-hydrolyzed aluminum chloride (PAC, PAX XL100). Areas of the lake where water column depth was between 6 and 9 m were treated with 30 g/m$^2$ Al injected into the sediment and additionally 10 g/m$^2$ dispersed in the water near the sediment water interface. Deeper areas (9–10 m) were dosed with 40 g/m$^2$ via sediment injection and 14 g/m$^2$ to the water. At the deepest point of the lake (10–14 m) 53 g/m$^2$ was injected and 17 g/m$^2$ added to the water (Table 1). Partial water application was conducted to precipitate P already released from sediment.

Trekanten is 13.5 ha lake with a catchment of 60 ha dominated by urban park areas. Al-treatment was performed in 2011 using

**Table 1**
Background information including morphology, sediment characteristics and aluminum treatment details. Al-treatment method includes sediment treatment (S) and water treatment (W). Sediment data reworked from Agstam-Norlin et al. (2020), with permission.

| Lake | Lejondalssjön | Flaten | Trekanten | Långsjön | Bagarsjön | Malmsjön |
|---|---|---|---|---|---|---|
| Max depth (m) | 14.0 | 13.1 | 6.6 | 3.3 | 5.6 | 6.8 |
| Mean depth (m) | 7.5 | 8.7 | 3.6 | 2.2 | 2.3 | 4.7 |
| Lake area (ha) | 272 | 63 | 14 | 29 | 6 | 89 |
| Osgood index | 4.6 | 11.0 | 9.5 | 3.7 | 8.8 | 5.0 |
| Watershed (ha) | 1660 | 403 | 60 | 243 | 135 | 1175 |
| Watershed:Lake area (WA:LA) | 6.1 | 6.4 | 4.3 | 8.4 | 22.5 | 13.2 |
| Volume (Mm$^3$) | 20.5 | 4.6 | 0.6 | 0.6 | 0.2 | 4.2 |
| Residence time (years) | 7.0 | 4.0 | 3.0 | 0.8 | 0.8 | 1.3 |
| Mean Al dose (g/m$^2$) | 25 | 61 | 60 | 75 | 50 | 60 |
| Treatment year(s) | 1991–1993 | 2000 | 2011 | 2006 | 1997 | 2007 |
| Al-treatment method | W | S | S | S | W | S |
| Treatment chemical | PAX XL60 | PAX XL 100 | PAX XL 100 | PAX XL100 | PAX XL 60 | PAX XL 100 |
| Mean Al:$P_{Al}$ | 11.4 | 9.0 | 10.7 | 12.9 | 16.0 | 14.6 |
| Mean Mob-P (g/m$^2$) | 7.3 | 4.9 | 8.6 | 2.8 | 5.3 | 3.5 |
| Al dose:Mob-P | 3.4 | 12.4 | 7.0 | 21.4 | 9.4 | 16.9 |

sediment injection of PAC (PAX XL 100). 60 g $Al/m^2$ was applied to all lake areas where water depth was > 4 m, with a total Al dose of 5 tons (Table 1).

Lejondalsjön has a surface area of 272 ha with a catchment of 1660 ha that is dominated by forest (58%), agriculture (crop- and pasture fields, 16%), and urban areas (2%). Al-treatment was performed during three consecutive years where a total of 28.6 tons of Al (as PAC, PAX XL 60) was applied to the hypolimnetic lake water. In 1991, a relatively low Al dose (10 $g/m^2$, total of 5 tons) was applied to the northern half of the lake where water depth was >10 m. In 1992, three small test areas of the northern part of the lake (total 12 ha) were treated with a total Al dose of 0.6 tons but with different areal doses (25, 50 and 75 $g/m^2$). The final PAC application in 1993 treated areas with water column depths exceeding 10 m (25 $g/m^2$, Table 1).

Bagarsjön is an urban, 6 ha lake with a catchment area of 135 ha that includes mainly residential (80%) and small park/forest areas. Al (as PAC, PAX XL 60) was applied in the hypolimnetic water at a total of 2 tons at a rate of 50 $g/m^2$ to areas with water depths greater than 2 m (Table 1).

#### 2.1.2. Polymictic lakes

Långsjön is a 29 ha urban lake. The 243 ha catchment is mainly residential with some recreational areas. Al (PAC, PAX XL100) was applied by direct sediment injection with different doses for different depth zones of the lake. A dose of 25 $g/m^2$ was applied at water column depths between 1.5 and 2 m, 50 $g/m^2$ at water column depths between 2 and 2.5 m, and 75 $g/m^2$ from a water depth of 2.5 m to the maximum depth (3.3 m, Table 1).

Malmsjön is an 89 ha lake in an 1175 ha catchment dominated by forest (57%), farmland (23%), and urban areas (11%). In total, 53 ton of Al (PAC, PAX XL100) was applied using sediment Al injection. The area specific Al dose was 60 $g/m^2$ at all lake locations with a water column depth exceeding 1 m (Table 1).

### 2.2. Data handling

Average, annual growing season (May-Aug) TP, Chlorophyll a (Chl_a), and Secchi disk depth (SD) were used to describe differences in surface water quality between pre- and post-treatment periods. In stratified lakes, the increase in hypolimnetic TP during the growing season and corresponding hypolimnetic water volumes were used to calculate internal P loading rates (Li, $mg/m^2/day$) (Nürnberg 2009). A 4-year pre-treatment period of annual means for SD, Chl_a, surface TP, and Li were compared with following post treatment 4-year periods to compare whether the parameters had changed due to Al-treatment. Four-year periods were used in order to limit the effect of natural interannual variation.

Longevity was estimated using three different methods. For lakes where longevity had been reached (i.e. no improvement was detected compared to pre-treatment after a period of time), longevity was defined as the year when annual mean (or single August data point, when seasonal data was lacking) TP concentration exceeded one standard error of mean (SEM) below mean pre-treatment TP (4-year period). For lakes where water quality improvements were still ongoing throughout the monitoring period (e.g., Malmsjön, Långsjön and Bagarsjön), but TP showed a linear increase post Al-treatment, we estimated longevity using linear regression and extrapolation. Longevity was estimated as the time point where the linear regression exceeded one SEM below pre Al-treatment mean TP (4-year period). Finally, the longevity predicting equation (LPEq) of Huser et al. (2016b) was used to estimate treatment longevity in cases where there was no deterioration of post-treatment water quality through the end of the dataset (Eq. (1)). Three variables described 82% of the variation in Al-treatment longevity in Eq. (1), including Al dose, watershed area to lake area ratio (WA:LA) and Osgood index (Osgood 1988). It should be noted that this method for calculating longevity is almost certainly an underestimate as the regression developed weighted lakes highest where no external nutrient reduction occured. In addition, longevity predicted using LPEq has an arbitrary endpoint (i.e., longevity is considered to end when surface TP is 50% of the pre-treatment value), and thus is not a true representation of longevity of positive treatment effects.

$$\log(Longevity) = -0.5 + 1.3 \times \log(Al\ dose) - 0.79 \times \log(WA : LA) + 0.37 \times \log(Osgood\ index) \quad (1)$$

Graphical exploration for effect size was used to evaluate effect and longevity of Al-treatment and Principal component analysis (PCA) was used to evaluate driving factors for longevity. All statistical analyses was performed in JMP (SAS institute Inc., version 11.0.0).

## 3. Results

Nutrient-related water quality variables improved after Al-treatment in all lakes (Table 2). Flaten, Långsjön, Lejondalssjön, Bagarsjön and Malmsjön had changes in all measured water quality variables during the first 4 years after Al-treatment, whereas

**Table 2**
Mean, standard error and percent change for each post 4 year period compared to pre aluminum treatment conditions. Pre denotes parameter mean values of a 4-year period prior to treatment. Following columns shows similar information for 4-year periods post Al-treatment (e.g. 1–4 y). Column "Δ" denotes the percent change in each parameter. TP and Chl a in μg/L, SD in meters and Li in $mg/m^2/day$. N denotes mean annual observations (Li: mean number of TP observations below hypolimnion).

| Lake | Parameter | Pre | 1–4 y | Δ | 5–8 y | Δ | 9–12 y | Δ | 13–16 y | Δ | 17–20 y | Δ | N |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Bagarsjön | SD | 2.2 ± 0.4 | 3.5 ± 0.4 | 59% | 3.1 ± 0.3 | 41% | 2.5 ± 0.3 | 14% | 2.7 ± 0.2 | 23% | 3.0 ± 0.2 | 36% | 1 |
| | TP | 60.3 ± 9.1 | 24.8 ± 4.4 | −59% | 37.3 ± 1.4 | −38% | 43.0 ± 2.5 | −29% | 23.8 ± 2.5 | 61% | 20.8 ± 2.4 | −66% | 1 |
| Flaten | SD | 2.9 ± 0.4 | 6.4 ± 0.4 | 121% | 6.3 ± 0.5 | 117% | 4.8 ± 0.5 | 66% | 5.5 ± 0.4 | 90% | *ND* | *ND* | 4 |
| | Chl a | 12.7 ± 2.8 | 2.8 ± 0.3 | −78% | 3.2 ± 0.6 | −75% | 5.4 ± 1.0 | −57% | 4.7 ± 1.0 | 63% | *ND* | *ND* | 4 |
| | TP | 33.3 ± 5.6 | 9.2 ± 0.4 | −72% | 8.9 ± 0.6 | −73% | 9.5 ± 0.6 | −71% | 11.0 ± 1.6 | 67% | *ND* | *ND* | 4 |
| | Li | 3.4 ± 0.4 | 0.2 ± 0.1 | −94% | 0.2 ± 0.1 | −94% | 0.3 ± 0.1 | −91% | 0.3 ± 0.1 | 91% | *ND* | *ND* | 19 |
| Lejondalssjön | SD | 2.9 ± 0.3 | 4.4 ± 0.2 | 52% | 4.3 ± 0.2 | 48% | 4.1 ± 0.2 | 41% | *ND* | ND | *ND* | *ND* | 4 |
| | TP | 35.3 ± 2.6 | 25.0 ± 1.4 | −29% | 27.2 ± 2.7 | −23% | 24.0 ± 1.4 | −32% | *ND* | ND | *ND* | *ND* | 4 |
| | Li | 5.0 ± 0.5 | 1.6 ± 0.4 | −68% | 2.9 ± 1.0 | −42% | 3.7 ± 0.2 | −26% | *ND* | ND | *ND* | *ND* | 11 |
| Långsjön | SD | 0.9 ± 0.1 | 1.5 ± 0.1 | 67% | 1.6 ± 0.1 | 78% | 1.7 ± 0.1 | 89% | ND | ND | *ND* | *ND* | 7 |
| | Chl a | 53.6 ± 7.5 | 22.7 ± 2.4 | −58% | 17.0 ± 4.9 | −68% | 16.4 ± 2.6 | −69% | ND | ND | *ND* | *ND* | 4 |
| | TP | 97.5 ± 5.7 | 33.3 ± 1.6 | −66% | 38.2 ± 3.1 | −61% | 46.5 ± 2.6 | −52% | ND | ND | *ND* | *ND* | 4 |
| Malmsjön | TP | 96.5 ± 25.6 | 19.3 ± 3.3 | −80% | 28.8 ± 4.5 | −70% | 43.8 ± 8.7 | −55% | ND | ND | ND | *ND* | 1 |
| Trekanten | SD | 3.0 ± 0.2 | 2.8 ± 0.2 | −7% | 3.6 ± 0.2 | 20% | *ND* | ND | *ND* | ND | *ND* | *ND* | 4 |
| | Chl a | 14.4 ± 2.6 | 7.2 ± 0.8 | −50% | 5.9 ± 0.8 | −59% | *ND* | ND | *ND* | ND | *ND* | *ND* | 5 |
| | TP | 53.1 ± 7.0 | 24.3 ± 2.5 | −54% | 23.4 ± 1.4 | −56% | *ND* | ND | *ND* | ND | *ND* | *ND* | 5 |
| | Li | 3.3 ± 0.9 | 0.2 ± 0. | −94% | 0.1 ± 0.1 | −97% | *ND* | ND | *ND* | ND | *ND* | *ND* | 12 |

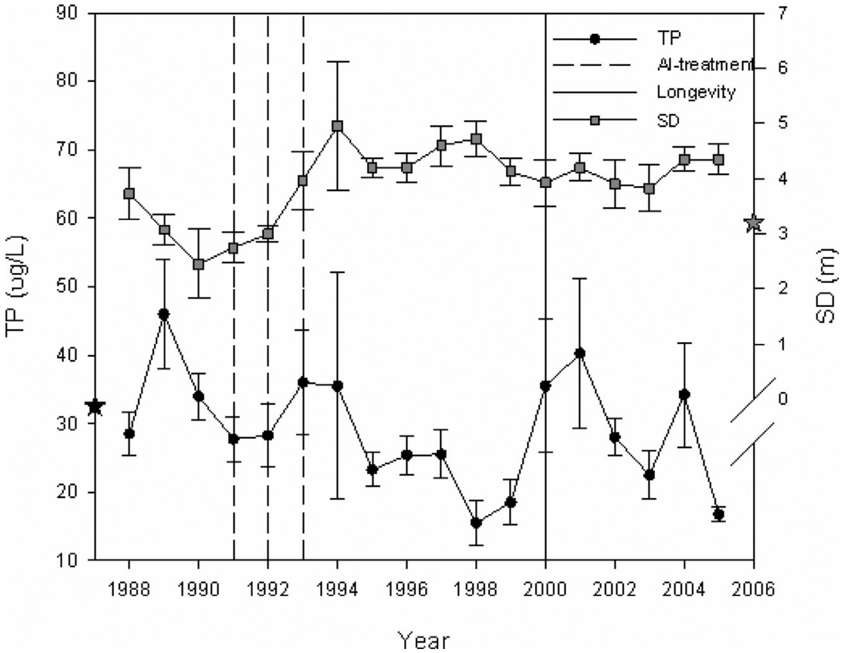


**Fig. 1.** Annual growing season surface water TP and SD mean and SEM (May-Aug) in Lejondalssjön. Longevity was estimated as the year when one SEM below mean pre-treatment (4-year period) was exceeded by annual post-treatment mean TP. Stars with corresponding color to time series at each Y-axis indicate one SEM below (or above for SD) mean for years before Al-treatment. Observations: SD=70, TP=74.

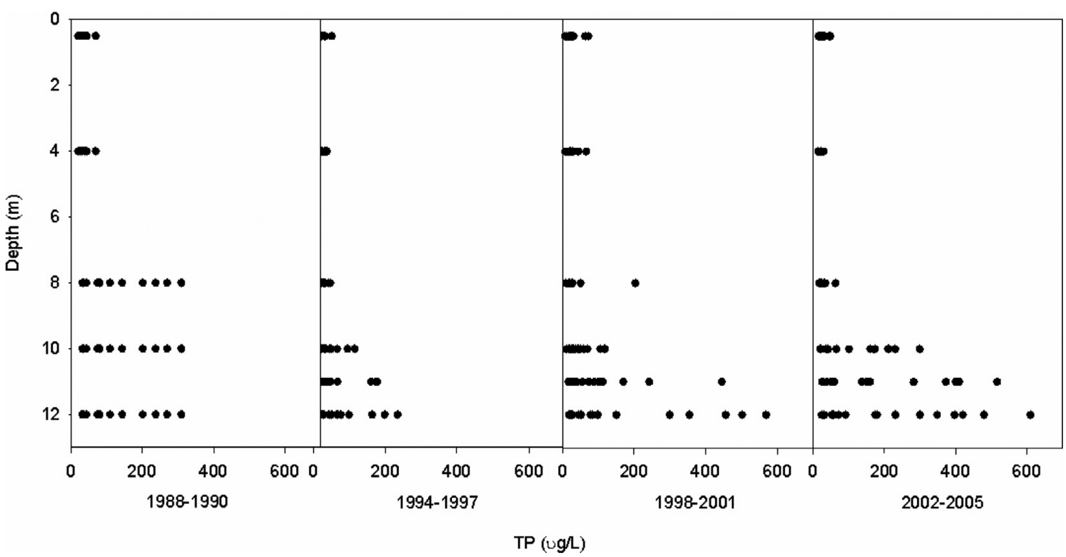


**Fig. 2.** Water column depth profiles showing concentrations of TP at different depth intervals in Lejondalssjön. 1988–1990 represents pre Al-treatment and following consecutive 4-year period represents post-treatment observations of TP. Pre-treatment data was collected as mixed water samples collected between 0 and 4 m, and 8–12 m. Post-treatment observations was collected at stated depths according to the graph. Observations: 1988–1990=62, 1994–1997=90, 1998–2001=95, 2002–2005=96.

Trekanten showed no initial increases in SD, but TP declined substantially (Table 2). Flaten, Långsjön, Malmsjön and Trekanten had changes in all measured water quality variables at the end of each available data series (16, 12, 12 and 12 years post treatment, respectively). Lejondalssjön had improved TP until 7 years post treatment and showed deeper SD throughout the monitoring period (Table 2).

## 3.1. Dimictic lakes

### 3.1.1. Lejondalssjön

TP decreased by 29% from 35.3 to 25.0 µg/L (mean values) during the first 4 year post-treatment. During the second post-treatment 4-year period the annual TP mean exceeded the pre-treatment breaking point (one SEM below annual TP mean) and longevity of the TP decline was 7 years (Table 2, Fig. 1). Li decreased as well, from a mean of 5.0 to 1.6 mg/m$^2$/day (68%) over the first 4-year period post Al-treatment, but during subsequent 4-year periods the Li rate started to return to pre-treatment conditions (2.9 mg/m$^2$/day) (Table 2, Fig. 2). A SD increase of >40% was sustained throughout the available data series, from an average of 2.9 m before treatment to an average of 4.1 m at the end of the data series (12 years post treatment).

### 3.1.2. Flaten

Surface TP averaged 33.5 µg/L before treatment and decreased to 9.2, 8.9, 9.5 and 11.0 µg/L during subsequent consecutive 4-year periods (4, 8, 12 and 16 years post-treatment), meaning the longevity of treatment with respect to surface water TP exceeded the end of this study. There was a decrease Chl_a, with con-

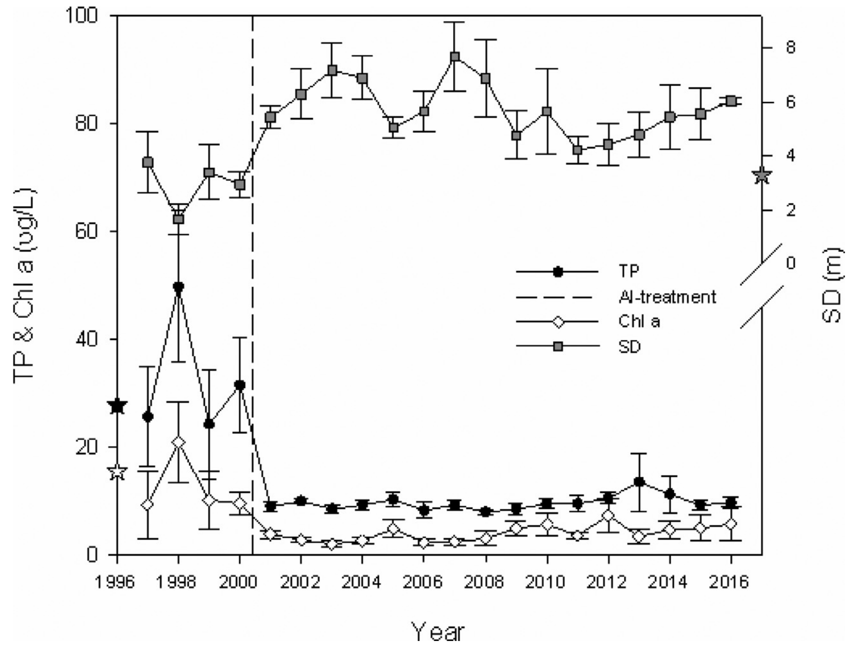


**Fig. 3.** Annual growing season surface water TP, Chl a and SD mean and SEM (May-Aug) in Flaten. Longevity was estimated using LPEq. Stars with corresponding color to time series at each Y-axis indicate one SEM below (or above for SD) mean for years before Al-treatment. Observations in total time series: Chl *a* = 85, SD=88, TP=77.

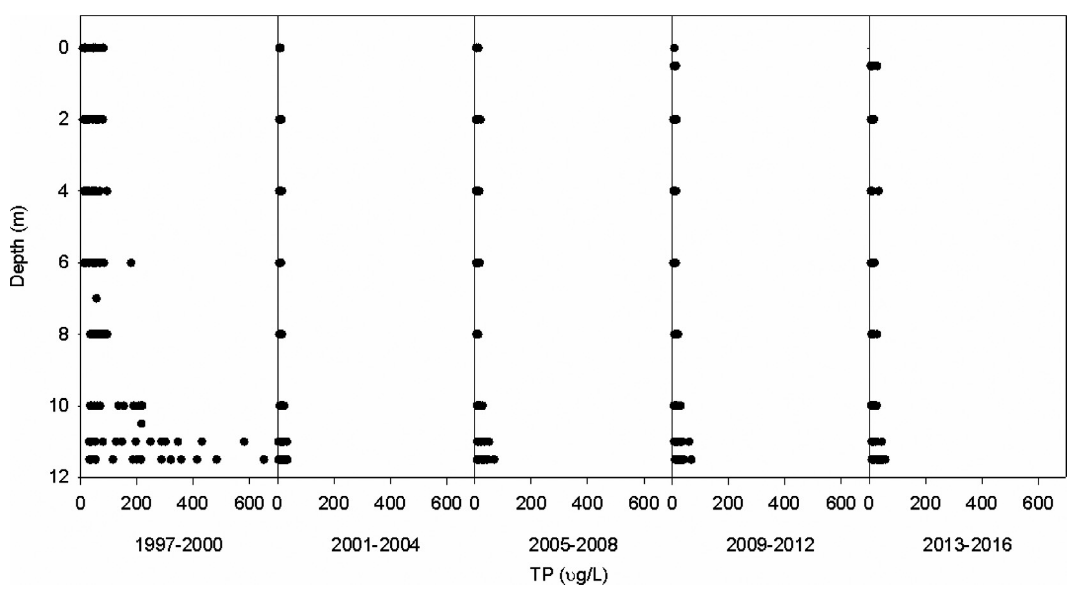


**Fig. 4.** Water column depth profiles showing concentrations of TP at different depth intervals in Flaten. 1997–2000 represents pre Al-treatment and following consecutive 4-year periods represents post-treatment observations of TP. Observations: 1997–2000=150, 2001–2004=125, 2005–2008=129, 2009–2012=129, 2013–2016=120.

centrations declining from a pre-treatment mean of 12.7 μg/L to 2.8, 3.2, 5.4, and 4.7 μg/L respectively for 4, 8, 12 and 16 years post-treatment. SD significantly improved as well, increasing from a pre-treatment mean of 2.9 m to 6.3, 6.4 4.8, and 5.5 m during 4, 8, 12 and 16 years post-treatment. Due to continued improvement of all nutrient related water quality variables (Fig. 3) throughout the duration of monitoring, treatment longevity could not be calculated but was instead estimated using LPEq to 37 years (Huser et al., 2016b). Li decreased (90%) for all available post-treatment data, with a pre-treatment mean of 3.4 mg/m$^2$/day and post-treatment averages were all below 0.3 mg/m$^2$/day (Table 2, Fig. 3 and 4).

### 3.1.3. Trekanten

Pre-treatment TP averaged 53.1 μg/L and decreased to a mean of 24.3 μg/L during the first 4-years post treatment period and decreased further to 23.4 μg/L through the end of the data set (5–8 years post treatment) (Table 2, Fig. 5). Chl_a decreased from a 14.4 μg/L pre-treatment mean to 7.2 μg/L and 5.9 μg/L, 4 and 8 years post treatment, respectively. SD, however, did not improve until the three last years of the study record (Table 2 and Fig. 5). Li decreased by over 90% throughout the available data series, initially at 3.3 mg/m$^2$/day before treatment and decreasing to 0.2 and 0.1 mg/m$^2$/day at 4 and 8 years post Al-treatment, respectively (Fig. 6, Table 2). Longevity was estimated to 47 years, using LPEq.

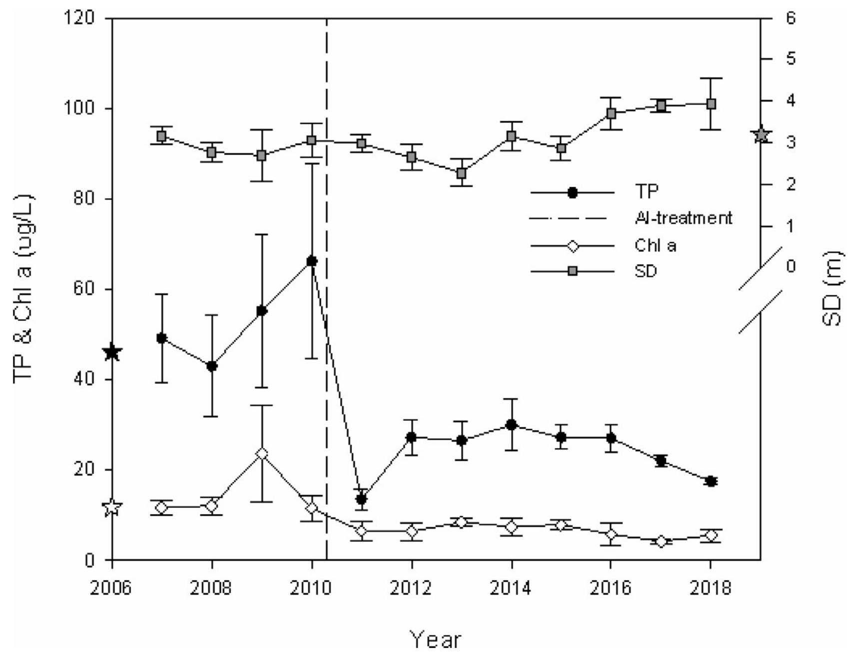


**Fig. 5.** Annual growing season surface water TP, SD and Chl a mean and SEM (May-Aug) in Trekanten. Longevity was estimated using LPEq. Stars with corresponding color to time series at each Y-axis indicate one SEM below (or above for SD) mean for years before Al-treatment. Observations in total time series: Chl $a$ = 54, SD=53, TP=55.

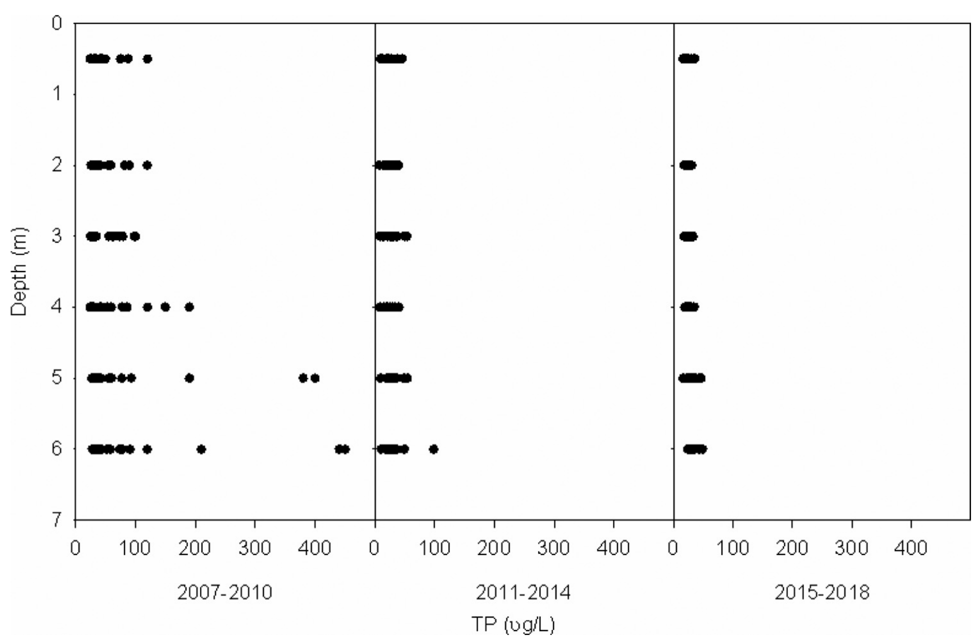


**Fig. 6.** Water column depth profiles showing concentrations of TP at different depth intervals in Trekanten. 1997–2000 represents pre Al-treatment and following consecutive 4-year period represents post-treatment observations of TP. Observations: 2007–2010=96, 2011–2014=96, 2015–2018=96.

*3.1.4. Bagarsjön*

During the first two 4-year post-treatment periods (e.g. eight years), TP was substantially lower compared to pre-treatment, decreasing from a mean of 60.3 μg/L to 24.8 and increasing to 37.3 μg/L respectively. During the following 4-year period (9–12 years post treatment) an increasing trend of TP was observed (mean=43.0 μg/L) (Table 2, Fig. 6). However, due to other management efforts such as freshwater dilution and hypolimnetic aeration during the 16, and 20 years post treatment periods, TP decreased again and was lower than pre-treatment conditions. SD did increased with 59% and 41% at the first and second 4-year period post Al-treatment, respectively. At years 9–12 post-treatment SD had decreased to 14% improvement. (Table 2, Fig. 7). Li was not analyzed due to lack of available data. Treatment longevity was determined to 14 years (Fig. 7).

*3.2. Polymictic lakes*

*3.2.1. Långsjön*

All variables (TP, SD, and Chl_a) improved by over 50% through the end of the study data series (12 years post Al-treatment). SD increased from a mean of 0.9 m to 1.7 m during the final post-treatment period. Chl_a decreased from a pre-treatment mean of 53.6 μg/L to 16.4 μg/L and TP decreased from a mean 97.5 μg/L to 46.5 μg/L 4, 8, and 12 years post-treatment. (Table 2, Fig. 8). Longevity using the post-treatment,

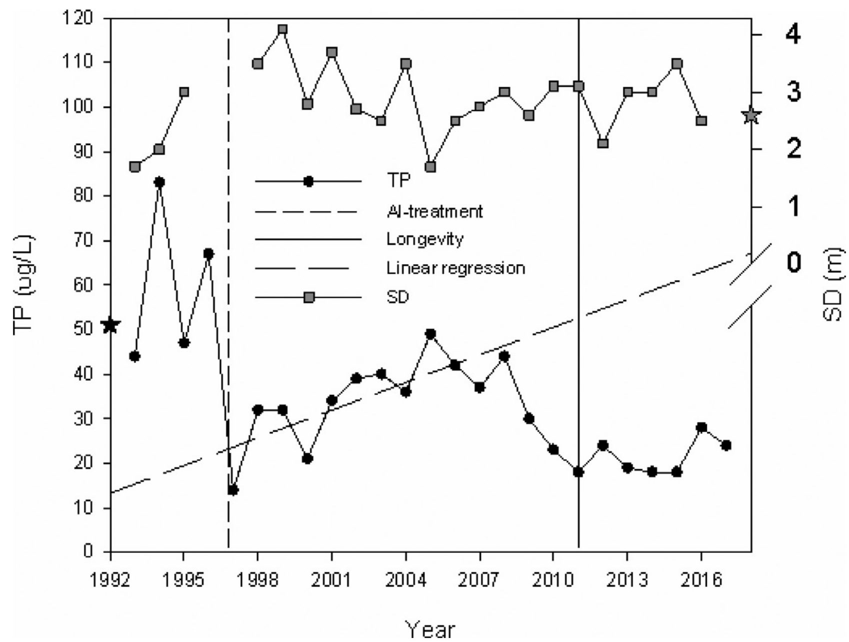


**Fig. 7.** Annual August surface water TP observation in Bagarsjön. Longevity was estimated as the time point where the linear regression exceeded one SEM below pre Al-treatment mean TP (4-year period). Star at each Y-axis indicate one SEM below (above for SD) mean pre-treatment TP and SD (4-years period). Observations in total time series: SD=22, TP=25. The decrease in TP after 2008 was likely due to installed hypolimnetic oxygenating system and not Al treatment.

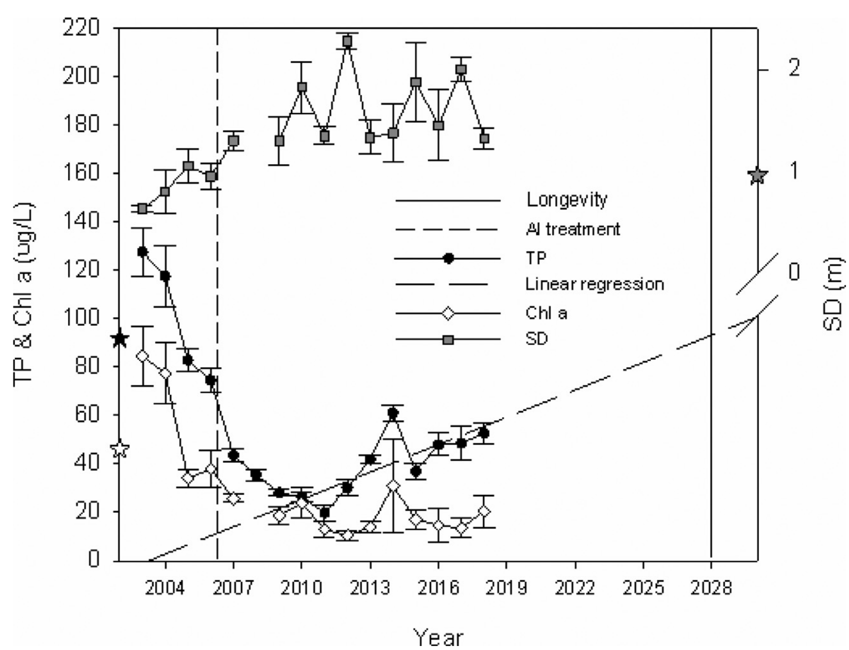


**Fig. 8.** Annual growing season surface water TP, SD and Chl a mean and SEM (May-Aug) in Långsjön. Longevity was estimated as the time point where the linear regression exceeded one SEM below pre Al-treatment mean TP (4-year period). Stars with corresponding color to time series at each Y-axis indicate one SEM below (or above for SD) mean for years before Al-treatment. Observations in total time series: Chl *a* = 57, SD=56, TP=114.

linear increase in surface water TP was estimated to be 21 years (Fig. 8).

### 3.2.2. Malmsjön

Surface water TP concentrations decreased by over 50% in the three post Al-treatment periods (4, 8 and 12 years post treatment, the entire dataset). Pre-treatment TP averaged 96.5 µg/L and was 43.8 µg/L at 12 years post treatment. (Table 2, Fig. 9). Longevity, using the linear increase in surface TP after treatment, was estimated to be 20 years. No other water quality data were available for Malmsjön.

## 3.3. Factors influencing longevity of Al-treatment

Principal Components Analysis (PCA) including factors in Table 1 was used to determine factors driving longevity and explained 79% of the variation in the data set with two components. Longevity was positively correlated to Osgood index suggesting that deeper lakes (dimictic) have higher longevity than small polymictic lakes. The analysis also showed that larger Al-doses were correlated with greater longevity. Al:$P_{Al}$ was negatively correlated to longevity, meaning that poor binding efficiency (i.e. high Al:$P_{Al}$) results in lower longevity. Finally, WA:LA also

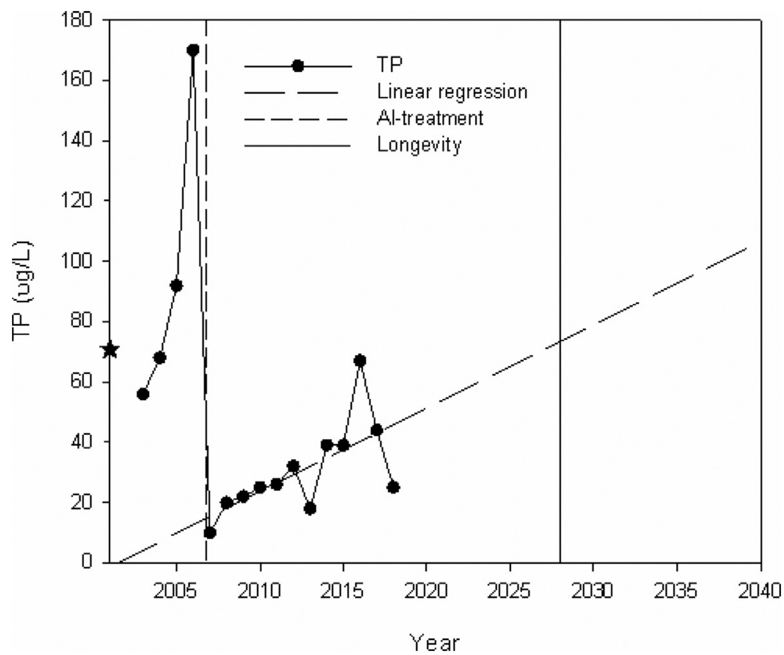


**Fig. 9.** Annual August surface water TP observation in Malmsjön. Longevity was estimated as the time point where the linear regression exceeded one SEM below pre Al-treatment mean TP (4-year period). Star at Y-axis indicate one SEM below mean pre-treatment TP (4-years period). Observations in total time series: TP=25.

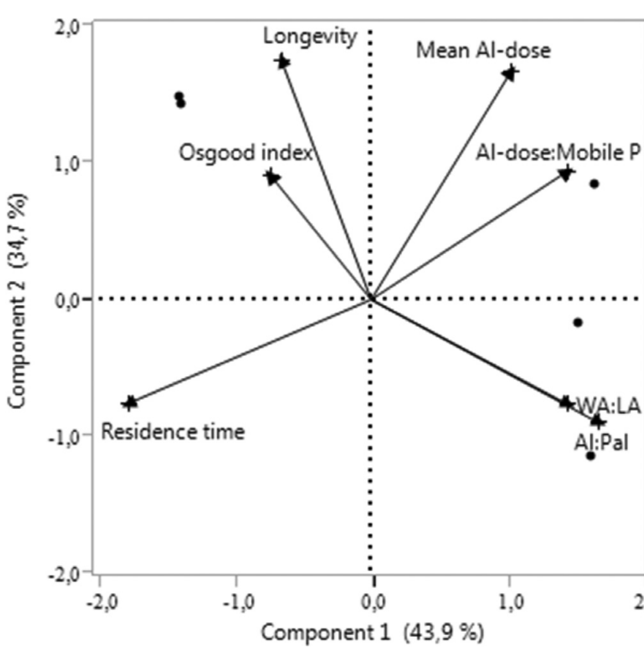


**Fig. 10.** Biplot from principal component analysis on variables from Table 1, explaining 79% of the total variation.

had a negative correlation to longevity, suggesting that lakes with greater watershed area relative to lake area have shorter longevities (Fig. 10).

## 4. Discussion

### 4.1. Water quality response to Al-treatment

#### 4.1.1. Dimictic lakes

Three dimictic lakes (Lejondalssjön, Trekanten and Flaten) were historically monitored at a level required determining the effect of Al-treatment on surface water quality. The data series from these lakes included monthly (May-September) measurements of Chl_a, SD and TP for both surface water and depth intervals covering the vertical water profile (except for Lejondalssjön, which lacked pre-treatment Chl_a data). This provided the possibility to include within-year (May to August) variation when studying historical trends in surface water quality. Vertical profiles with samples taken from different water depths enabled calculation of historical internal loading rates, which are generally a better indicator of the effects of Al-treatment. However, this direct quantification of internal loading is only applicable to dimictic lakes, where the lake remains stratified and hypolimnion remains stable throughout the growing season.

*4.1.1.1. Lejondalssjön.* Internal loading rates decreased 68% during the first 4-year period post Al-treatment. During the following 4-year post-treatment periods, however, internal loading gradually returned to levels not different to pre-treatment conditions (Table 2, Fig. 1& 2). This is likely due to a combination of factors. The high amount of legacy (mobile) sediment P remaining after Al-treatment (Agstam-Norlin et al., 2020) (Table 1) likely overwhelmed the Al-treatment, as was the case for numerous historical treatments where Al doses were added that were inadequate to bind the entire excess mobile P pool (Huser et al., 2016b; Welch and Cooke 1999). Even though Li improved substantially after treatment (at least initially), surface water TP only decreased by 29% during the first 4-year period. This can likely be explained by ongoing, excess external P inputs.

Limited treatment longevity has been seen elsewhere when external loading was not reduced adequately or inadequate Al doses were used (Egemose et al., 2011; Garrison and Knauer 1984; Huser et al., 2011; Huser et al., 2016b; James et al., 1991; Mehner et al., 2008). Thus the limited treatment longevity in Lejondalssjön is likely explained by a combination of continued excess external loading and the low Al dose (25 g/m$^2$) added. Dose calculation methods based on other variables besides sediment P were common before 2000. For Lejondalssjön, Al dose was calculated based on the internal P loading rate (Kennedy et al., 1987). One problem with this approach is that it only accounts for sediment P release during one year, even though the sediment often contains enough mobile P to support many years of internal loading. Without looking at sediment P content, it is impossible to determine how much mobile P exists in the sediment, and thus underdosing of Al is a likely result (Kuster et al., 2020).

*4.1.1.2. Flaten.* When this lake was treated in 2000, the understanding of Al dosing had improved substantially and was based on the available pool of mobile P (labile organic P, P bound to Fe, and loosely sorbed/porewater P) in the lake sediment (Rydin 2000). The mean dose of Al across the lake was 61 g/m$^2$ (Schütz et al., 2017), however dose was adjusted higher or lower depending on the amount of mobile sediment P in different locations. The application technique employed was also different compared to most Al-treatments. A combination of sediment injection and hypolimnetic water application was used, where approximately a fourth of the Al dose was applied to the water and the rest injected into the sediment. Surface water TP concentrations decreased by over 60% due to Al-treatment and this decrease was sustained through the end of the data series (Table 2, Fig. 3 & 4). Chl_a had a similar response and remain reduced throughout the study period. SD increased more than two fold, which also was sustained through the end of the data series.

*4.1.1.3. Trekanten.* The most recent treatment presented in this study was performed in 2011 in Trekanten, where a similar dose to that used in Flaten was applied (60 g/m$^2$). However, the application technique differed slightly and only sediment injection was used (i.e., no water application). As with Flaten, the Al dose was determined from the amount of mobile P in the sediment. The water quality data covered only two 4-year periods post treatment (i.e. 8 years) and showed a twofold decrease of TP and Chl_a throughout the measurement period (Table 2, Fig. 5). Because internal loading rates decreased by nearly 100%, but the lake water still has a TP concentration of nearly 25 µg/L, it seems likely that external sources of P are still elevated. This may, to some extent, be due to sewage water pipes that had been incorrectly connected to storm water runoff pipes between 2006 and 2013, unfortunately contributing excess external P load during these years. The excess external loading does not seem to have had an effect on internal loading (Fig. 6), but if not controlled, it is likely internal loading will return (Huser et al., 2011).

While SD did not initially improve post treatment, there was an increasing trend from 7% during the first 4-year period to 20% increase at the end of the data set (5–8 years). Similar, delayed effects have been seen after Al-treatment in a number of US lakes (Huser et al., 2011). The lack of immediate response in SD does not seem to be controlled by algal presence, because Chl_a decreased by 50%. This suggests a number of alternatives, e.g., sediment resuspension due to winds or benthic feeding fish causing sediment mixing (bioturbation), turbid storm water inflows, or increasing water color (total/dissolved organic carbon) that has been shown to be occurring in boreal regions across Sweden and elsewhere (de Wit et al. 2016; Huser et al., 2012; Monteith et al., 2007). Unfortunately, the lack of data did not allow us to explore any of these possible effects.

*4.1.1.4. Bagarsjön.* Whereas the previously described dimictic lakes were monitored in enough detail to calculate historical trends for Li, data for Bagarsjön only included surface water TP and SD at one occasion per year (August). Thus, it was not possible to calculate Li and within year variation (seasonal mean) could not be included in the analysis. However, the effect of Al-treatment on surface water TP was clear for the initial 4-year period (Fig. 7). Surface water TP began to increase after treatment but decreased again during the last two 4-year periods post treatment (from 2009). This decrease is likely a result of another separate management action starting in 2009, when an aeration pump was installed in order to keep hypolimnetic water oxygenated. There was no clear change in SD after 6 years post Al-treatment (including after aeration started).

#### 4.1.2. Polymictic lakes

Because polymictic lakes are generally shallow and well or partially mixed, the lack of a well-defined hypolimnion makes it is difficult to calculate internal loading rates. Dynamic modeling would be needed in such cases but monitoring of inflows and outflows was not conducted or data available were too sparse to use dynamic or even simple mass balance models. Surface water quality trends, however, could be analyzed and used to estimate treatment effect, as has been done in previous studies (Huser et al., 2016b; Welch and Cooke 1999).

*4.1.2.1. Långsjön.* Historical water chemical data for TP, SD and Chl_a, together with water profile measurements of TP were available. Because the lake is polymictic, often no accumulation of TP occurs in the bottom water, which limits the possibility to estimate Li. However, there was a positive effect for all measured parameters for all 12 years of post-treatment data, with all variables improving by >50% (Table 2, Fig. 8). It should be noted that other management actions including freshwater dilution were performed simultaneously to Al-treatment. These actions will, of course, improve water quality due to dilution of nutrients in the lake. External loading remains elevated, however (Table 2), and will likely overwhelm the effects of Al-treatment in the future (see Longevity section below).

*4.1.2.2. Malmsjön.* Available water chemical data included surface water TP concentrations from single samples taken in August each year. The lack of water column profile measurements of TP precluded calculation of internal loading, making quantification of the direct effect of Al-treatment impossible. Despite the limited data available, the effect of Al-treatment on surface water TP was clear for the complete data record. Surface water TP started to increase after treatment (Table 2, Fig. 9), and TP was estimated to return to pre-treatment levels after 20 years.

### 4.2. Al-treatment longevity in polymictic and dimictic lakes

If external P sources are reduced sufficiently and Al is added at a dose necessary to inactivate the entire legacy sediment P pool, treatment longevity would theoretically be infinite. Once P is bound to Al, it is considered permanently inactivated (Welch and Cooke 1999) and the sediment layer with elevated aluminum bound P will, over time, be buried by new sediment. For most lakes discussed herein, however, external loading remains elevated, in some cases substantially (i.e., Malmsjön and Långsjön (Table 2)). In these cases, estimates of longevity are mainly a factor of how much external loading has been reduced, with Al dosing and treatment methods that affect how much Al was added relative to legacy sediment P (Kuster et al., 2020) and binding efficiency between Al and P (Agstam-Norlin et al., 2020; Huser 2012) having a smaller effect.

#### 4.2.1. Dimictic lakes

Nutrient cycling in Flaten is mainly controlled by in-lake processes rather than external sources due to the low WA:LA ratio (6.4) and low intensity of land use in the watershed. Further, the Al dose was relatively high (mean 61 g/m$^2$) and mostly applied directly into the sediment rather than to the water column, which likely increased binding efficiency (mean 9.0, Table 1) (Agstam-Norlin et al., 2020). Given that no increase has been detected for Li or surface water TP after Al-treatment, longevity in this case may be infinite. The LPEq-estimated treatment longevity in Flaten was

37 years (i.e. surface water TP increases to 50% of pre-treatment), which is above the mean treatment longevity (21 years) for dimictic lakes (Huser et al., 2016b). However, treatment longevity will likely be substantially longer than predicted using LPEq given no significant increase in Li or surface water TP has been detected to date.

The treatment of Trekanten was in many ways similar to the situation in Flaten, with an even lower WA:LA (4.3), similar sediment binding efficiency (mean Al:$P_{Al}$ = 10.7) (Agstam-Norlin et al., 2020), and a relatively high Al dose (mean = 60 g/$m^2$) injected exclusively into the sediment. Because water quality was still improved with no decline throughout the measurement period, a determination of total longevity was not possible. With an LPEq-estimated treatment longevity of 47 years, it is possible that total longevity (e.g., the time it takes for surface water TP to reach pre-treatment levels) may be a century or longer.

The treatment of Bagarsjön differed from the previous two cases (Al was applied exclusively to the water column), resulting in a lower binding efficiency (mean Al:$P_{Al}$=16.0, (Agstam-Norlin et al., 2020)). Additionally, nutrient cycling and availability is likely controlled mostly by external sources because the lake has a relatively small area compared to the watershed (WA:LA = 22.5). These two factors likely decreased longevity (14 years), which is lower than what would be predicted for an average dimictic lake (Huser et al., 2016b). Similarly, Lejondalssjön also had a shorter than expected longevity (7 years), which can be explained by other factors, namely a low dose (25 g Al/$m^2$, calculated using outdated methods) applied to the water column together with moderately high binding efficiency (mean Al:$P_{Al}$ = 11.4).

*4.2.2. Polymictic lakes*

The longevity of the Långsjön treatment was estimated to be 20 years using linear increase and extrapolation (Fig. 8) of TP. This is above the previously reported Al-treatment longevity of polymictic lakes (mean 5.7 years) according to Huser et al. (2016b). The much longer predicted longevity can partly be explained by the relatively low WA:LA (8.4), meaning that in lake processes are likely more important than external sources for nutrient cycling. Långsjön was treated with a relatively high Al dose (mean = 60 g/$m^2$) directly applied to the sediment, which led to a moderately high binding efficiency (mean Al:$P_{Al}$ = 12.9). Further, another restoration effort was simultaneously performed using freshwater dilution (30 l/s, since 2002), likely increasing longevity.

Treatment longevity for Malmsjön, which was estimated at 20 years using the linear increase in surface water TP after treatment (Fig. 9), was well over the typical range for polymictic lakes (Huser et al., 2016b). Similar to Långsjön, direct sediment Al application was done with a relatively high dose (60 g/$m^2$). Binding efficiency (mean Al:$P_{Al}$= 14.6) and the WA:LA (13.2) were moderate in comparison to the other study lakes. The generally shorter longevity in shallow, polymictic lakes is not solely due to lake characteristics, but because historically these types of lakes usually received lower Al doses, which per se results in short longevity. Because Långsjön and Malmsjön received relatively high Al doses (calculated using the mobile sediment P pool) applied directly into the sediment (increasing binding efficiency), longevity exceeded expectations compared to earlier treatments (Agstam-Norlin et al., 2020; Huser et al., 2016b).

*4.2.2. Factors influencing Al-treatment longevity*

In this study we showed that observed factors generally controlling longevity for the most part follow current understanding in the literature. Al-dose, WA:LA, and Osgood index (i.e. polymictic versus dimictic lakes) drove longevity for the study lake, similar to previous conclusions from Huser et al., al.(2016b) where 114 lakes were analyzed for factors controlling longevity, and 82% of the variation in longevity was explained by WA:LA, Al-dose and Osgood index. This is also supported by PCA analysis of the study lakes (Fig. 10), where a higher Osgood index (i.e. a greater chance for stratification) was positively related to longevity and WA:LA ratios were negatively correlated. Al-dose had a weak positive correlation to longevity, possibly explained by the similarity of dose across the study lakes (Table 1). A new factor, Al:$P_{Al}$ ratios, was also shown to control longevity, with greater ratios (i.e. lower binding efficiency) being negatively correlated to longevity of treatment. This is plausible, as a lower binding efficiency means that less sediment P will be bound per unit Al added.

*4.3. Recommendations for assessing Li with water quality monitoring*

The duration and intensity of lake water quality monitoring before and after Al-treatment constrains the evaluation of effectiveness and longevity. In many situations it may not be sufficient to only measure changes in surface water nutrient related parameters such as SD, Chl_a, and TP in the lake. This is because other restoration efforts may be used simultaneously and/or external loading may remain high, potentially masking the effect of Al (or other treatments) designed to reduce internal phosphorus loading. Three dimictic lakes in this study were monitored in a sufficient manner to determine changes to Li associated with Al-treatment (Lejondalssjön, Flaten and Trekanten). Monthly water profile measurements of TP, with multiple samples collected through the vertical water profile and temperature every meter, made it possible to calculate the volume of the hypolimnion and the mass of TP in hypolimnetic water and follow the increase throughout the growing season when lake was stratified (Nürnberg 2009). Higher resolution monitoring is likely needed in polymictic systems, as in Huser et al. (2011) where biweekly observations were used to assess Li. This is due to the fact that internal loading events are likely shorter (i.e. periodic stratification and/or high pH events). The polymictic Långsjön was monitored monthly with vertical water chemical profiles and temperature at least every meter, however assessing Li using the same methods as for dimictic lakes was not possible due to the lack of stabile stratification during the growing season. Thus, in polymictic/shallow lakes, monitoring of flow and nutrient concentrations is needed in the inlets as well so that external and internal loading can be differentiated. Further, to assess Li accurately, it will generally be necessary to use dynamic modeling approaches, including spatially representative information on sediment P forms and potential flux, with data series covering pre and post periods of Al-treatment.

## 5. Conclusions

An Al dose adequate to bind legacy sediment P and appropriate treatment methods, depending on lake morphological characteristics, are crucial for sustainable restoration of surface water quality using Al-treatment. Al dose needs to be large enough to inactivate the entire pool of excess mobile P in the lake sediment, which drives the long term potential for internal loading. External load reduction is also important, as continued excess P loading will eventually overwhelm any measure designed to reduce sediment P release, as we showed in this study. Based on available data, the positive effects from Al-treatment in two lakes (Flaten and Trekanten) are likely to last a century or longer. On the other hand, lakes with high levels of excess external loading had (or will have) shorter treatment longevity ranging from 7 to 21 years. In addition, elevated external loading will limit the positive effects of reduced internal loading. In Lejondalssjön, internal loading was reduced substantially after Al-treatment (before the treatment was overwhelmed by new P), but surface water TP decreased by a much smaller amount due to continued elevated external loading.

Assuming adequate reduction of external loads, Al dose becomes the most important factor for restoration success. Newer methods, based on sediment P, have been developed and have led to substantially greater treatment longevities primarily due to greater Al doses being added. However, care should be taken with Al doses added during one treatment as high, single treatment doses decrease the overall efficiency of Al-treatment, as was seen in Malmsjön, and can limit the amount of sediment P that is inactivated per unit Al added. Doses should be split into smaller subtreatments to improve binding efficiency and increase the amount of P inactivated per unit Al added.

Finally, Al-treatment success was based (in most cases) on surface water quality improvement in this study, however the main goal of Al-treatment is to reduce P release from sediment. We attempted to evaluate reduction in internal loading for all lakes, but too often adequate data to do this were lacking. In cases where variables such as TP, Chl_a, and SD have unexpected responses, it is important to be able to evaluate whether these responses are due to internal, external or both types of loading. Proper monitoring, both before and after treatment, is a necessary requirement to be able to evaluate and optimize methods to reduce P loading and eutrophication in surface waters in the future.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgements

This study was funded by the Oscar and Lilli Lamm foundation (DO2014–0057) and FORMAS (The Swedish Research Council) grant number 2016–00846. The authors thank Fredrik Erlandsson at Stockholm Vatten, Dan Arvidsson at Botkyrka kommun, Eva Herlitz and Maria Elfström at Upplands-Bro kommun for sharing water quality monitoring data for the study lakes. The authors are grateful for helpful suggestions from two anonymous reviewers. Thanks to Sten-Åke Carlsson for sharing detailed information regarding Al-dosing and Al application procedures for the study lakes. All authors have contributed with data collection, data analysis and writing.

## References

Agstam-Norlin, O., Lannergård, E.E., Futter, M.N., Huser, B.J., 2020. Optimization of aluminum treatment efficiency to control internal phosphorus loading in eutrophic lakes. Water Res. 185, 116150 2020.

Bajer, P.G., Sorensen, P.W., 2014. Effects of common carp on phosphorus concentrations, water clarity, and vegetation density: a whole system experiment in a thermally stratified lake. Hydrobiologia 746 (1), 303–311.

Berkowitz, J., Anderson, M.A., Amrhein, C., 2006. Influence of aging on phosphorus sorption to alum floc in lake water. Water Res. 40 (5), 911–916.

Conley, D.J., Pearl, H.W., Howarth, P.W., Boesch, D.F., Seitzinger, S.P., Havens, K.E., Lancelot, C., Likens, G.E., 2009. Controlling Eutrophication: nitrogen and Phosphorus. Science 323, 1014–1015.

Cooke, D.W., Welch, E.B., Peterson, S.A., Nichols, S.A., 2005. Restoration and Management of Lakes and Reservoirs. CRC Press, Boca Raton.

de Wit, H.A., Valinia, S., Weyhenmeyer, G.A., Futter, M.N., Kortelainen, P., Austnes, K., Hessen, D.O., Räike, A., Laudon, H, Vuorenmaa, J., 2016. Current Browning of Surface Waters Will Be Further Promoted by Wetter Climate. Environ. Sci. Technol. Lett. 3 (12), 430–435.

deVicente, I., Huang, P., Andersen, F.O., Jensen, H.S., 2008. Phosphate adsorbtion by fresh and aged aluminum hydroxide. Environ. Sci. Technol. 42 (17), 6650–6655.

Egemose, S., de Vicente, I., Reitzel, K., Flindt, M.R., Andersen, F.O., Lauridsen, T.L., Sondergaard, M., Jeppesen, E., Jensen, H.S., 2011. Changed cycling of P, N, Si, and DOC in Danish Lake Nordborg after aluminum treatment. Canadian J. Fisheries and Aquatic Sci. 68 (5), 842–856.

Egemose, S., Reitzel, K., Andersen, F.Ø., Jensen, H.S., 2012. Resuspension-mediated aluminium and phosphorus distribution in lake sediments after aluminium treatment. Hydrobiologia 701 (1), 79–88.

Garrison, P.J., Knauer, D.R., 1984. Long ter evaluation of three alum treated lakes. Lake Reserv. Manag. 1 (1), 513–517.

Huser, B., Brezonik, P., Newman, R., 2011. Effects of alum treatment on water quality and sediment in the Minneapolis Chain of Lakes, Minnesota, USA.. Lake Reserv. Manag. 27 (3), 220–228.

Huser, B.J., 2012. Variability in phosphorus binding by aluminum in alum treated lakes explained by lake morphology and aluminum dose. Water Res. 46 (15), 4697–4704.

Huser, B.J., 2017. Aluminum application to restore water quality in eutrophic lakes: maximizing binding efficiency between aluminum and phosphorus. Lake Reserv. Manag. 33 (2), 143–151.

Huser, B.J., Bajer, P.G., Chizinski, C.J., Sorensen, P.W., 2016a. Effects of common carp (Cyprinus carpio) on sediment mixing depth and mobile phosphorus mass in the active sediment layer of a shallow lake. Hydrobiologia 763, 23–33.

Huser, B.J., Egemose, S., Harper, H., Hupfer, M., Jensen, H., Pilgrim, K.M., Reitzel, K., Rydin, E., Futter, M., 2016b. Longevity and effectiveness of aluminum addition to reduce sediment phosphorus release and restore lake water quality. Water Res. 97, 122–132.

Huser, B.J., Futter, M., Lee, J.T., Perniel, M., 2016c. In-lake measures for phosphorus control: the most feasible and cost-effective solution for long-term management of water quality in urban lakes. Water Res. 97, 142–152.

Huser, B.J., Fölster, J., Köhler, S.J., 2012. Lead, zinc, and chromium concentrations in acidic headwater streams in Sweden explained by chemical, climatic, and land-use variations. Biogeosciences 9 (11), 4323–4335.

James, W.F., Barko, J.W., Taylor, W.D., 1991. Effects of Alum Treatment on Phosphorus Dynamics in a North-Temperate Reservoir. Hydrobiologia 215 (3), 231–241.

Jensen, H.S., Reitzel, K., Egemose, S., 2015. Evaluation of aluminum treatment efficiency on water quality and internal phosphorus cycling in six Danish lakes. Hydrobiologia 751 (1), 189–199.

Kennedy, R.H., Barko, J.W., James, W.F., Taylor, W.D., Godshalk, G.L., 1987. Aluminum Sulfate Treatment of a Eutrophic Reservoir: rationale, Application Methods, and Preliminary Results. Lake Reserv. Manag. 3 (1), 85–90.

Kuster, A.C., Kuster, A.T., Huser, B.J., 2020. A comparison of aluminum dosing methods for reducing sediment phosphorus release in lakes. J. Environ. Manage. 261, 110195.

Lewandowski, J., Schauser, I., Hupfer, M., 2003. Long term effects of phosphorus precipitations with alum in hypereutrophic Lake Süsser See (Germany). Water Res. 37 (13), 3194–3204.

Mehner, T., Diekmann, M., Gonsiorczyk, T., Kasprzak, P., Koschel, R., Krienitz, L., Rumpf, M., Schulz, M, Wauer, G., 2008. Rapid Recovery from Eutrophication of a Stratified Lake by Disruption of Internal Nutrient Load. Ecosystems 11 (7), 1142–1156.

Monteith, D.T., Stoddard, J.L., Evans, C.D., de Wit, H.A., Forsius, M., Hogasen, T., Wilander, A., Skjelkvale, B.L., Jeffries, D.S., Vuorenmaa, J., Keller, B., Kopacek, J., Vesely, J., 2007. Dissolved organic carbon trends resulting from changes in atmospheric deposition chemistry. Nature 450 (7169), 537–U539.

Nürnberg, G.K., 2009. Assessing internal phosphorus load – Problems to be solved. Lake Reserv. Manag. 25 (4), 419–432.

Osgood, R.A., 1988. Lake mixis and internal phosphorus dynamics. Arch. Hydrobiol. 113 (4), 629–638.

Parkos, J.J., Santucci, V.J., Wahl, D.H., 2003. Effects of adult common carp (Cyprinus carpio) on multiple trophic levels in shallow mesocosms. Canadian J. Fisheries and Aquatic Sci. 60 (2), 182–192.

Pilgrim, K.M., Huser, B.J., Brezonik, P.L., 2007. A method for comparative evaluation of whole-lake and inflow alum treatment. Water Res. 41 (6), 1215–1224.

Reitzel, K., Hansen, J., Andersen, F.O., Hansen, K.S., Jensen, H.S., 2005. Lake restoration by dosing aluminum relative to mobile phosphorus in the sediment. Environ. Sci. Technol. 39 (11), 4134–4140.

Rydin, E., 2000. Potentially mobile phosphorus in Lake Erken sediment. Water Res. 34 (7), 2037–2042.

Rydin, E., Huser, B., Welch, E.B., 2000. Amount of phosphorus inactivated by alum treatments in Washington lakes. Limnol. Oceanogr. 45 (1), 226–230.

Rydin, E., Welch, E.B., 1999. Dosing alum to wisconsin lake sediments based onin vitroformation of aluminum bound phosphate. Lake Reserv Manag 15 (4), 324–331.

Sas, H., 1990. Lake restoration by reduction of nutrient loading: expectations, experiences, extrapolations. Int. Assoc. Theor. Appl. Limnol. Proc. 24, 247–251.

Schütz, J., Rydin, E., Huser, B.J., 2017. A newly developed injection method for aluminum treatment in eutrophic lakes: effects on water quality and phosphorus binding efficiency. Lake Reserv. Manag. 33 (2), 152–162.

Sondergaard, M., Jensen, J.P., Jeppesen, E., 2003. Role of sediment and internal loading of phosphorus in shallow lakes. Hydrobiologia 506 (1–3), 135–145.

Weber, M.J., Brown, M.L., 2009. Effects of common carp on aquatic ecosystems 80 years after “carp as a dominant”: ecological insights for fisheries management. Rev. Fisheries Sci. 17 (4), 524–537.

Welch, E.B., Cooke, G.D., 1999. Effectiveness and Longevity of Phosphorus Inactivation with Alum. Lake Reserv. Manag. 15 (1), 5–27.

Welch, E.B., Kelly, T.S., 1990. Internal Phosphorus Loading and Macrophytes: an Alternative Hypothesis. Lake Reserv. Manag. 6 (1), 43–48.