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1 **Suppression of bloom-forming colonial cyanobacteria by**
2 **phosphate precipitation: a 30 years case study in Lake Barleber**
3 **(Germany)**

4

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21

22 Abstract

23 Although the treatment of eutrophied lakes with aluminium (Al) compounds has been
24 established for more than 40 years, publications reporting on long-term consequences
25 for phytoplankton are rare. Here we present observations from Lake Barleber for the
26 period 1985-2016. The lake was treated in autumn 1986 because of high phosphorus
27 (P) concentrations and cyanobacteria blooms, which limited the lake's recreational
28 use. Within six weeks 480 t of Al sulphate solution (37 t of Al) were applied to the lake.
29 This was equivalent to a dose of $36 \text{ g Al}^{3+} \text{ m}^{-2}$ or $5.7 \text{ mg Al}^{3+} \text{ L}^{-1}$. Already after having
30 applied half of the Al sulphate, the concentration of soluble reactive phosphorus (SRP)
31 reached its analytical limit of quantification ($3 \mu\text{g L}^{-1}$). Removal rates calculated after
32 completion of the treatment were 98% for SRP and 90% for total phosphorus (TP). In
33 the following 13 years from 1987 to 1999, cyanobacteria were almost absent. In the
34 years 2000 to 2003 as well as in 2005 and 2014 they appeared in low abundances. In
35 the period 1987-2014, almost complete absence of cyanobacteria and high
36 transparency provided good conditions for recreational use of Lake Barleber.
37 Compared to pre-treatment conditions, phytoplankton biomass increased temporarily
38 from 1987-2016. This increase in biomass did not interfere with the use for bathing
39 and swimming, because phytoplankton community composition changed towards a
40 dominance of chlorophytes and dinophytes. In 2016, however, P concentration and
41 cyanobacterial biomass rose again to the level of the last pre-treatment years (TP 134
42 $\mu\text{g L}^{-1}$, cyanobacterial biomass 1 mg L^{-1} ; averages for the period May-October). We
43 conclude that Al treatment is effective and can last for decades. For recreational lake

44 use, the effects of the alum treatment on phytoplankton community composition
45 showed to be more important than its effects on total phytoplankton biomass.

46

47 Dedication statement

48 This paper is dedicated to Prof. Dr. Helmut Klapper (June 2, 1932 to June 19, 2019).

49 Until his retirement in 1997, he was the leader of the team that designed the
50 treatment of Lake Barleber in 1986 and that did the accompanying monitoring.

51

52 Key words: eutrophication, diazotrophic cyanobacteria, phytoplankton composition,
53 lake management, aluminium application, cyanoHAB

54

55

56

57 1 Introduction

58 Growing incidences of cyanobacterial blooms in lakes worldwide place a serious
59 burden on water resources (Pearl and Scott, 2010; Ibelings et al., 2016a).
60 Cyanobacterial blooms cause a range of ecological, economic and health effects
61 (Huisman et al., 2005; Sharma et al., 2013), which can lead to serious problems,
62 especially in water bodies used primarily for drinking water supply or recreational
63 purposes. The negative side effects of such blooms can include the formation of algal
64 mats, taste and odour compounds as well as the production of cyanotoxins, which can
65 have far-reaching effects on aquatic and terrestrial organisms (Chorus and Bartram,
66 1999; Backer et al., 2015). Moreover, if water bodies are used primarily for bathing,
67 direct exposure to algal toxins poses a health risk to the public (Chorus et al., 1999).
68 Anthropogenic eutrophication is considered to be the main reason for the increase in
69 cyanobacterial blooms (Carpenter, 2008; Watson et al., 2008).

70 The addition of aluminium (Al), which permanently binds P and removes it by
71 sedimentation, is recognized as one of the most effective strategies for controlling
72 internal P loads (Cooke et al. 1982, 2005; Klapper, 1991; Wagner 2017). In P-rich lakes,
73 the addition of Al can reduce the internal P loading and thereby limit primary
74 production and suppress toxic cyanobacterial blooms (Cooke et al. 2005). The addition
75 of Al sulphate ($\text{Al}_2(\text{SO}_4)_3$; referred to as alum) to the water body rapidly leads to
76 hydrolysis and the formation of amorphous flocs of $\text{Al}(\text{OH})_3$, which exhibit a high
77 adsorption affinity for P (Huang et al., 2002). They react with PO_4^{3-} to form insoluble
78 AlPO_4 (Nogaro et al., 2013), whereby a high binding capacity for inorganic P is reached

79 at a pH of 6 to 8. In addition to reducing the P concentration, the application of Al
80 sulphate also reduces cyanobacterial blooms by flocculation (Cooke et al., 2005;
81 Brattebo et al., 2017).

82 Since the early 1970s, hundreds of lakes have been treated with Al (Landner, 1970;
83 Cooke et al. 2005; Jensen et al., 2015; Huser et al., 2016). Most of those treatments
84 have been carried out in the USA. In addition to P precipitation in the pelagic zone, the
85 formed Al hydroxide can also increase P-retention in the sediment once the flocs have
86 settled to the sediment (Lewandowski et al., 2003; Huser et al., 2011). According to
87 Cook et al. (2005) an increase of the P retention capacity of the sediment even is the
88 main goal of P inactivation when using alum. Unless overcritical external inputs or
89 burial of new sediments interfere with the sustained adsorption of P at the sediment-
90 water interface, alum treatment thereby can lead to a long-lasting improvement of
91 water quality (Lewandowski et al., 2003; Huser et al., 2016). Recently, Huser et al. (2016)
92 reviewed cases from 114 lakes of different morphology to determine key parameters for a
93 long-term success of alum treatment. They analysed treatment results with respect to the
94 long-term effects on total P concentration (TP), water clarity (Secchi depth) and
95 phytoplankton biomass (chlorophyll *a*). Decisive criteria for long-term success were:
96 the applied Al dose, the watershed-to-lake area ratio, and the morphology of the lakes.
97 The duration of success varied widely, ranging from a few months to more than 30
98 years (Welch and Cooke, 1999; Huser et al., 2016).

99 Lake Barleber (Germany) was treated with alum (autumn 1986; Rönicke and Bahr,
100 1989; Klapper and Geller, 2001) in order to re-establish conditions allowing for safe

101 recreational use after years of increasing dominance of N₂-fixing cyanobacteria
102 (*Anabaena*, *Aphanizomenon*; Fig. 1). The reduction of the P concentration lasted until
103 2014 and for almost 30 years the occurrence of cyanobacteria could be limited to a
104 level acceptable for recreational use. Although reviews, individual reports and even
105 three special issues of journals dealing with alum treatment of lakes have been
106 published in recent years (Zamparas and Zacharias, 2014; Lürling et al., 2016; Huser et
107 al., 2016; Ibelings et al., 2016a,b; Wagner, 2017), long-term studies including the
108 response of the plankton have rarely been reported. By presenting results from the
109 long-term monitoring (1985-2016) of Lake Barleber (Germany), we intend to
110 contribute to fill this gap. We further discuss the criteria for evaluating the success of P
111 precipitation with particular emphasis on the role of phytoplankton community
112 composition.

113

114 2 Methods

115 2.1 Study Site

116 Lake Barleber is located in the north of the city of Magdeburg, Germany (Table 1). It is
117 a gravel pit lake, created mainly between 1929 and 1931 (Bauch, 1953). After
118 excavation, it filled rapidly with groundwater. The lake does not have any surface
119 inflow nor outflow, but groundwater is flowing through the lake (ca. 640,000 m³a⁻¹
120 inflow, ca. 530,000 m³a⁻¹ outflow; Hannappel and Strom, 2020). The surrounding area
121 is used for agriculture, i.e., mainly the cultivation of grains and root vegetables. The

122 most important morphometric parameters are presented in Table 1. Fig. S1 (see
123 Supplementary Material) provides a bathymetric map of Lake Barleber.

124 Around Magdeburg, Lake Barleber is the most heavily used lake for recreation. It is
125 fully developed for tourism and mainly used for bathing, swimming, diving and angling.
126 Since the 1970s, a large camping site, an anglers' allotment and a main beach for
127 bathing have been established.

128 Lake Barleber is monomictic, with stable thermal stratification during summer (Fig. S2
129 in Supplementary Material). Only exceptionally, in case of complete ice cover, the lake
130 is stratified inversely in winter. The shallow depth of Lake Barleber results in a very
131 small hypolimnion, with a waterbody volume of only 117,000 m³ below 7 m depth, i.e.
132 1.5% of the total volume.

133 Soluble reactive phosphorus (SRP) has been measured in Lake Barleber since the 1950s
134 (Fig. 1; Bauch 1953). Until 1962, SRP concentrations remained below the analytical
135 detection limit of 3 µg L⁻¹. The lake was in clear-water stage with extensive underwater
136 grasslands (*Chara* and *Elodea* populations; Bauch 1953). From the mid-1960s to the
137 mid-1970s, SRP concentrations steadily increased up to 50 µg L⁻¹. This increase in SRP
138 concentrations was linked to the onset of turbidity as a result of the increased
139 presence of planktonic algae (greens and diatoms). In the following decade (1975 to
140 1985), a rapid increase in the SRP concentration was observed which, in the mid-80s,
141 led to a concentration above 150 µg L⁻¹. In the same period, planktonic cyanobacteria
142 appeared during the summer months, culminating in cyanobacteria mass
143 developments and a high turbidity of the water body (Rönicke and Bahr, 1989).

144 To reduce the internal P loading sufficiently enough to inhibit summer cyanobacterial
145 blooms, alum was applied to Lake Barleber in autumn 1986 (for details see Table 2).
146 Within a period of six weeks, the alum treatment was accomplished by surface
147 spraying (Rönicke and Bahr, 1989; Klapper and Geller, 2001). Since there were no
148 surface inflows and no known inflows of waste water, a reduction of the external
149 phosphorus load was not possible.

150

151 2.2 Sampling and analytical methods

152 Sampling of Lake Barleber was done at the deepest point of the lake at intervals of 2-3
153 weeks during the vegetation period (May to October) from 1986 to 2000. Since 2001,
154 sampling was done at monthly intervals. Until 1992, water samples were taken with a
155 Ruttner sampler; from 1993 onwards, a Limnos sampler (LIMNOS, Finland) was used.
156 In general, water samples were taken at depths of 0, 1, 2.5, 5 and 7.5 m and the
157 analytical data were averaged over the water column. To examine the water-sediment
158 contact zone (9 m), a specially developed sediment corer (Rönicke and Bahr 1990) that
159 enabled the collection of water samples right above the sediment was used. The chemical
160 parameters Al, sulphate, soluble reactive phosphorus (SRP), total phosphorus (TP) and
161 dissolved inorganic nitrogen (DIN; calculated by summing up of nitrogen analysed as
162 nitrate, nitrite and ammonia) were analysed as described in Herzsprung et al. (2005,
163 2006). We calculated DIN:TP ratios according to Dolman et al. (2016), who identified
164 this ratio as most informative with respect to nutrient limitation of primary
165 production. They defined a critical value of 1.6, with a DIN:TP ratio of < 1.6

166 corresponding to N limitation and a DIN:TP ratio of > 1.6 to P limitation. Chlorophyll-*a*
167 concentration was analysed photometrically according to the German standard DIN
168 39412. Qualitative and quantitative determination of the phytoplankton and ciliate
169 abundance was done by microscopic analysis following Utermöhl (1958), after fixation
170 with Lugol's solution. Cell sizes were measured and their biovolume calculated based
171 on suitable geometric shapes. The biomass of the individual species was determined
172 under the assumption of a specific density of 1 g cm⁻³ (Premazzi, 1980; Benndorf et al.,
173 1983). Zooplankton samples were taken by vertical hauls from 7.5 m depth to the
174 surface using a conical plankton net (opening diameter 25 cm, length 0.5 m, mesh size
175 55 µm, corrected for filtering efficiency). These samples were also fixed with Lugol's
176 solution and species were identified and quantified under the microscope. Rotifer
177 volume was calculated according to Ruttner-Kolisko (1977). Crustacean biomass was
178 calculated according to published length-weight relationships assuming that dry
179 weight equals 15% of wet weight.

180 Macrophytes were investigated four times during the study period (July 23, 2007;
181 August 26, 2010; June 29, 2014; July 13, 2016) following the requirements of the EU
182 Water Framework Directive (WFD, Guideline 2000/60/EG) by contractors of
183 Landesbetrieb für Hochwasserschutz und Wasserwirtschaft Sachsen-Anhalt. A semi-
184 quantitative methodology was applied following the German assessment standard that
185 is based on Schaumburg et al. (2004) and Stelzer et al. (2005).

186 First, a survey of the occurrence of macrophytes, macrophyte species and their
187 estimated abundance was done (according to a five degree scale: 1 - very rare; 2 - rare;

188 3 - common; 4 - frequent; 5 - abundant, predominant). Occurrence was mapped along
189 transects (four in 2007 and 5 during the other years) and separated by depth zones 0-1
190 m, 1-2 m, 2-4 m and >4 m according to Melzer (1999). Abundances were converted
191 into plant quantities using the function $y = x^3$ (Melzer 1999). From these, the reference
192 index and the class of ecological potential were determined following Schaumburg et
193 al. (2004) (the five status classes are: 1 - very good, 2 - good; 3 - moderate; 4 - poor
194 and 5 – bad).

195 In order to visualize changes in the abundance of the dominating species (abundance
196 degrees 5-3), the calculated plant quantities of all transects were averaged per depth
197 zone and sampling date.

198

199 3 Results

200 3.1 Effects of alum addition on P, Al, and sulphate concentrations

201 At the start of the alum treatment at 3rd of October 1986, the SRP concentration in the
202 pelagic zone was $180 \mu\text{g L}^{-1}$ and the TP concentration was $190 \mu\text{g L}^{-1}$ indicating that
203 95% of the P was present in soluble, mineralized form. By October 22nd after
204 approximately half of the total precipitant quantity had been applied, the detection
205 limit for SRP ($3 \mu\text{g L}^{-1}$) was reached in the pelagic zone (Fig. 2). The remaining 50% of
206 the Al sulphate solution was applied as excess precipitant to condition the sediment
207 with $\text{Al}(\text{OH})_3$ flocs in order to enhance the P binding capacity. After having completed
208 the precipitation, 98% of SRP had been removed and TP had been reduced by over

209 90%. The mean SRP concentrations in the subsequent years until 2014 fluctuated
210 between $6 \mu\text{g L}^{-1}$ (1989) and $15 \mu\text{g L}^{-1}$ (1995), and the mean TP concentration ranged
211 between $24 \mu\text{g L}^{-1}$ (1997) and $54 \mu\text{g L}^{-1}$ (1998) (Fig. 3). In 2016, however, a substantial
212 increase in P was observed (SRP $86 \mu\text{g L}^{-1}$, TP $134 \mu\text{g L}^{-1}$). Concentrations of Al and
213 sulphate increased during the treatment, but reached similar levels to the ones observed
214 before the treatment by the end of the year 1986 (Table 3).

215

216 3.2 Phosphorus accumulation in the bottom layer

217 In the summer of 1986, before the alum was applied, a massive release of dissolved P
218 from the lake sediment was detected (Fig. 4). Already in April, a rapid decline in
219 oxygen concentrations occurred near the bottom of the lake, and led to anaerobic
220 conditions during the summer months. This oxygen decline was accompanied by a
221 marked increase in SRP: in mid-April (16 Apr) 1986 the SRP concentration was $49 \mu\text{g L}^{-1}$
222 and at the end of September (30 Sept) a concentration of almost 1 mg L^{-1} ($980 \mu\text{g L}^{-1}$)
223 was recorded. These SRP concentrations at the lake bottom were several times higher
224 than those measured in the surface water (0.18 mg L^{-1}) indicating a substantial release
225 of P from the phosphate-rich sediment during the summer stratification period.

226 In the years after the alum treatment, even during periods of low oxygen levels, the
227 release of P was almost entirely inhibited and SRP concentration remained low (Fig. 4).
228 From 1987 to 2014, no or only very low P release rates from the sediments were
229 recorded (Fig. 4). In late September 2016, however, 1.15 mg L^{-1} SRP was measured
230 above the sediment indicating significant P release from the sediment.

231

232 3.3 N:P ratios

233 During the summer period of 1986 (June-August), before the phosphate precipitation
234 took place, the mean mass-related DIN:TP ratios remained below the critical value of
235 1.6, indicating N-limitation of the phytoplankton (Fig. 5). After the application of alum,
236 DIN:TP ratios increased as a result of the reduction in the pelagic P concentration. In
237 the following years 1987 to 2014, DIN:TP ratios were mostly above the critical value of
238 1.6, indicating P-limitation. In 1987, the value even rose to 5.3. In summer 2016, the
239 DIN:TP ratio again dropped below the critical value of 1.6.

240

241 3.4 Secchi disc transparency and chlorophyll-*a* concentration

242 Bi-weekly to monthly Secchi depths measurements were averaged over the six-month
243 vegetation period. The mean Secchi depths in 1985 and 1986 prior to application of
244 the precipitant, were 1.3 m and 2 m, respectively (Fig. 6). During the summer months
245 and the mass development of diazotrophic cyanobacteria, the values dropped to well
246 below 1 m. In the years after the application of the precipitant, Secchi depths
247 increased. The highest mean values were calculated for the years 1998, 1999, 2002
248 and 2013 at approximately 6 m. Even individual values ≥ 8 m were observed: 8.6 m (15
249 June 1994), 8 m (19 June 2000), 8.5 m (20 June 2013) and 9.0 m (9 June 2016). In
250 addition to the general increase in Secchi depths, also the duration of periods of high
251 transparency (> 5 m) increased. It continually lasted over several weeks in the post-

252 treatment years. In 1994, for instance, the period of high Secchi depths lasted over
253 two months from 15 June until 24 August 1994. In August 2016, however, Secchi
254 depths decreased to 0.5 m again, caused by an algal mass development.

255 In the six-month vegetation period of the years 1985 to 2016, mean chlorophyll-*a*
256 concentrations overall remained at a low level (Fig. S1). Significant fluctuations
257 occurred during the study period as a whole. The lowest recorded chlorophyll-*a*
258 concentration was 2.0 µg L⁻¹ in 1998. The highest annual average (10.1 µg L⁻¹) was
259 measured in 2001. The highest single concentration peak during the years 2000 and
260 2001 corresponded with the mass occurrence of *Ceratium hirundinella* (dinophytes)
261 (see below).

262

263 3.5 Development of phytoplankton

264 In the years 1985 and 1986, the summer phytoplankton community was largely
265 characterized by diazotrophic cyanobacteria (Fig. 6, Fig. S2). The dominant species
266 were *Anabaena flos-aquae*, *A. lemmermannii* and *A. circinalis*. The genus
267 *Aphanizomenon* was represented by the species *Aphanizomenon flos-aquae* and *A.*
268 *gracile*. The average percentage contribution of cyanobacteria to the total
269 phytoplankton biomass was 45% in 1985 and 58% in 1986. In the summer months June
270 to August, cyanobacteria dominated the phytoplankton community with maximum
271 percentages of 96% (11 July 1985, mean 85%) and 98% (09 July 1986, mean 88%).
272 These mass developments led to the formation of extensive floating algal mats. In the
273 subsequent 13 years after P precipitation (autumn 1986), N₂-fixing cyanobacteria were

274 observed rarely. The low abundances of cyanobacteria detected in autumn 1987 and
275 spring 1988 were composed by the species *Limnothrix redekei* and *Microcystis*
276 *aeruginosa*. Diazotrophic cyanobacteria (*Anabaena* and *Aphanizomenon* species) were
277 first detected again in the years 2000, 2002, 2003 and 2005. However, their
278 percentage contribution to the total phytoplankton was significantly lower than
279 measured in 1985 and 1986. During the six-month vegetation period they accounted for
280 6% in 2000, 13% in 2002, 15% in 2003 and 3% in 2005. Besides cyanobacteria, over the
281 entire study period dominant representatives of chrysophytes (*Kephyrion spec.*,
282 *Ochromonas spec.*, *Chrysochromulina parva*, *Dinobryon divergens*), greens
283 (*Chlamydomonas spec.*, *Ankistrodesmus angustus*, *A. falcatus*, *Scenedesmus*
284 *longispina*, *S. quadricauda*, *Kirchneriella obesa*, *Crucigenia rectangularis*), diatoms
285 (*Stephanodiscus astraea*, *S. hantzschii*, *Nitzschia acicularis*, *Tabellaria fenestrata*,
286 *Diatoma elongatum*, *D. vulgare*, *Fragilaria crotonensis*, *Synedra acus*), dinophytes
287 (*Ceratium hirundinella*, *Gymnodinium spec.*, *Peridinium umbonatum*) and cryptophytes
288 (*Cryptomonas erosa*, *C. ovata*, *Rhodomonas pusillum*) were detected. During the years
289 2000 to 2005, dinophytes were increasingly present. *Ceratium hirundinella*, a species
290 with very high cell volume, dominated the phytoplankton community. Its mean
291 percentage share during the six-month vegetation period ranged between 66% and
292 99%. During the *Ceratium*-dominated years, the total phytoplankton biovolume was
293 high compared to the years before or afterwards (Fig. 6). Starting in 2014, the
294 contribution of cyanobacteria to the total phytoplankton biomass increased again
295 (2014: 6%, 2016: 20%; Fig. 6), parallel to the increase of P concentrations (Fig. 3).
296 However, on average dinophytes still dominated during the years 2014 and 2016.

297

298 3.6 Dynamics of zooplankton

299 Zooplankton total biomass was $1.3 \text{ mm}^3 \text{ L}^{-1}$ in spring and summer 1986, but decreased
300 to $0.5 \text{ mm}^3 \text{ L}^{-1}$ during the alum treatment (Fig. 7). During treatment, cladocerans
301 disappeared and both rotifers and ciliates declined in number. Copepods were less
302 affected. From 1987 onwards, cladocerans re-established and contributed significantly
303 to the algal biomass (*Daphnia hyalina*, *D. galeata*, *Diaphanosoma brachyurum*,
304 *Bosmina longirostris*). With the exception of 1990, copepods (*Eudiaptomus gracilis*,
305 *Cyclops spp.*) remained dominant in terms of biovolume. The average annual
306 zooplankton biomass was somewhat lower ($0.6 - 1.2 \text{ mm}^3 \text{ L}^{-1}$) in the years after the
307 treatment compared to 1986 (pre-treatment; Fig. 7).

308

309 3.7 Macrophytes

310 The macrophyte status of Lake Barleber evaluated according to the WFD was
311 moderate (2007), good (2010 and 2014) and moderate (2016) (Tab. 4).

312 The species reaching the abundance degree of 5 in at least one transect and at least
313 once were *Phragmites australis*, *Typha angustifolia*, *Potamogeton pectinatus*,
314 *Ranunculus circinatus*, *Elodea canadensis*, and *Chara contraria*. The abundance degree
315 of 4 was reached in at least one transect and at least once by *Fontinalis antipyretica*,
316 *Potamogeton lucens*, *Potamogeton perfoliatus*, *Ceratophyllum demersum*, *Lemna*
317 *trisulca*, *Chara globularis* and *Nitellopsis obtuse*. Detailed results of the macrophyte

318 assessment are provided as supplementary material
319 (Roenicke_etal_Macrophytes_LakeBarleber_2007-2016.xls). There was no consistent
320 trend in the lake-wide averages of plant quantities separated by depth zones (Fig. S5).
321 At all investigation dates, the entire lake bottom was also colonised by the filamentous
322 green alga *Vaucheria spec.* down to the deepest parts of the lake basin.

323

324 4 Discussion

325 4.1 Suitability of aluminium sulphate treatment for Lake Barleber

326 From the available approaches for suppressing cyanobacterial blooms alum treatment
327 was chosen for Lake Barleber. Under the conditions in Magdeburg (former GDR, i.e.
328 East Germany) in 1986, alum treatment was seen as the most effective and efficient
329 approach with the highest longevity. However, the use of alum is often seen critically
330 and subject of discussion in the public due to the well-known toxicity of Al. Al and low
331 pH values can have direct toxic effects on plankton (Havens, 1990, Wauer et al., 2004),
332 as demonstrated for *Daphnia* at Al concentrations of 1 mg L⁻¹ (Havas and Likens, 1985).
333 Also indirect negative effects on zooplankton can occur. The non-selective filter
334 feeding daphnids are particularly vulnerable to decreases in food quality such as
335 increasing amounts of abiotic seston (e.g. Al flocs) (Kirk, 1992). Smaller zooplankton
336 organisms (ciliates, rotifers) may also be removed from the water column directly by
337 flocculation. In Lake Barleber, only a slight and short increase in Al concentrations was
338 observed (Table 3). The the sufficiently high alkalinity in Lake Barleber (≥ 2 meq L⁻¹)
339 kept pH at a natural level where Al solubility is very low. The occurrence of a diverse

340 and abundant zooplankton community in the years following the treatment implies
341 that there was no sustainable toxic effect. Also no damage of fish was reported by
342 anglers.

343 High concentrations of sulphate can promote the formation of iron sulphide under
344 anoxic conditions and, thus, reduce the P binding capacity. In Lake Barleber, the alum
345 treatment did not change the sulphate concentration considerably (Table 3). Thus,
346 selection of Al sulphate for phosphate precipitation in Lake Barleber was adequate.
347 Due to the improvement of Secchi depths and the suppression of cyanobacteria mass
348 developments of cyanobacteria allowing for undisturbed recreational use for almost
349 30 years, the treatment of Lake Barleber was a success.

350

351 4.2 Comparison of the treatment results with other lakes

352 In the 114 case studies examined by Huser et al. (2016) for the long-term impact and
353 effectiveness of alum treatment, the applied quantities of alum ranged between 5 and
354 122 g Al m⁻². In six Danish lakes treated with polyaluminium chloride during the years
355 2001 to 2009, the applied Al doses ranged between 10 and 54 g Al m⁻² (Jensen et al.,
356 2015). Thus, the Al dose of 36 g Al m⁻² applied to Lake Barleber is in the middle range
357 of the published data.

358 The reduction of the phosphate concentration reached immediately after the
359 treatment was very high (98% for SRP, 90% for TP). Although the average reductions
360 for the years 1987-2014 were only 89% for SRP and 68% for TP, this was still enough to

361 cause considerable changes in the phytoplankton composition (Fig. 7) and was in the
362 range reached in other lakes (Huser et al., 2016). The reduction lasted at least until
363 2014, i.e. 28 years. This is much longer than Huser et al. (2016) found as average for
364 unstratified (6 years) and stratified lakes (21 years). Obviously, in Lake Barleber, the
365 intended surplus of binding capacity of the sediment, which resulted from the second
366 half of the applied alum amount worked well until it was exhausted in 2015 or 2016.
367 Nevertheless, our results indicate that the water quality improvements after the alum
368 treatment were limited in time – a statement that was also clearly made by Huser et
369 al. (2016).

370 The absence of surface inflows and the small catchment area of Lake Barleber
371 probably has not provided enough sediment to bury the Al hydroxide rapidly as
372 reported for Lake Süßer See (Lewandowski et al., 2003). Bioturbation during times of
373 sufficient supply of oxygen at the sediment surface (Fig. 4) may have contributed to
374 the long lasting reduction of P concentrations in Lake Barleber. Firstly, the consequent
375 mixing of the sediment can have prevented a too deep burial of the Al hydroxide as
376 observed in Lake Süßer See (Lewandowski et al., 2003). Secondly, bioturbation can
377 allow for deeper oxidation of the sediment surface (Lewandowski and Hupfer, 2005).
378 Future sediment investigations and a detailed P balance are needed to reliably identify
379 the reasons for the end of the success phase of the alum treatment in Lake Barleber.

380

381 4.3 Evaluation of changes in phytoplankton

382 Comparing chlorophyll-*a* concentrations and phytoplankton biomass before and after
383 the treatment may not lead to the conclusion that the measure was a success. The
384 biomass and also the concentration of chlorophyll-*a* remained in the same range
385 before and after the treatment, in particular when considering the interannual
386 fluctuations. However, there were remarkable changes in the community composition
387 of the phytoplankton. Green algae and increasingly dinophytes were dominant after
388 the treatment. The mass occurrence of dinophytes in the years 2000 to 2005 mainly
389 consisted of the species *Ceratium hirundinella*. *Ceratium* has the ability to migrate
390 from surface waters during the day to the metalimnion during the night in order to
391 take up P at depth (Whittington et al., 2000). This may have given the species a
392 competitive advantage under P limitation. The change in phytoplankton community
393 composition was accompanied by increases in Secchi depths and cell volumes of the
394 dominant algal species.

395 Relevant phytoplankton consumers are in the group of crustaceans rather than rotifers
396 or ciliates. For the onset of the clear-water phase, Lampert (1988) quoted a critical
397 biomass of crustaceans of 1.5 g m⁻² dry weight, which corresponds to a value of 1.3 mg
398 L⁻¹ fresh weight for our study lake. This biomass was reached or exceeded on 16
399 sampling days, i.e. in 15% of the samples. Within the group of crustaceans, large
400 *Daphnia* species are the most efficient phytoplankton grazers. Earlier studies observed
401 significant changes in the abundance of available prey at a *Daphnia* biomass of 0.7 mg
402 L⁻¹ fresh weight (Jürgens 1994, Tittel et al. 1998). This value was exceeded in 12% of
403 the sampling days. Smaller cladocerans (*Bosmina*) were included in our calculations,
404 which are less efficient phytoplankton consumers than *Daphnia* species. High

405 biomasses occurred only in summer with maximum values in July 1987 (2.1 mg L⁻¹) as
406 well as in June and August 1988 (1.9 mg L⁻¹). The well available and likely the most
407 affected prey can be expected to be in the size of up to ca. 30 µm length. These were
408 green algae, chrysophytes and partly diatoms, but not dinophytes (*Ceratium*) or
409 filamentous cyanobacteria. We conclude that in about 30% to 40% of all summer days
410 the zooplankton contributed significantly to high transparencies.

411 The phagotrophy of *Ceratium* was subjected to a debate among phycologists. Earlier
412 reports of Hofeneder (1930) and of Dodge und Crawford (1970) have provided
413 evidence of phagotrophy. Later on phagotrophy of *Ceratium* has been questioned
414 (Chapman et al., 1981, Hansen und Calado, 1999, Stoecker 1999). However, there is
415 clear evidence of phagotrophy by another species of the genera (*C. furcoides*,
416 Bockstahler and Coats 1993). Further work is needed to address this issue (Moestrup
417 and Calado 2018). If *C. hirundinella* was capable of ingesting bacteria this phagotrophy
418 might enlarge the accessible P pool for the phytoplankton and contribute to the
419 relatively high phytoplankton biovolume as was observed after the alum treatment. No
420 decline in phytoplankton biomass after P reductions also was reported for other lakes,
421 including the progressing dominance of phagotrophic algae (mixotrophs) (Jeppesen et
422 al., 2005; Weyenmeyer and Broberg, 2014; Wentzky et al., 2018). Therefore, the total
423 biovolume of phytoplankton often is a poor indicator of successful eutrophication
424 control, while community composition and transparency are usually more powerful
425 indicators.

426 The predominant cyanobacteria in Lake Barleber were *Anabaena* and *Aphanizomenon*
427 in Lake Barleber. Both genera have the potential to fix nitrogen, to form surface scums
428 and to produce toxins. Due to the formation of filaments and large colonies, both
429 genera can hinder efficient grazing by filter feeders. Diazotrophic cyanobacteria are
430 likely favoured by low nitrogen concentrations, but require high P levels for mass
431 development (Reynolds et al., 2002).

432 To understand their dominance, the N:P ratio plays an important role. In a series of
433 studies, the DIN:TP ratio in the water column was shown to be the best indicator for
434 limiting conditions (e.g. Morris and Lewis, 1988; Bergström, 2010; Dolman et al., 2016).
435 According to this ratio, the conditions in Lake Barleber changed from N-limitation to P-
436 limitation after the phosphate precipitation. This counteracts the dominance of
437 diazotrophic cyanobacteria and can be regarded as the major reason for the
438 suppression of cyanobacterial blooms after Al treatment (Brattebo et al. 2017).

439 Unfortunately, no quantitative data exist for the occurrence of macrophytes in Lake
440 Barleber before the restoration in autumn 1986 and in the years immediately after
441 the treatment. However, property owners at Lake Barleber reported that there was a
442 considerable increase of the abundance of submerged macrophytes in 1987 and the
443 following years. This is consistent with the memories of the author H. Rönicke.

444 Macrophytes are well known to stabilize clear water conditions in shallow lakes
445 (Scheffer and van Nes 2007). According to Hilt (2015) this stabilizing effect may be
446 found in deep lakes, too. Therefore, stabilizing effects can also be expected for Lake
447 Barleber. The status class “good” found for macrophytes in 2010 and 2014 suggests

448 that the macrophytes were abundant enough to contribute to the high water clarity.
449 The decline in abundance for a number of species in 2016 already may have been a
450 consequence of the re-occurrence of considerable P release from the sediment and,
451 thus, an indicator for a shift back to a turbid state.

452

453

454 5 Conclusion

455 The precipitation of P using Al sulphate, as carried out in Lake Barleber in autumn
456 1986, has proven to be an effective restoration method. The P content of the water
457 body was largely reduced and harmful cyanobacterial blooms were suppressed over a
458 period of almost thirty years. This pronounced long-term effect had been achieved by
459 applying an excess Al dose. By conditioning the P-rich sediment with $\text{Al}(\text{OH})_3$ flocks, the
460 release of P (internal fertilization) in the following years was considerably reduced.
461 Consequently, only at the end of the study period, a significant increase in P
462 concentrations was observed. In well-buffered eutrophic standing waters with little
463 through-flow and low external input of P, precipitation with Al can lead to a marked
464 long-term improvement in water quality. The missing reduction of the phytoplankton
465 biomass and even its increase above the pre-treatment level did not hinder the
466 recreational use of Lake Barleber since the transparency of the water was high and
467 toxin producing cyanobacteria did not reach relevant abundances. We conclude that
468 the success of the measure was based on a shift in the phytoplankton community
469 composition caused by the strong reduction of P availability.

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478

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658

659 Table 1 Morphometric parameters for Lake Barleber

660

Lake parameter	Value
Location	52° 13' 15'' N, 11° 39' 0'' O
Area	103 ha
Volume	6.9 x 10 ⁶ m ³
Mean depth	6.7 m
Maximum depth	10.0 m
Inflow	groundwater

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664 Table 2 Application of aluminium sulphate to the surface of Lake Barleber

Period:	3 October to 13 November 1986
Precipitating agent:	aluminium sulphate
Quantity added:	480 t of solution containing 37 t Al ³⁺
Al dose per volume:	5.7 mg Al ³⁺ L ⁻¹
Al dose per area:	36 g Al ³⁺ m ⁻²
Distribution technique:	floating vessel with tank for the aluminium sulphate solution and a perforated tube (with 10 mm holes) pushed by a motor boat

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668 Table 3 Aluminium and sulphate concentrations as well as pH values (mean values 0-
669 7.5 m) before, during and after the application of aluminium sulphate (n.d. - not
670 analysed)

Date	Al ³⁺ (mg L ⁻¹)	SO ₄ ³⁻ (mg L ⁻¹)	pH
11/09/1986	n.d.	505	8.3
29/09/1986	0.04	495	8.1
08/10/1986	0.12	510	7.8
15/10/1986	0.17	513	7.6
23/10/1986	0.25	525	7.5
29/10/1986	0.5	534	7.4
12/11/1986	0.4	651	7.6
19/11/1986	0.1	611	7.7
10/12/1986	0.05	454	7.8

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673 Table 4 Results of macrophyte investigations summarized per transect (for all depth
 674 zones) and evaluation in terms of status classes

Date	Transect	Location of transect	Lower limit of macrophytes (m)	Species number of transect	Total species in Lake Barleber	Status class of transect	Status class of Lake Barleber
23.07.2007	1	West	5.4	5	16	moderate	moderate
23.07.2007	2	East	6.5	12	16	good	moderate
23.07.2007	3	North-west	6.0	11	16	moderate	moderate
23.07.2007	4	South	6.5	8	16	good	moderate
26.08.2010	1	West	7.0	7	15	good	good
26.08.2010	2	East	7.7	7	15	moderate	good
26.08.2010	3	North-west	7.0	5	15	good	good
26.08.2010	4	South	7.0	8	15	good	good
26.08.2010	5	South-east	7.2	6	15	very good	good
29.06.2014	1	West	4.8	6	19	moderate	good
29.06.2014	2	East	6.1	8	19	good	good
29.06.2014	3	North-west	5.8	10	191	good	good
29.06.2014	4	South	4.8	9	19	good	good
29.06.2014	5	South-east	5.8	10	19	very good	good
13.07.2016	1	West	8.3	5	18	moderate	moderate
13.07.2016	2	East	8.2	13	18	good	moderate
13.07.2016	3	North-west	6.5	11	18	good	moderate
13.07.2016	4	South	6.5	8	18	good	moderate
13.07.2016	5	South-east	7.2	7	18	good	moderate

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679 Figure captions

680 Figure 1 Concentrations of phosphorus (SRP, spring overturn values, 0-7.5 m), 1950 to
681 2017 (based on Röncke et al. 1995, extended)

682 Figure 2 SRP concentration (0-7.5 m) and oxygen in the deep water layer (9 m), 1986
683 and 1987 (IC – ice cover, AP – aluminium precipitation)

684 Figure 3 Alum application and the effects on phosphorus concentration (mean values
685 0-7.5 m), 1986 to 2016

686 Figure 4 Dynamics of SRP- and oxygen concentration in the deep water layer (9 m) in
687 Lake Barleber, 1986 to 1992, 1999, 2010, 2013, 2014, and 2016

688 Figure 5 Mean values of N:P ratio (DIN:TP) in the summer months (June to August) in
689 Lake Barleber, 1986 to 2016

690 Figure 6 Mean values of phytoplankton biomass and composition (0-7.5 m) and Secchi
691 depths during the vegetation period (May to October) in Lake Barleber, 1985 to 2016

692 Figure 7 Zooplankton biovolume and community composition. Upper part: total
693 biovolume, means from 9-19 samplings per year; lower part: composition derived from
694 proportions at individual sampling days. * before precipitation (February to October 2,
695 14 samples), # during precipitation (October 15 to December, 6 samples)

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Electronic supplementary material for

Suppression of bloom-forming colonial cyanobacteria by phosphate precipitation: a 30 years case study in Lake Barleber See (Germany)

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The supplementary material consists of Figures S1-S4, provided below in this document and a MS Excel file named SupplementaryMaterial_Roenicke_etal.xlsx. The latter provides the data used in the paper.

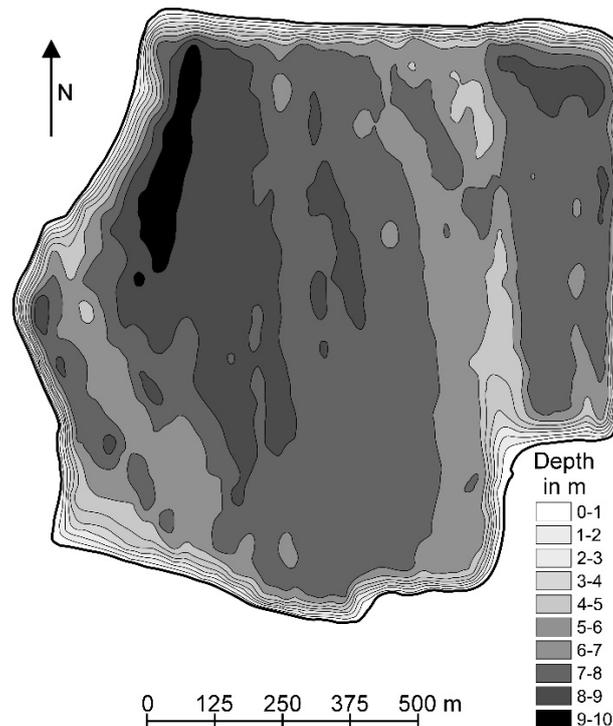


Figure S1 Bathymetric map of Lake Barleber (redrawn based on the map prepared by Fisch und Umwelt e.V., Rostock, Germany by order and for account of Landesbetrieb für

Hochwasserschutz und Wasserwirtschaft Sachsen-Anhalt, Gewässerkundlicher Landesdienst, Magdeburg, Germany in 2003).

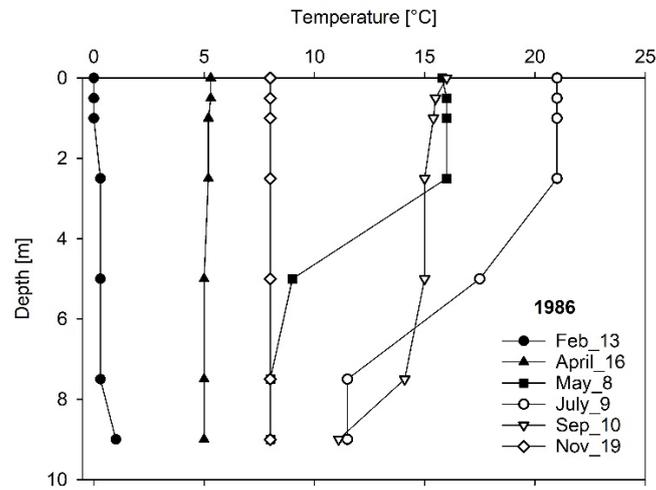


Figure S2 Results of selected temperature measurements from surface to bottom in 1986 at the deepest site of Lake Barleber.

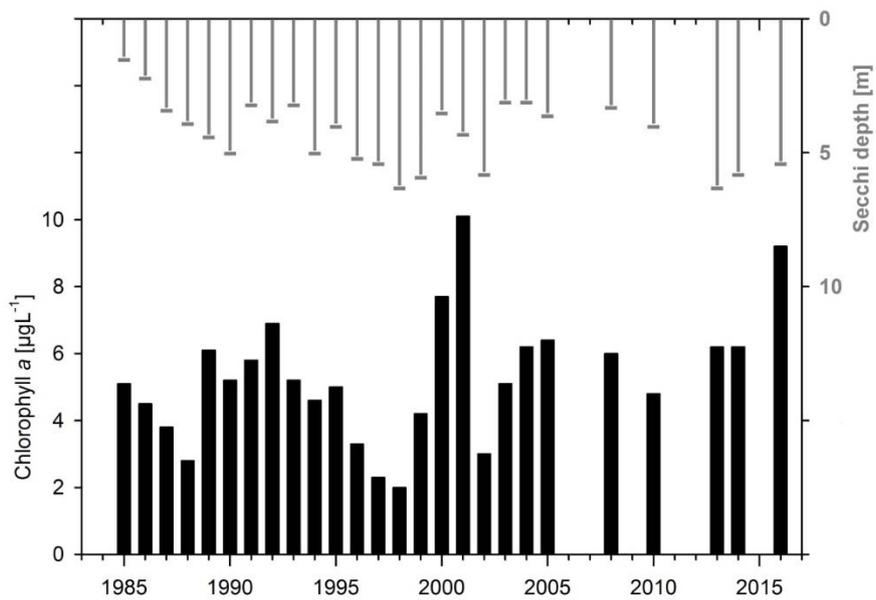


Figure S3 Mean values of chlorophyll-*a* concentration (0-7.5 m) and Secchi depths during the vegetation period (May to October) in Lake Barleber, 1985 to 2016.

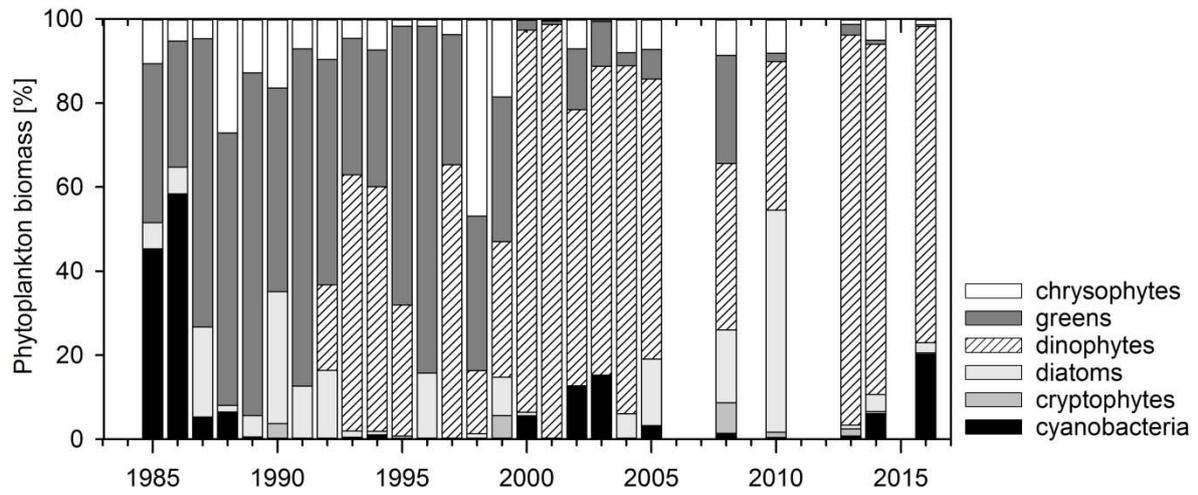


Figure S4 Percentage of phytoplankton biomass classified into the major taxonomic groups (annual mean values, 0-7.5 m) during the vegetation period (May to October) in Lake Barleber, 1985 to 2016.