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1 **Reduced acid deposition leads to new start for brown trout (*Salmo***  
2 ***trutta*) in an acidified lake in southern Norway**

3  
4 Espen Lund<sup>A</sup>, espen.lund@niva.no, corresponding author, phone +47 47400858, ORCID 0000-0003-  
5 4089-9068

6  
7 Øyvind A. Garmo<sup>A</sup>, oyvind.garmo@niva.no

8  
9 Heleen A. de Wit<sup>A</sup>, heleen.de.wit@niva.no, ORCID 0000-0001-5646-5390

10

11 Torstein Kristensen<sup>B</sup>, torstein.kristensen@nord.no, ORCID 0000-0002-2640-4260

12

13 Kate L. Hawley<sup>A,C</sup>, kate.hawley@niva.no

14

15 Richard F. Wright<sup>A</sup>, richard.wright@niva.no

16

17 <sup>A</sup> Norwegian Institute for Water Research (NIVA), Gaustadalléen 21, 0349 Oslo, Norway.

18 <sup>B</sup> Nord University, Universitetsalléen 11, 8026 Bodø, Norway.

19 <sup>C</sup> Current: Faculty of Environmental Sciences and Natural Resource Management, Norwegian

20 University of Life Sciences, 1432 Ås, Norway. kate.louise.hawley@nmbu.no

21

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23

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## Abstract

Acid deposition has led to acidification and loss of fish populations in thousands of lakes and streams in Norway. Since the peak in the late 1970s acid deposition has been greatly reduced, and acidified surface waters have shown chemical recovery. Biological recovery, in particular fish populations, however, has lagged behind. Long-term monitoring of water chemistry and fish populations in Lake Langtjern, south-eastern Norway, show that around 2008 chemical recovery had progressed to the point at which natural reproduction of brown trout (*Salmo trutta*) reoccurred. The stocked brown trout reproduced in the period 2008–2014, probably for the first time since the 1960s, but reproduction and/or early life stage survival was very low. The results indicate that chemical thresholds for reproduction in this lake are approximately  $\text{pH} = 5.1$ ,  $\text{Al}_i = 26 \mu\text{g/l}$ ,  $\text{ANC} = 47 \mu\text{eq/l}$ , and  $\text{ANC}_{\text{Oaa}} = 10 \mu\text{eq/l}$  as annual mean values. These thresholds agree largely with the few other cases of documented recovery of brown trout in sites in Norway, Sweden and the UK. Occurrence and duration of acidic episodes have decreased considerably since the 1980s, but still occur and probably limit reproduction success.

## 54 Introduction

55           During the 20<sup>th</sup> century acid deposition caused environmental damage in large regions of  
56 Europe and eastern North America. In Norway, thousands of lakes and streams were acidified, with  
57 the resultant loss and damage to freshwater fish populations (Hesthagen et al. 1999). Southern  
58 Norway is particularly vulnerable to acid deposition due to the highly siliceous and weathering-  
59 resistant bedrock and overburden, and thin and patchy organic-rich soils (Wright and Henriksen  
60 1978). Acid deposition in Europe peaked in the late 1970s and has declined sharply over the past 30  
61 years (Schöpp et al. 2003), largely as a result of implementation of international agreements to  
62 reduce the emissions of acidifying air pollutants (UNECE 2014). Acidified freshwaters in Norway have  
63 shown dramatic improvements in water chemistry as a response to declining acid deposition  
64 (Skjelkvåle et al. 1998; Garmo et al. 2014; Gray et al. 2016). In many cases, however, biological  
65 recovery has lagged behind chemical recovery (Hesthagen et al. 2011; Hesthagen et al. 2016).

66

67           Damage to salmonid fish populations, in particular the brown trout (*Salmo trutta*), in acidified lakes  
68 is usually due to recruitment failure. The eggs and young fry are the most sensitive life stages, and  
69 they are often exposed to the acidic water during snowmelt (Overrein et al. 1980; Serrano et al.  
70 2008). In marginally-acidified lakes stocking with young fish may be successful, but reproduction  
71 often fails. Toxicity is largely due to elevated concentrations of inorganic aluminium species (termed  
72 here  $Al_i$ ) (Baker and Schofield 1982; Rosseland et al. 1990). Toxicity of  $Al_i$  is ameliorated by dissolved  
73 organic carbon (DOC) in the water through formation of Al-humus complexes (Cronan et al. 1986).

74

75            $Al_i$  is mobilized from soils by acidic water. The strong acid anions sulphate ( $SO_4$ ) and nitrate ( $NO_3$ ) in  
76 acid deposition acidify the soil and mobilize  $Al_i$  (Reuss et al. 1990). Acid neutralising capacity (ANC),  
77 defined as the equivalent sum of the concentrations of base cations minus the equivalent sum of the  
78 concentrations of strong acid anions, is commonly used as a measure of the acidification of  
79 freshwaters (Reuss et al. 1987). There was a close relationship between ANC and the brown trout

80 population status in lakes in Norway during the 1980s when acidification was near its peak (Lien et  
81 al. 1996). Inclusion of organic strong acids ( $ANC_{Oaa}$ ; “organic acid adjusted”) slightly improved the  
82 correlation (Lydersen et al. 2004).

83

84 Documentation of chemical and biological recovery in acidified lakes requires systematic long-term  
85 monitoring. In southern Norway, Lake Langtjern is one such monitoring site where water chemistry  
86 and fish populations have been monitored since the 1970s (Henriksen and Wright 1977; Henriksen  
87 and Grande 2002; De Wit et al. 2014). The native brown trout population disappeared around 1960,  
88 probably because of acidification (Henriksen and Grande 2002). pH in the lake was generally below  
89 5.0 in the 1970s. Since then, the lake has been stocked several times for research purposes with  
90 brown trout, brook trout (*Salvelinus fontinalis*) and rainbow trout (*Oncorhynchus mykiss*) to study  
91 the relative tolerance of various fish species to acid water (Grande et al. 1978). The stocking did not  
92 result in natural reproduction in the lake, until recent findings of small, non-stocked brown trout.  
93 Here we investigate the recent fish recovery in relation to changes in the water chemistry of Lake  
94 Langtjern over the 42-year period 1973–2014.

95

## 96 Materials and methods

### 97 The catchment and the lake

98 Lake Langtjern (<https://www.niva.no/en/services/environmental-monitoring/langtjern>) is a  
99 small, headwater lake located in Flå township, Buskerud county, about 75 km north of Oslo, south-  
100 eastern Norway (Fig. 1). Lake Langtjern has been a site for monitoring and research on acidification  
101 since 1972. It is undisturbed from direct human influence and has never been limed. The lake and its  
102 catchment are included in several national and international monitoring programs. The lake is 0.23  
103 km<sup>2</sup> with catchment including the lake 4.7 km<sup>2</sup>. The lake is relatively shallow, with maximum depth  
104 12 m and mean depth 2 m. The catchment is mixed sparse forest of pine, spruce and birch. Soils are

105 thin and organic-rich podsols developed on weathering-resistant glacial moraine and bedrock of  
106 gneisses and granites. The catchment is 63 % forest, 16 % peatland and 16 % exposed bedrock. Apart  
107 from a minor amount of forest harvesting, there is no other human disturbance or sources of  
108 pollution to the lake or the catchment. The lake and its catchment including exclusive fishing rights  
109 have been leased to the Norwegian Institute for Water Research for research and monitoring  
110 purposes since 1973.

111           The lake has three inflowing streams; only the largest of these (LAE02) and the outlet  
112 (LAE01) provide suitable habitat as spawning beds for trout. The width of the outlet stream is 1–2 m.  
113 The outlet has two dam constructions: an old large stone dam previously used to float timber and a  
114 newer concrete dam downstream with v-notch weir installed in 1973 for measuring discharge. The  
115 older stone dam restricts free water flow when discharge is large and has an opening at the bottom  
116 which allows fish passage both ways at all levels of water discharge. The v-notch weir has a waterfall  
117 that effectively prevents any upstream migration of fish. From the lake outlet, it is about 30 m to the  
118 stone dam and 100 m to the concrete dam. The main inlet (LAE02) has a width of about 0.5–1 m.  
119 Both streams have sections of shallow water, but also some deeper pools. The substrate varies from  
120 fine to coarse. Suitable brown trout spawning substrates are more common in the inlet (LAE02) than  
121 in the outlet.

122 Deposition data

123           The nearby stations Brekkebygda (1998–2015) and Gulsvik (1974–1997) are part of the  
124 national atmospheric monitoring programme run by the Norwegian Institute for Air Research (NILU).  
125 Bulk deposition is collected in weekly samples, and volume and concentrations of major ions  
126 measured at NILU and reported annually (Aas et al. 2016).

127 Discharge data

128           Langtjern is a station in the national hydrology monitoring programme run by The  
129 Norwegian Water Resources and Energy Directorate (NVE). Discharge is measured continuously by  
130 water-level recorder at the v-notch weir. The data are reported as mean daily discharge.

131 Water chemistry data

132           Samples for water chemistry have been collected weekly from the outlet since 1973, except  
133 during an 18-month period with no funding in 1984–1985. Water chemistry parameters relevant to  
134 acidification have been analysed; these include major cations and anions, aluminium species (from  
135 1986), and dissolved organic carbon (from 1986). The inlets and the lake itself have also been  
136 sampled, but not as frequently or as systematically as the outlet. The monitoring data and analytical  
137 methods used are reported annually (Garmo et al. 2016).

138 Fish data

139           Fish catch and stocking information for the period 1906–1971 was derived from log books  
140 kept by the local fishermen and other anecdotal information from local people. Beginning in 1972  
141 experimental fish stockings and gill net catches were systematically recorded (Henriksen and Grande  
142 2002). During the period 2000–2003, investigations were conducted to assess the potential for  
143 natural recruitment of brown trout in the lake. In autumn 2000, 216 fertilized brown trout eggs of  
144 the Nordmarka (Oslo) strain were placed at potential spawning sites in the outlet (72 eggs in 3  
145 boxes) and main inlet (144 eggs in 6 boxes) and then inspected periodically for survival rates to

146 hatching in May 2001. In September 2002, the streams were sampled by electrofishing with a  
147 backpack apparatus (Bohlin et al. 1989). In August 2003, the lake was sampled by 9 multi mesh  
148 survey nets of 32 m and 4 single mesh nets of 25 m with mesh widths 10 mm, 12.5 mm, 16 mm and  
149 22 mm.

150 In August 2010, October 2011, October 2012, August 2013 and August 2014, the outlet and  
151 main inlet were sampled by one or two passes of electrofishing, except 2014 when only outlet was  
152 sampled. The outlet stream was sampled in an area of ca. 150 m<sup>2</sup>, covering the full width of the  
153 stream from the outlet down to the V-notch weir about 100 m downstream. The inlet was sampled  
154 in an area of ca. 130 m<sup>2</sup>, covering the full width of the stream from the lake to about 130 m  
155 upstream. Length and weight of the captured fish were measured, and the fish then released into  
156 the same stream. Young-of-the-year (age 0+) were counted to assess the year specific reproduction.  
157 This age class was separated from older classes (>0+) mainly by fish lengths, but also by reading  
158 scales samples of some individuals. Gender and sexual maturation were determined only when  
159 possible.

## 160 Statistical methods

161 We analysed for break points in the various data series by means of the software package  
162 Change-Point Analyzer ver. 2.3 (Taylor Enterprises, Inc.), where annual means were analysed for  
163 significant changes with 95 % confidence level using 1000 bootstraps without replacements. In some  
164 series, data were grouped to avoid violation of the assumptions of independent errors.

165 Duration of extremes was estimated by counting days between consecutive measurements  
166 of extreme values, assuming the values to be extreme in the period between the actual  
167 measurements. We used the thresholds of ANC 10 µeq l<sup>-1</sup>, ANC<sub>0aa</sub> -5 µeq l<sup>-1</sup>, pH 4.8 and Al<sub>i</sub> 50 µg l<sup>-1</sup>.  
168 Days between two consecutive measurements were counted if both measured values were below  
169 the thresholds for ANC, ANC<sub>0aa</sub> or pH and above the threshold for Al<sub>i</sub>. Measurements were weekly  
170 and the expected count between two such values was therefore seven.



## 171 Results

### 172 Water chemistry

173           The concentrations of  $\text{SO}_4$  in the outlet have decreased sharply since the peak years in the  
174 1970s (Fig. 2). This has been in response to the large decrease in  $\text{SO}_4$  deposition – from about 40  
175  $\text{meq m}^{-2}\text{yr}^{-1}$  in the late 1970s to about  $10 \text{ meq m}^{-2}\text{yr}^{-1}$  in the 2010s (Fig. 2). The large decrease in  $\text{SO}_4$   
176 deposition from 1991 to 1997 corresponds to the decrease in  $\text{SO}_4$  concentrations in the outlet from  
177 1993 to 2000, with a delayed response of less than two years (Table 1) (Fig. 2). The decreasing  
178 concentrations of  $\text{SO}_4$  in the outlet have been accompanied in part by lower concentrations of base  
179 cations such as Ca, and in part by higher pH and lower concentration of  $\text{Al}_i$ . The ANC and  $\text{ANC}_{\text{Oaa}}$  have  
180 increased, and the water has become less toxic to fish (Table 1). At Langtjern concentrations of  
181 nitrate ( $\text{NO}_3$ ) are in the range  $1\text{--}2 \mu\text{eq l}^{-1}$ , showing lower values in recent decades. Thus nitrogen  
182 deposition plays a minor role relative to sulphur in surface water acidification at this site. TOC has  
183 increased since the mid 1980s, which is the reason for the slight decline in  $\text{ANC}_{\text{Oaa}}$  in the period  
184 2008–2014.

185           The 42-year record of annual  $\text{SO}_4$  deposition and mean concentrations of  $\text{SO}_4$ , Ca, ANC,  
186  $\text{ANC}_{\text{Oaa}}$  and pH in the outlet had change-points during 1997–2001, with decreased  $\text{SO}_4$  deposition,  
187  $\text{SO}_4$  and Ca and increased ANC,  $\text{ANC}_{\text{Oaa}}$  and pH (Fig. 2). A second change point was found in 1991–  
188 1992. The change-points for ANC and  $\text{ANC}_{\text{Oaa}}$  in 2000 had confidence intervals which included 2002  
189 and 2001, respectively. The decrease in  $\text{SO}_4$  of  $41 \mu\text{eq l}^{-1}$  was associated with in a decrease of Ca of  
190  $11 \mu\text{eq l}^{-1}$  and an increase of ANC of  $28 \mu\text{eq l}^{-1}$ .

191           The occurrence and duration of acidic episodes have decreased considerably since the 1980s  
192 (Fig. 3). During 1973–1992 in the Langtjern outlet, there were many periods of more than 90  
193 consecutive days of  $\text{pH} < 4.8$ . Before 1995 it was usual to have more than 14 days of  $\text{pH} < 4.8$  every  
194 year. Then, in the period 1996–2014, there were 11 years without consecutive weekly  
195 measurements of  $\text{pH} < 4.8$ . Periods of  $\text{Al}_i > 50 \mu\text{g l}^{-1}$  were more frequent and much longer before

196 1995 than after. Since 1995, only two years had high  $Al_i$  periods of 14 days or more. Periods of  $ANC <$   
197  $10 \mu eq l^{-1}$  were more frequent and of much longer duration before 1990 than after. After 1990, only  
198 three years had such periods: 1991, 1994 and 2000. For  $ANC_{0aa} < -5 \mu eq l^{-1}$ , there were fewer and  
199 shorter periods after year 2001.

200

201 Fish

202           According to anecdotal information from the local fishermen and log books on fish catches in  
203 Lake Langtjern, the lake lost its population of brown trout during the 1960s, probably due to  
204 acidification. This “original” population was a result of several stockings of brown trout since ca.  
205 1906 and the natural offspring of these. Gill net catches were relatively good for the first decades of  
206 the 1900s, but then very poor in 1967–1969. From 1972, the lake was managed for research  
207 purposes and repeatedly stocked with brown trout and also brook trout usually aged 1+ (Fig. 4)  
208 (Henriksen and Grande 2002). The last stocking was of 400 brown trout in June 2006. There was no  
209 systematic tagging of the stocked fish, but most were fin clipped. The stocked fish were also  
210 captured (and killed), usually by use of gill nets each summer, with the last gill net catch conducted  
211 in 2011. The gill net catches usually corresponded to the previous stocking of fish, *i.e.* the stocked  
212 fish were recognized (by size and fin clippings) in the catches some 2–4 years after release (Fig. 4).  
213 Captured fish were mainly marked (fin clipping) confirming recaptures of stocked fish. Unmarked  
214 captures, which could be wild or stocked, were very rare and did not point to an ongoing natural  
215 reproduction. During the period 1992–2000, recapture of stocked fish was estimated to 20%  
216 (Henriksen and Grande 2002). The remaining 80% of the stocked fish usually disappeared after 6–8  
217 years, presumably owing to natural causes, but possibly as a result of the acidification. There usually  
218 were mature individuals among the captured fish. Probably, there have been mature fish in the lake  
219 in varying numbers since the 1970s, but the gill net captures have not indicated any successful  
220 reproduction of trout.

221 The studies of possible trout recruitment starting in the winter 2000-01 gave negative  
222 results. In May 2001, all eggs in the experimental boxes in the substrate were dead when inspected.  
223 At the same time, dead eggs from natural spawning were also observed. The September 2002  
224 electrofishing resulted in no catches in the streams. The August 2003 lake sampling by gill nets gave  
225 no catch of non-stocked fish. Studies were resumed in 2010, and for the first time recruitment of  
226 young brown trout was observed. The electrofishing in the outlet (LAE01) and the major inlet  
227 (LAE02) in 2010–2014 resulted in captures of non-stocked brown trout each year (Table 2). These  
228 fish were not fin clipped, and they were much smaller than the expected size of the stocked fish  
229 from 2006. In 2011, 2012 and 2014 there were electrofishing catches of young-of-the-year brown  
230 trout, with fish lengths < 8 cm in August catches and < 9 cm in October. Mature individuals of both  
231 sexes, and of both wild (< 27 cm, n=3) and stocked origin (> 38 cm, n=2), were captured in the outlet  
232 in 2011 and 2012.

233

## 234 Discussion

235 The chemical recovery at Langtjern follows the well-documented pattern seen in acidified  
236 lakes and streams in many parts of Europe and eastern North America (Stoddard et al. 1999; Jeffries  
237 et al. 2003; Skjelkvåle et al. 2007; Futter et al. 2014; Monteith et al. 2014; Rask et al. 2014; Driscoll et  
238 al. 2016). The reduced deposition of  $\text{SO}_4$  has led to lower concentrations of  $\text{SO}_4$  in surface waters. pH  
239 and ANC have increased while  $\text{Al}_i$  has decreased. In addition, TOC has increased, which is a result of  
240 increasing organic matter solubility related to lower electrolyte concentrations and reduced acidity  
241 (De Wit et al. 2007). Also, concentrations of  $\text{NO}_3$  decreased, probably as a combined result of climate  
242 warming (less snow cover) and lower N deposition (de Wit et al. 2008).

243 The lag time of <2 years between reductions in  $\text{SO}_4$  deposition and decrease in  
244 concentrations of  $\text{SO}_4$  in the lake observed at Langtjern is also not unexpected, as soil processes such  
245 as adsorption and desorption of  $\text{SO}_4$  are minor in young, organic-rich soils such as those at Langtjern  
246 (Reuss and Johnson 1986). The in-lake processes that also act to dampen changes in  $\text{SO}_4$

247 concentrations are also apparently of minor importance relative to the through-flux of SO<sub>4</sub> in the  
248 lake (Couture et al. 2016).

249         The fish catches indicate an important qualitative change in the brown trout ecology in Lake  
250 Langtjern: the stocked fish now reproduce, *albeit* to a very low extent and maybe not every year.  
251 Thus, water quality in the outlet and inlet streams is apparently close to a critical limit for successful  
252 brown trout reproduction.

253         The exact year of the first successful reproduction in recent times cannot be ascertained, but  
254 we know from the electrofishing that young-of-the-year brown trout were produced in 2011, 2012  
255 and 2014 (Table 2). The estimated age of older captured fish suggests that brown trout reproduced  
256 also in 2008 and 2009. Gill net sampling, electrofishing and egg exposure experiments indicate that  
257 reproduction probably did *not* occur in in 2001 and 2002. The August 2003 gill net sampling in the  
258 lake would not have been able to capture potential young-of-year, as they would have been residing  
259 in the stream; thus there are no data on the trout reproduction in 2003. Hence, the first successful  
260 reproduction was probably in the period 2003–2008.

261         Langtjern is one of a few acid water monitoring sites in Norway at which long-term data  
262 record the recovery of the brown trout following reductions in acid deposition. Hesthagen et al.  
263 (2011) have documented the revitalisation of the brown trout population in Lake Saudlandsvatn,  
264 southernmost Norway. Here the native population was severely depleted, but never completely lost,  
265 and was able to naturally reproduce when water quality improved in the 1990s. Similarly, brown  
266 trout recruitment became increasingly successful in streams in the River Vikedal catchment  
267 (Hesthagen et al. 2001) during the 1990s, *albeit* with occasional setbacks due to acidic episodes  
268 (Hesthagen et al. 2016).

269         There are only a few documented cases of recovery of fish populations from acidified waters  
270 elsewhere in Europe and eastern North America. This appears to be because of the paucity of long-  
271 term data monitoring fish populations, but perhaps also because of factors acting to delay biological  
272 recovery in response to chemical recovery. In Sweden, the thousands of acidified lakes have shown

273 chemical recovery since the 1980s (Futter et al. 2014), but there are apparently few lakes in which  
274 the long-term data are sufficient to document recovery of fish populations (Holmgren 2014). Valinia  
275 et al. (2014) found that in a dataset of 28 Swedish lakes, the roach (*Rutilus rutilus*) population had  
276 been lost in 14 lakes due to acidification in the 1980s, but in 2010 it had reappeared in 5 of these in  
277 response to chemical recovery. In Finland there has been widespread recovery of perch (*Perca*  
278 *fluviatilis*) populations, but the more acid-sensitive roach shows much less recovery (Rask et al.  
279 2014). In the United Kingdom two of the 22 sites in the acid waters monitoring network (AWMN)  
280 now show recovery of brown trout populations (Malcolm et al. 2014) in response to the general  
281 improvement in water quality due to reduced sulphur deposition (Monteith et al. 2014). In the  
282 eastern United States long-term monitoring data from 43 lakes in the Adirondack Mountains, New  
283 York, show reduced acidity in response to decreased sulphur deposition, but so far there have been  
284 no major improvements in populations of brook trout (Baldigo et al. 2016). Likewise in eastern  
285 Canada there have been several reports of improved fish populations in acidified lakes and streams  
286 of Atlantic Canada (Lacoul et al. 2011) and in acidified lakes near Sudbury, Ontario (Gunn and Keller  
287 1990, Snucins et al. 2001).

288         Chemical recovery proceeds along a continuum, whereas biological recovery is often marked  
289 by thresholds. There have been many studies of the tolerance of fish species to acidified waters, in  
290 particular the brown trout. Empirical data for water chemistry and brown trout populations from  
291 synoptic surveys of 1000 lakes in Norway show that there were rather sharp thresholds of ANC for  
292 the transition between “not affected” and “reduced” populations, and between “reduced” and  
293 “extinct” populations in the 1980s (Bulger et al. 1993, Lien et al. 1996). Fitting a logistic expression to  
294 the data explained 54% of the variance. Including organic acids in the expression for  $ANC_{Oaa}$   
295 increased the strength of these relationships to 56% (Lydersen et al. 2004).  $ANC_{Oaa}$  is particularly  
296 appropriate in humic lakes, such as Langtjern. A threshold for  $ANC_{Oaa}$  of 8  $\mu\text{eq/l}$  gave a 95%  
297 probability for no population damage to brown trout in Norwegian lakes based on the survey data  
298 from 1986.

299 Hesthagen et al. (2008)) revisited these thresholds based on a new survey of the Norwegian  
300 lakes conducted in 1995. Their analysis indicates that in 1995 the threshold for 95% probability for  
301 no population damage to brown trout was  $ANC_{Oaa}$  48  $\mu\text{eq/l}$ , substantially higher than the value for  
302 the 1986 data. They suggest that the higher  $ANC_{Oaa}$  threshold found for the 1995 data might be  
303 caused by a lower pH and a higher  $Al_i$  concentration at a given ANC value in 1995 than in the 1980s.  
304 But this difference could also be caused by the lag times between changes in water chemistry and  
305 population status in lakes.

306 Based on the long-term field data from the streams in the River Vikedal catchment  
307 Hesthagen et al. (2016) suggest that recruitment of brown trout can give low density of fry at  $ANC_{Oaa}$   
308 levels of -18 to -5  $\mu\text{eq l}^{-1}$ , increased but unstable densities at  $ANC_{Oaa}$  -5 to +10  $\mu\text{eq l}^{-1}$ , and steady  
309 increase in density at  $ANC_{Oaa}$  above 10  $\mu\text{eq l}^{-1}$ . They indicate that  $ANC_{Oaa}$  of 20–25  $\mu\text{eq l}^{-1}$  is necessary  
310 for significant recovery of young brown trout in streams. This value is consistent with the observed  
311 fish recovery at Lake Saudlandsvatn (Hesthagen et al. 2011). The UK data of Malcolm et al. (2014)  
312 indicate threshold value of  $ANC_{Oaa}$  in the range 7 to 38  $\mu\text{eq l}^{-1}$ , for 80% probability of brown trout fry  
313 present in two of three sampled stream reaches.

314 The data from Langtjern fit this picture. For the period 2008–2014 during which  
315 reproduction occurred, the outlet water chemistry mean values of  $ANC_{Oaa}$  was 10  $\mu\text{eq l}^{-1}$  (Table 1),  
316 the threshold indicated by Hesthagen et al. (2016)) for unstable densities of young brown trout in  
317 running water. Further, the mean values for 2008–2014 were similar to the mean values for 2000–  
318 2007, suggesting that the conditions were close to the critical limits also prior to 2008.

319 The ANC in Lake Langtjern is not likely to further increase appreciably soon. There is little  
320 room for further reductions in  $SO_4$  deposition as levels in 2015 were only 7% of those in the peak  
321 year 1980 (Aas et al. 2016). Nitrate makes only a minor contribution to ANC and appears to be  
322 declining (De Wit et al. 2008). Thus any major increase in ANC will have to come from increasing  
323 concentrations of base cations caused by replenishment of soil base cation pools due to natural  
324 weathering, a process that typically takes decades (Hodson and Langan 1999).  $ANC_{Oaa}$ , on the other

325 hand, could increase with a decline in TOC. ANCoaa is the organic acid adjusted ANC, where organic  
326 acids (TOC) are subtracted from the base cation concentration to give an adjusted, and reduced,  
327 ANC<sub>oaa</sub>. However, TOC concentrations do not show any sign of levelling off and may increase further  
328 under a wetter climate (de Wit et al. 2016).

329 The change-points for ANC and ANC<sub>oaa</sub> in 2000 may explain why the successful reproduction  
330 of brown trout started at some point after 2002. Although the estimated change-point in 2000 does  
331 not fit with the different investigations indicating no reproduction during 2001–2002, the confidence  
332 interval for the ANC change-point was 2000–2002, and the upper limit makes it possible that the  
333 positive change in ANC level occurred shortly after the known period of non-successful  
334 reproduction. The 1997 change-point in SO<sub>4</sub> indicates that the reduced SO<sub>4</sub> concentration was the  
335 main cause for the ANC upward change, although the two changes were not estimated to occur at  
336 the same time.

337 ANC is a convenient measure of lake acidification. Toxicity to fish, however, is caused by Al<sub>i</sub>  
338 and/or H<sup>+</sup>. The recent reproductive success might better be explained by lower frequency, severity  
339 and duration of toxic episodes rather than increased mean ANC levels (Baker et al. 1982). In the  
340 1980s, Lake Langtjern experienced long periods of low pH, low ANC and high Al<sub>i</sub> (Fig. 3). Episodes of  
341 pH < 4.8 decreased considerably both in duration and frequency since the peak in 1989, but periods  
342 of pH < 4.8 still occur, *e.g.* a possible 15-day period in 2012. If these periods cause mortality in the  
343 youngest individuals or the fertilized eggs in the stream substrate, the population would still have  
344 irregular setbacks in producing offspring. Serrano et al. (2008) proposed that pH, not Al, is most  
345 important for trout survival in organic-rich boreal streams. Juvenile brown trout mortality in such  
346 streams was modelled with 80 % mortality during 14 days of pH 4.8. The Lake Langtjern outlet had  
347 episodes in 2007 and 2009 where pH possibly was below 4.8 for 63 and 35 days, respectively. These  
348 episodes may have inflicted high mortality in several year-classes of brown trout, even though the  
349 yearly means for pH were 5.0 and 5.1, respectively. Longer episodes of Al<sub>i</sub> > 50 µg l<sup>-1</sup> have been few

350 since 1997, but week-long episodes probably occurred in both 2006 and 2007. Such episodes may  
351 have caused high mortality in young-of-the-year brown trout.

352 In addition to the water chemistry, habitat characteristics probably limit the brown trout  
353 population in Lake Langtjern. Suitable spawning areas are few and the number of spawning fish is  
354 low, as the remaining individuals from the stocking in 2006 and 2003 probably only exist in small  
355 numbers. The inlet stream (LAE02) has more suitable substrate, but it is also a smaller stream than  
356 the outlet. Both streams are subject to winter and summer droughts. Our catches document that  
357 successful spawning is occurring. The number of spawners has been higher before, without resulting  
358 in successful reproductions. The present reproduction therefore indicates that the change in water  
359 chemistry is the crucial factor.

360

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## 367 References

- 368 Aas, W., Fiebig, M., Platt, S., Solberg, S., and Yttri, K.E. 2016. Monitoring of long-range transported  
369 air pollutants in Norway, annual report 2015. *Miljødirektoratet rapport, M-562/2016, NILU*  
370 *report, 13/2016*. Norwegian Institute for Air Research, Kjeller, Norway.
- 371 Baker, J.P., and Schofield, C.L. 1982. Aluminum toxicity to fish in acidic waters. *Water Air Soil*  
372 *Pollution 18(1-3): 289-309*.



373 Baldigo, B.P., Roy, K.M., and Driscoll, C.T. 2016. Response of fish assemblages to declining acidic  
374 deposition in Adirondack Mountain lakes, 1984-2012. *Atmospheric Environment* 146: 223-  
375 235.

376 Bohlin, T., Hamrin, S., Heggberget, T.G., Rasmussen, G., and Saltveit, S.J. 1989. Electrofishing - theory  
377 and practice with special emphasis on salmonids. *Hydrobiologia* 173: 9-43.

378 Bulger, A.J., Lien, L., Cosby, B.J., and Henriksen, A. 1993. Brown trout (*Salmo trutta*) status and  
379 chemistry from the Norwegian thousand lake survey: statistical analysis. *Canadian Journal of*  
380 *Fisheries and Aquatic Sciences* 50: 575-585.

381 Couture, R.M., Fischer, R., Van Cappellen, P., and Gobeil, C. 2016. Non-steady state diagenesis of  
382 organic and inorganic sulfur in lake sediments. *Geochimica Et Cosmochimica Acta* 194: 15-33.

383 Cronan, C.S., Walker, W.J., and Bloom, P.R. 1986. Predicting aqueous aluminium concentrations in  
384 natural waters. *Nature* 324: 140-143.

385 De Wit, H.A., Mulder, J., Hindar, A., and Hole, L. 2007. Long-term increase in dissolved organic  
386 carbon in streamwaters in Norway is response to reduced acid deposition. *Environmental*  
387 *Science & Technology* 41(22): 7706-7713.

388 de Wit, H.A., Hindar, A., and Hole, L. 2008. Winter climate affects long-term trends in stream water  
389 nitrate in acid-sensitive catchments in southern Norway. *Hydrology and Earth System*  
390 *Sciences* 12(2): 393-403.

391 De Wit, H.A., Granhus, A., Lindholm, M., Kainz, M.J., Lin, Y., Braaten, H.F.V., and Blaszcak, J.R. 2014.  
392 Forest harvest effects on mercury in streams and biota in Norwegian boreal catchments.  
393 *Forest Ecology and Management* 324: 52-63.

394 de Wit, H.A., Valinia, S., Weyhenmeyer, G.A., Futter, M.N., Kortelainen, P., Austnes, K., Hessen, D.O.,  
395 Raike, A., Laudon, H., and Vuorenmaa, J. 2016. Current Browning of Surface Waters Will Be  
396 Further Promoted by Wetter Climate. *Environmental Science & Technology Letters* 3(12):  
397 430-435.

398 Driscoll, C.T., Driscoll, K.M., Fakhraei, H., and Civerolo, K. 2016. Long-term temporal trends and  
399 spatial patterns in the acid-base chemistry of lakes in the Adirondack region of New York in  
400 response to decreases in acidic deposition. *Atmospheric Environment* 146: 5-14.

401 Futter, M.N., Valinia, S., Lofgren, S., Kohler, S.J., and Folster, J. 2014. Long-term trends in water  
402 chemistry of acid-sensitive Swedish lakes show slow recovery from historic acidification.  
403 *Ambio* 43: 77-90.

404 Garmo, O.A., Skjelkvale, B.L., de Wit, H.A., Colombo, L., Curtis, C., Folster, J., Hoffmann, A., Hruska, J.,  
405 Hogasen, T., Jeffries, D.S., Keller, W.B., Kram, P., Majer, V., Monteith, D.T., Paterson, A.M.,  
406 Rogora, M., Rzychon, D., Steingruber, S., Stoddard, J.L., Vuorenmaa, J., and Worsztynowicz,  
407 A. 2014. Trends in Surface Water Chemistry in Acidified Areas in Europe and North America  
408 from 1990 to 2008. *Water Air Soil Pollut.* 225(3).

409 Garmo, Ø., Skancke, L.B., and Høgåsen, T. 2016. Monitoring long-range transboundary air pollution.  
410 Water chemical effects 2015. *NIVA-rapport 7078, Miljødirektoratet-rapport M-613*.  
411 Norwegian Institute for Water Research, Oslo.

412 Grande, M., Muniz, I.P., and Andersen, S. 1978. Relative tolerance of some salmonids to acid waters.  
413 *Verhandlungen des Internationalen Verein Limnologie* 20: 2076-2084.

414 Gray, C., Hildrew, A.G., Lu, X., Ma, A., McElroy, D., Monteith, D., O'Gorman, E., Shilland, E., and  
415 Woodward, G. 2016. Recovery and Nonrecovery of Freshwater Food Webs from the Effects  
416 of Acidification. In *Advances in Ecological Research, Vol 55: Large-Scale Ecology: Model  
417 Systems to Global Perspectives*. Edited by A.J. Dumbrell, R.L. Kordas and G. Woodward.  
418 Elsevier Academic Press Inc, San Diego. pp. 475-534.

419 Gunn, J.M., and Keller, W. 1990. Biological recovery of an acid lake after reductions in industrial  
420 emissions of sulphur. *Nature* 345 (6274): 431-433

421 Henriksen, A., and Grande, M. 2002. Lake Langtjern - fish studies in the Langtjern area 1966-2000.  
422 *Acid Rain Research Report 54/02 SNO 4537-2002*. NIVA, Oslo.

423 Henriksen, A., and Wright, R.F. 1977. Effects of acid precipitation on a small acid lake in southern  
424 Norway. *Nordic Hydrology* 8: 1-10.

425 Hesthagen, T., Sevaldrud, I.H., and Berger, H.M. 1999. Assessment of damage to fish populations in  
426 Norwegian lakes due to acidification. *Ambio* 28: 112-117.

427 Hesthagen, T., Forseth, T., Saksgard, R., Berger, H.M., and Larsen, B.M. 2001. Recovery of young  
428 brown trout in some acidified streams in southwestern and western Norway. *Water Air Soil*  
429 *Pollut.* 130(1-4): 1355-1360.

430 Hesthagen, T., Fiske, P., and Skjelkvale, B.L. 2008. Critical limits for acid neutralizing capacity of  
431 brown trout (*Salmo trutta*) in Norwegian lakes differing in organic carbon concentrations.  
432 *Aquatic Ecology* 42(2): 307-316.

433 Hesthagen, T., Fjellheim, A., Schartau, A.K., Wright, R.F., Saksgård, R., and Rosseland, B.O. 2011.  
434 Chemical and biological recovery of Lake Saudlandsvatn, a highly acidified lake in  
435 southernmost Norway, in response to decreased acid deposition. *Science of the Total*  
436 *Environment* 409: 2908-2916.

437 Hesthagen, T., Fiske, P., and Saksgard, R. 2016. Recovery of young brown trout (*Salmo trutta*) in  
438 acidified streams: What are the critical values for acid-neutralizing capacity? *Atmospheric*  
439 *Environment* 146: 236-244.

440 Hodson, M.E., and Langan, S.J. 1999. Considerations of uncertainty in setting critical loads of acidity  
441 of soils: the role of weathering rate determination. *Environmental Pollution* 106(1): 73-81.

442 Holmgren, K. 2014. Challenges in assessing biological recovery from acidification in Swedish lakes.  
443 *Ambio* 43: 19-29.

444 Jeffries, D.S., Clair, T.A., Couture, S., Dillon, P.J., Dupont, J., Keller, W., McNicol, D.K., Turner, M.A.,  
445 Vet, R., and Weeber, R. 2003. Assessing the recovery of lakes in southeastern Canada from  
446 the effects of acid deposition. *Ambio* 32: 176-182.

447 Lacoul, P., Freedman, B., and Clair, T. 2011. Effects of acidification on aquatic biota in Atlantic  
448 Canada. *Environmental Reviews* 19: 429-460.

449 Lien, L., Raddum, G.G., Fjellheim, A., and Henriksen, A. 1996. A critical limit for acid neutralizing  
450 capacity in Norwegian surface waters, based on new analyses of fish and invertebrate  
451 responses. *Science of the Total Environment* 177: 173-193.

452 Lydersen, E., Larssen, T., and Fjeld, E. 2004. The influence of total organic carbon (TOC) on the  
453 relationship between acid neutralizing capacity (ANC) and fish status in Norwegian lakes.  
454 *Science of the Total Environment* 362: 63-69.

455 Malcolm, I.A., Bacon, P.J., Middlemas, S.J., Fryer, R.J., Shilland, E.M., and Collen, P. 2014.  
456 Relationships between hydrochemistry and the presence of juvenile brown trout (*Salmo*  
457 *trutta*) in headwater streams recovering from acidification. *Ecological Indicators* 37: 351-364.

458 Monteith, D.T., Evans, C.D., Henrys, P.A., Simpson, G.L., and Malcolm, I.A. 2014. Trends in the  
459 hydrochemistry of acid-sensitive surface waters in the UK 1988-2008. *Ecological Indicators*  
460 37: 287-303.

461 Overrein, L., Seip, H.M., and Tollan, A. 1980. Acid precipitation - Effects on forest and fish. *Final*  
462 *report of the SNSF-project 1972-1980. FR 19-80. SNSF project, Ås, Norway.*

463 Rask, M., Vuorenmaa, J., Nyberg, K., Tammi, J., Mannio, J., Olin, M., Kortelainen, P., Raitaniemi, J.,  
464 and Vesala, S. 2014. Recovery of acidified lakes in Finland and subsequent responses of  
465 perch and roach populations. *Boreal Environment Research* 19(3): 222-234.

466 Reuss, J.O., and Johnson, D.W. 1986. *Acid Deposition and the Acidification of Soils and Waters.*  
467 Springer Verlag, New York.

468 Reuss, J.O., Cosby, B.J., and Wright, R.F. 1987. Chemical processes governing soil and water  
469 acidification. *Nature* 329: 27-32.

470 Reuss, J.O., Roswall, E.C., and Hopper, R.W.E. 1990. Aluminium Solubility, Calcium-Aluminum  
471 Exchange, and pH in acid forest soils. *Soil Science Society of America Journal* 54: 374-380.

472 Rosseland, B.O., Eldhuset, T.D., and Staurnes, M. 1990. ENVIRONMENTAL-EFFECTS OF ALUMINUM.  
473 *Environmental Geochemistry and Health* 12(1-2): 17-27.

474 Schöpp, W., Posch, M., Mylona, S., and Johansson, M. 2003. Long-term development of acid  
475 deposition (1880-2030) in sensitive freshwater regions in Europe. *Hydrology and Earth  
476 System Sciences* 7: 436-446.

477 Serrano, I., Buffam, I., Palm, D., Brannas, E., and Laudon, H. 2008. Thresholds for survival of brown  
478 trout during the spring flood acid pulse in streams high in dissolved organic carbon.  
479 *Transactions of the American Fisheries Society* 137: 1363-1377.

480 Skjelkvåle, B.L., Wright, R.F., and Henriksen, A. 1998. Norwegian lakes show widespread recovery  
481 from acidification: results of national surveys of lakewater chemistry 1986-1997. *Hydrology  
482 and Earth System Sciences* 2: 555-562.

483 Skjelkvåle, B.L., Borg, H., Hindar, A., and Wilander, A. 2007. Large scale patterns of chemical recovery  
484 in lakes in Norway and Sweden: Importance of seasalt episodes and changes in dissolved  
485 organic carbon. *Applied Geochemistry* 22(6): 1174-1180.

486 Snucins, E., Gunn, J., Keller, B., Dixit, S., Hindar, A., and Henriksen, A. 2001. Effects of regional  
487 reductions in sulphur deposition on the chemical and biological recovery of lakes within  
488 Killarney Park, Ontario, Canada. *Environmental Monitoring and Assessment* 67(1-2): 179-194.

489 Stoddard, J.L., Jeffries, D.S., Lükewille, A., Clair, T.A., Dillon, P.J., Driscoll, C.T., Forsius, M.,  
490 Johannessen, M., Kahl, J.S., Kellogg, J.H., Kemp, A., Mannio, J., Monteith, D., Murdoch, P.S.,  
491 Patrick, S., Rebsdorf, A., Skjelkvåle, B.L., Stainton, M.P., Traaen, T.S., van Dam, H., Webster,  
492 K.E., Wieting, J., and Wilander, A. 1999. Regional trends in aquatic recovery from  
493 acidification in North America and Europe 1980-95. *Nature* 401: 575-578.

494 UNECE. 2014. Convention on Long-range Transboundary Air Pollution.  
495 [http://www.unece.org/env/lrtap/lrtap\\_h1.html](http://www.unece.org/env/lrtap/lrtap_h1.html)

496 Valinia, S., Englund, G., Moldan, F., Futter, M.N., Kohler, S.J., Bishop, K., and Folster, J. 2014.  
497 Assessing anthropogenic impact on boreal lakes with historical fish species distribution data  
498 and hydrogeochemical modeling. *Global Change Biology* 20(9): 2752-2764.

499 Wright, R.F., and Henriksen, A. 1978. Chemistry of small Norwegian lakes with special reference to  
500 acid precipitation. *Limnology and Oceanography* 23: 487-498.

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504 **Table 1** Lake Langtjern outlet water chemistry mean values  $\pm$  SD in five periods from 1973 to 2014. No data for  
505 1984–1985. Data for  $Al_i$ , TOC and  $ANC_{Oaa}$  from 1986

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Period	$SO_4$ $\mu eq\ l^{-1}$	$NO_3$ $\mu eq\ l^{-1}$	pH	$Al_i$ $\mu g\ l^{-1}$	ANC $\mu eq\ l^{-1}$	$ANC_{Oaa}$ $\mu eq\ l^{-1}$	TOC $mg\ C\ l^{-1}$	Ca $\mu eq\ l^{-1}$
1973–1979	$72 \pm 14$	$1.9 \pm 1.3$	$4.9 \pm 0.2$		$27 \pm 11$			$69 \pm 12$
1980–1989	$66 \pm 14$	$1.7 \pm 1.3$	$4.8 \pm 0.2$	$74 \pm 19$	$13 \pm 8$	$-15.3 \pm 8.8$	$8.8 \pm 1.6$	$57 \pm 11$
1990–1999	$51 \pm 14$	$1.5 \pm 1.2$	$5.0 \pm 0.2$	$46 \pm 20$	$32 \pm 12$	$-2.3 \pm 9.1$	$10.1 \pm 2.0$	$55 \pm 10$
2000–2007	$25 \pm 6$	$0.9 \pm 0.7$	$5.1 \pm 0.2$	$27 \pm 9$	$47 \pm 12$	$10.6 \pm 8.7$	$10.8 \pm 1.9$	$47 \pm 10$
2008–2014	$17 \pm 4$	$0.8 \pm 0.7$	$5.1 \pm 0.2$	$26 \pm 7$	$47 \pm 9$	$9.7 \pm 6.9$	$11.0 \pm 2.0$	$42 \pm 8$

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510 **Table 2** Number of non-stocked fish caught by electrofishing in the outlet and major inlet to Lake Langtjern in  
511 the period 2010–2014. No data for inlet 1014

		Age class		Year		
		2010	2011	2012	2013	2014
Outlet	0+	0	2	13	0	4
	>0+	6	5	1	2	0
Inlet	0+	0	0	1	0	n.a.
	>0+	1	1	2	2	n.a.

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517 **Fig. 1** Map showing the position and the catchment (dotted line) of Lake Langtjern (60.37 N, 9.73 E), a research  
518 station for studying acidification of surface waters in Norway. Outlet (LAE01) and inlets (LAE03, LAE02 and  
519 LAE08) are indicated

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521

522 **Fig. 2** Annual deposition of  $\text{SO}_4$ , and yearly mean concentrations of  $\text{SO}_4$ , Ca, ANC,  $\text{ANC}_{\text{Oaa}}$ , pH and  $\text{Al}_i$  in the  
523 outlet at Langtjern over the period 1973–2014. Deposition data from Aas et al. (2016)). No outlet data for  
524 1984–1985. TOC and  $\text{Al}_i$  analysed from 1986. Dotted lines indicate levels of significant changes in the data  
525 series

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528 **Fig. 3** Count of consecutive days of  $\text{pH} < 4.8$ ,  $\text{ANC} < 10 \mu\text{eq l}^{-1}$ ,  $\text{ANC}_{\text{Oaa}} < -5 \mu\text{eq l}^{-1}$  and  $\text{Al}_i > 50 \mu\text{g l}^{-1}$  at the  
529 outlet of Lake Langtjern 1973–2014. No data for 1984–1985, and data for  $\text{ANC}_{\text{Oaa}}$  and  $\text{Al}_i$  from 1986

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532 **Fig. 4** Number of stocked (top) and captured (bottom) brown trout ( $n/100 \text{ m}^2$ ) in Lake Langtjern in the period  
533 1972–2011.

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