Water chemistry and acidification recovery in Rogaland County

By Espen Enge

Espen Enge is senior engineer, Environmental Division, County Governor of Rogaland.

Sammendrag

Redusert forsuring av fjellvann i Rogaland fylke. Her bearbeides resultater fra i alt 1144 prøver fra 2002, 2007 og 2012. Tre ulike forsuringsmodeller antydet at forsuringen i fjellområdene i Rogaland (>500 m) i dag er begrenset, og at vannkvaliteten trolig er nær en opprinnelig uforsuret vannkvalitet. Mange lavereliggende innsjøer var tilsynelatende fortsatt forsuret, men disse estimatene kan være forbundet med usikkerhet. Ioneinnholdet i vannet i fjellområdene er ekstremt lavt, noe som i seg selv trolig utgjør en begrensning for utbredelsen av aure.

Summary

The current study compiles data from 1144 water samples from three large regional water chemical surveys, performed in 2002, 2007 and 2012. Three different models suggested that the acidification of the mountain lakes in Rogaland (>500 m) has declined to a minimum, and subsequently that the water chemistry is close to "pre-acidification" conditions. Many low altitude lakes were apparently still acidified, but these estimates were associated with somewhat uncertainty. The conductivity of the water in the mountain areas is extremely low, and is possibly restricting the distribution of brown trout.

Introduction

Due to exceptionally slow weathering bedrock, the concentrations of ions in river and lake water in the mountain areas of southern Norway are generally very low (Wright and Henriksen 1978). In Rogaland, 12% of the surveyed lakes in 1974-1979 (Sevaldrud and Muniz 1980) had conductivity of <10 µS/cm, and the minimum value was 4.2 µS/cm.

Dilute, weakly buffered water is highly sensitive to acidification. Thus, as early as the 1870s the brown trout population (*Salmo trutta*) in Sandvatn in Hunnedalsheiene declined close to extinction (Huitfeldt-Kaas 1922), possibly due to emerging acidification (Qvenild et al. 2007). In the 1920s massive fish kills due to acidic water were observed in the salmon rivers Dirdal, Frafjord and Espedal (Huitfeldt-Kaas 1922). Coincidentally, mass death of brown trout was observed in some of the mountain lakes, e.g. in Månavatn (Huitfeldt-Kaas 1922). In the following decades the acidification problems increased (Hesthagen et al. 1999), and in 1970-1980 as much as 41% of the county area was severely affected by acidification (Sevaldrud and Muniz 1980).

Several large regional water chemical surveys have documented the water chemical effects of the acidification in Norway. The most important of these surveys were the comprehensive samp-

Sandvatn (altitude 910 m). In these remote, barren mountain areas the water is extremely dilute. According to Huitfeldt-Kaas (1922) the brown trout population in Sandvatn was close to extinction as early as the 1870s (Photo: Anne E. Carlsen).

ling of 700 lakes in southern Norway in 1974-1975 (Wright and Snekvik 1978), the "1000 lakes survey" in 1986 (Lien et al. 1987) and the regional survey of 1500 lakes in 1995 (Skjelkvåle et al. 1996).

Due to several international emission control agreements (unece.org 2013), the acid deposition over Norway have been considerably reduced over the last 2-3 decades. Sulfate in surface water has decreased by 43-86% throughout 1980-2010, and a distinct increase in pH and ANC has been observed (Skjelkvåle 2011). In 14 lakes in "Sør-Vestlandet", including Rogaland, a 74% decrease in non-marine sulfate has been observed between 1986 and 2010 (Skjelkvåle 2011).

Over the last decade, the County Governor of Rogaland has performed several large scale water chemical surveys. In the current study this data set has been used to update the acidification status in the county and predict the remaining recovery potential.

Material and methods

The sampling included lakes from all over Rogaland County, from sea level up to an altitude of 1324 m. To assure a convenient map overlap to the neighboring counties in the east (Aust- and Vest-Agder), a few samples from these two counties, located along the border, were also included.

The intention was to sample the same lakes all three years (2002, 2007 and 2012), but minor changes in the selection were, however, unavoidable (will be discussed later). Primarily, lake locations were sampled, but occasionally rivers and streams were sampled too. Water was sampled at the lake outlets, but in some lakes, however, the samples had to be collected as surface samples along the shoreline of the lakes. In 2002 the water was sampled in new 125 ml HDPE bottles ("Nalgene"), in 2007 in 250 ml PE bottles ("Assistent") and in 2012 in 150 ml HDPE bottles ("Mellerud Plast").

Sampling was performed by local environ-

mental authorities ("kommuner"), landowners and landowners associations, power companies, tourist associations and other volunteers. The sampling period was set to June, July and August. Sampling beyond this period was also accepted, e.g. sampling in September/October for lakes in remote high altitude areas having snowmelt throughout the entire summer. The samples were stored cold, and sent to the laboratory within a few days after sampling.

The median time between sampling and the samples being received at the laboratory, was 3 days. The median time further to analysis of pH, conductivity, color and alkalinity was 1 day (2012). The other three parameters accept a certain storage time (Eaton et al. 1995), but were anyhow analyzed within a few days.

pH, conductivity, color and Ca were measured on all samples in all years, and alkalinity, Na and Cl in 2012 only. There have been some minor changes in analytical equipment throughout the years, but except for the determinations of color, the instrumentation are comparable. pH was measured with pH-meters Radiometer PHM92 and VWR "pHenomenal". Calibration was performed with standard buffers (pH=4.01 and 6.86/7.00). The applied electrodes were Metrohm "Aquatrode" (2002), Radiometer GK2401C (2007&2012) and Radiometer pHC2001 (2012), all excellent for low conductivity water measurements. Intercalibration of the electrodes showed agreement in the range of ±0.02 pH. The conductometers used, Petracourt PCM1 (2002), HACH CO150 (2002), Amber Science mod. 1056 (2007&2012) and Cyberscan PC300 (2007&2012) were calibrated with standard KCl-solutions. Intercalibration of the conductometers showed agreement to ±0.5 µS/cm in the current measuring range. Color was measured with comparator HACH CO-1 (2002&2007) and photometric (445 nm) without filtration (2012). Thus, the color results are not directly comparable between 2002/2007 and 2012. Alkalinity was titrated with sulfuric acid to fixed endpoint pH=4.50, and equivalence alkalinity ("ALKe") was calculated according to Henriksen (1982a). Na and Cl were measured with Radiometer and Metrohm ion selective electrodes (ISE), according to the electrode manuals. Ca was measured with Radiometer electrode ISE25Ca according to Enge et al. (in prep.). Non marine fractions of ions (notation: *) were calculated by subtracting the marine fractions, estimated from Cl, from the measured concentrations (Skartveit 1980). H^+ adjusted conductivity was calculated by subtracting 0.35 µS/cm per µeq/l H^+.

The chemical measurements were subject to internal and external quality assurance. Participation at "SIFF-Ringtest" in 2002, demonstrated results comfortably within the acceptance limits for all parameters and test samples.

Several advanced water chemical models for assessing acidification have been developed, e.g. the frequently used "MAGIC" model (Cosby et al. 1985). Such models are, however, highly demanding with respect to both water chemical and catchment input data. Thus, three simpler models were applied at the current data material:

1. A general evaluation of the acidification was performed with the classical "Henriksen acidification indicator" (Henriksen 1978).
2. Acidification was also estimated as loss of alkalinity (Henriksen 1978). Due to the lack of Mg-data, the original alkalinity ("ALK_0") was estimated using non marine Ca only, according to Henriksen (1980): $ALK_0=1.21Ca^*$. Henriksen (1978) suggests a "background acidification" of 10-20 µeq/l in granitic areas in southern Norway. Thus, the lakes that had an alkalinity loss >20 µeq/l were considered as "acidified", while those with a loss of <10 µeq/l were considered as "not acidified".
3. Estimated pre-acidification pH-values (pH_0), according to Hindar and Wright (2002), were compared to observed pH. p_{CO2} was set to 4x the atmospheric partial pressure (Enge 2009).

Estimations of pre-acidification pH require analysis of all major constituents in water. The "1000-lakes survey" in 1986 (Lien et al. 1987) is

one of the few large regional surveys meeting this requirement. The 32 lakes included in both the 1000-lakes survey and the current study, were used.

Mapping was performed with ArcGIS/ArcInfo (v.10.1). The Thiessen polygon method was applied for map presentations. A "redraw" of an old map of pH-values in the lakes in Rogaland from the mid 1980s (Enge and Lura 2003) has been included in the current study. Originally, this map was drawn by hand, but due to the requirement of an objective basis for comparison, this map was redrawn by using Thiessen polygons.

Results

Water chemistry

A total of 1144 samples, table 1, were collected. This included 21 samples from the neighboring counties, located along the border. Small changes in the lake selections throughout the 2000s were unavoidable. However, due to the very large number of lakes in all the selections, evenly scattered throughout the county, the effects of slightly different selections will statistically be negligible.

A general observation was that the chemistry of many of the low altitude lakes showed serious deviations from the natural state. Generally, these lakes were located in areas heavily affected by human activities (populated areas, extensively exploited farming land, industry, etc.), causing pollution and possibly other severe water chemical interferences. Pollution may also directly affect the ISE-measurements (Ion Selective Electrodes).

The water chemistry showed distinct geographic gradients, figure 1 and figure 2. Most apparent was the decreasing conductivity, Ca and color along a southwest to northeast gradient, while pH decreased along a northwest to southeast gradient.

The marine exposure was highly correlated to geographic parameters. Multiple regression of log{Cl} against UTM-East, UTM-North and altitude yielded r^2=0.87 ($p<0.001$, n=407).

Na was highly correlated to Cl (r^2=0.96, $p<0.001$, n=407), but the observed slope deviated slightly from the Na:Cl ratio in seawater ($p<0.001$). When examining the lakes above an altitude of 500 m separately, the regression (r^2=0.93, $p<0.001$, n=128) yielded a slope not different from the seawater ratio ($p>0.05$). On the latter selection, the non marine sodium (Na*) was estimated to 0.17±0.15 mg/l (n=128). These findings were in accordance with other studies in southern Norway (Wright and Henriksen 1978).

Multiple regression demonstrated that the marine ion contribution, represented by Cl, and the "geologic" ion contribution, represented by Ca, fully explained the conductivity (H^+ adjusted) in the studied lakes (r^2=0.98, $p<0.001$, n=407).

Year	Altitude	n	Sampling date	pH	Cond.	Cond*	Color	Ca	Cl	Na	ALKe
	m		(median)		µS/cm	µS/cm	mg Pt/l	mg/l	mg/l	mg/l	µeq/l
2002	≤500	262	25.07.2002	6.16	38.2	37.4	15	0.96			
2007	≤500	237	12.08.2007	5.99	41.1	40.5	23	0.99			
2012	≤500	279	29.07.2012	6.23	39.9	38.6	22	0.95	7.6	4.3	32
2002	>500	128	12.08.2002	5.64	11.8	10.9	5	0.30			
2007	>500	110	17.08.2007	5.55	12.4	11.1	17	0.27			
2012	>500	128	18.08.2012	5.71	9.4	8.7	5	0.23	1.4	0.91	7
2002	(all)	390	01.08.2002	5.91	26.5	24.2	10	0.60			
2007	(all)	347	15.08.2007	5.79	29.0	27.4	20	0.64			
2012	(all)	407	07.08.2012	6.01	26.4	24.4	17	0.61	4.8	2.8	19

Table 1. Median values (cond = conductivity adjusted for the H^+ contribution).*

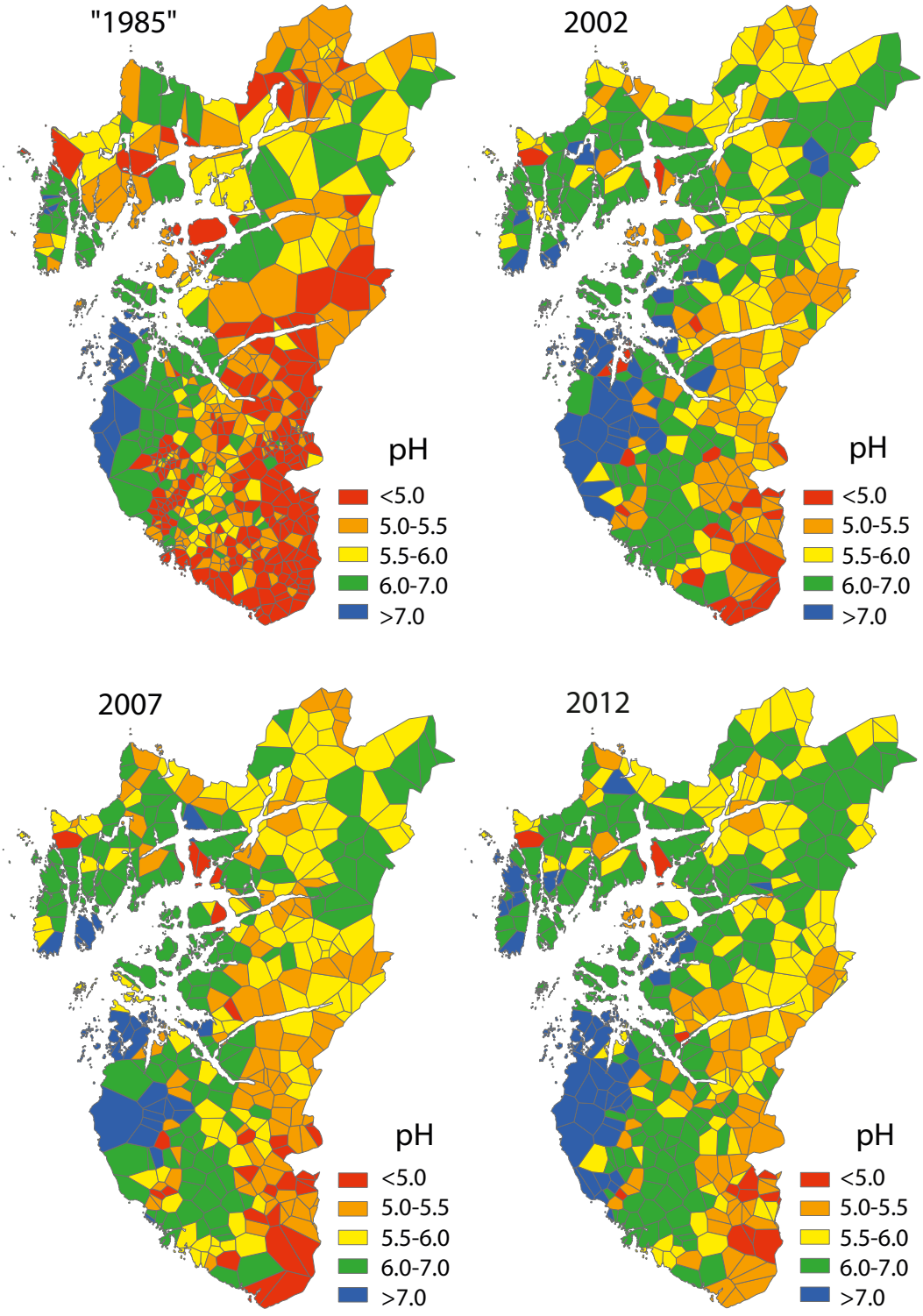

Figure 1. pH in 347-407 lakes in Rogaland in 2002-2012. The map from "1985" is a redraw of a map from the mid 1980s (Enge and Lura 2003), using Thiessen polygons.

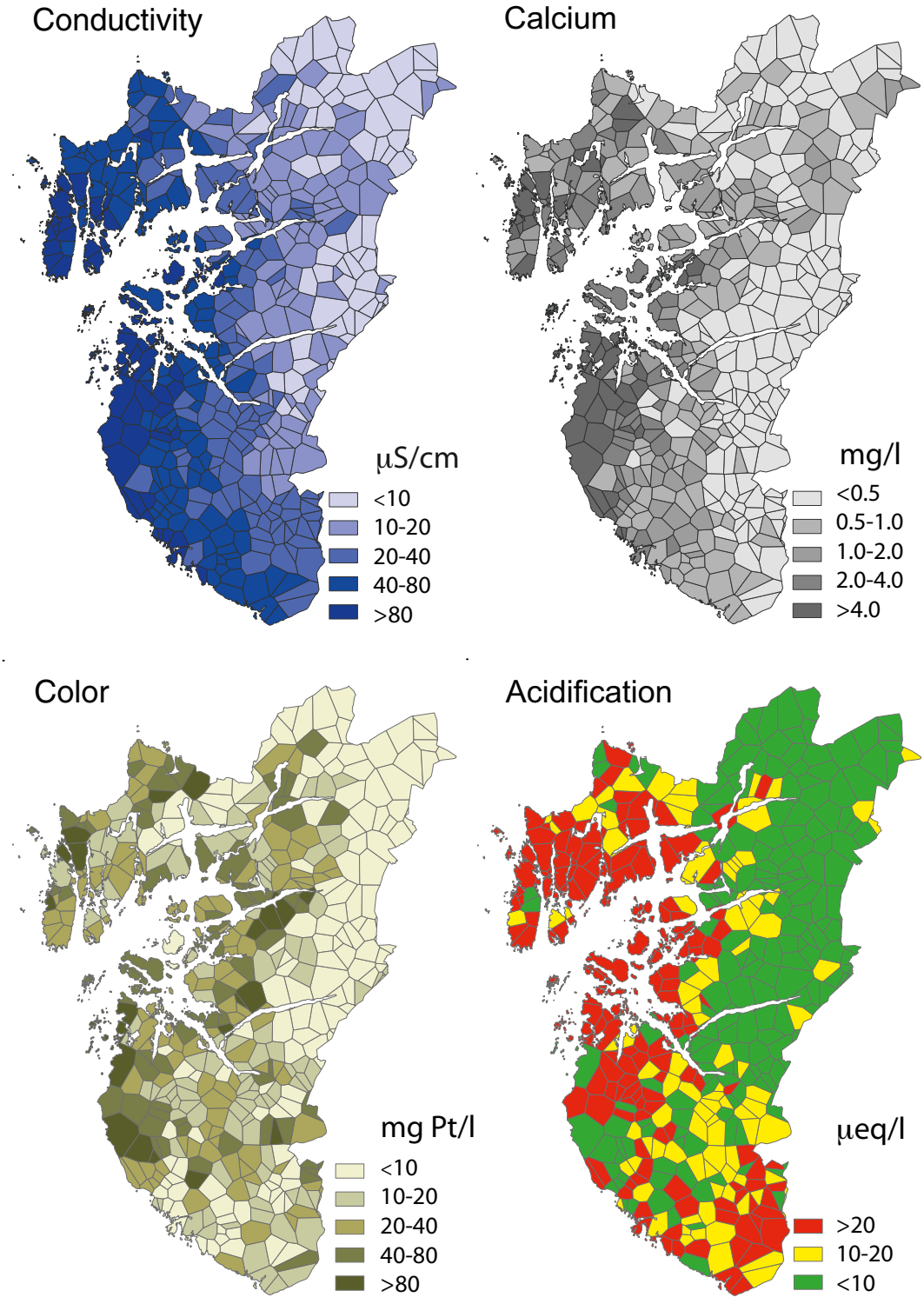

Figure 2. Conductivity, color, calcium and "apparent acidification" (discussed in the text) in 407 lakes in Rogaland in 2012.

When studying the effects separately, Cl correlated somewhat better to conductivity (r^2=0.92) than Ca (r^2=0.82). Calculations based on Cl, suggest that the marine ions represent a conductivity of 19.2 µS/cm in the "median" lake in Rogaland, about 3/4 of the measured conductivity. When examining the lakes above an altitude of 500 m separately (n=128), the conductivity was apparently explained sufficiently by the marine ion contribution, represented by Cl (r^2=0.88). In this selection of lakes, poor correlation between Ca and conductivity was found (r^2=0.10).

Acidification and pre-acidification water chemistry

Presented in a "Henriksen diagram", figure 3, all lakes above an altitude of 500 m and most of the lower altitude lakes were located below the curved line, suggesting none or limited acidification.

The estimated acidification, based on the ALK_0 approach, was limited (17±43 µeq/l, n=407). According to this model, the lakes above an altitude of 500 m were hardly acidified at all (4±7 µeq/l, n=128). Surprisingly, declining acidification along an eastern gradient (UTM-East) was found ($p<0.001$), while there was apparently no effect of the other geographic parameters ($p>0.05$).

The observed pH-values in 2002, 2007 and 2012 (Table 2) were slightly below the estimated pre-acidification pH-values ($p<0.05$), but possibly within the uncertainty of the model. The estimated "F"-factors were 0.17±0.13 (n=32). When separating the lakes by altitude and analyzing all years simultaneously, the lakes above 500 m were apparently not acidified ($pH>pH_0$, $p<0.001$, n=24), while the low altitude lakes were significantly acidified ($p<0.001$, n=72).

Discussion

The first signs of possible acidification became apparent as early as the mid 1800s (Qvenild et al. 2007). Subsequently, an exact determination of acidification and recovery is not possible due to the lack of water chemical references prior to the acidification. Several models have been suggested for calculating "pre-acidification" water chemistry, but the major limitation of all these models is the

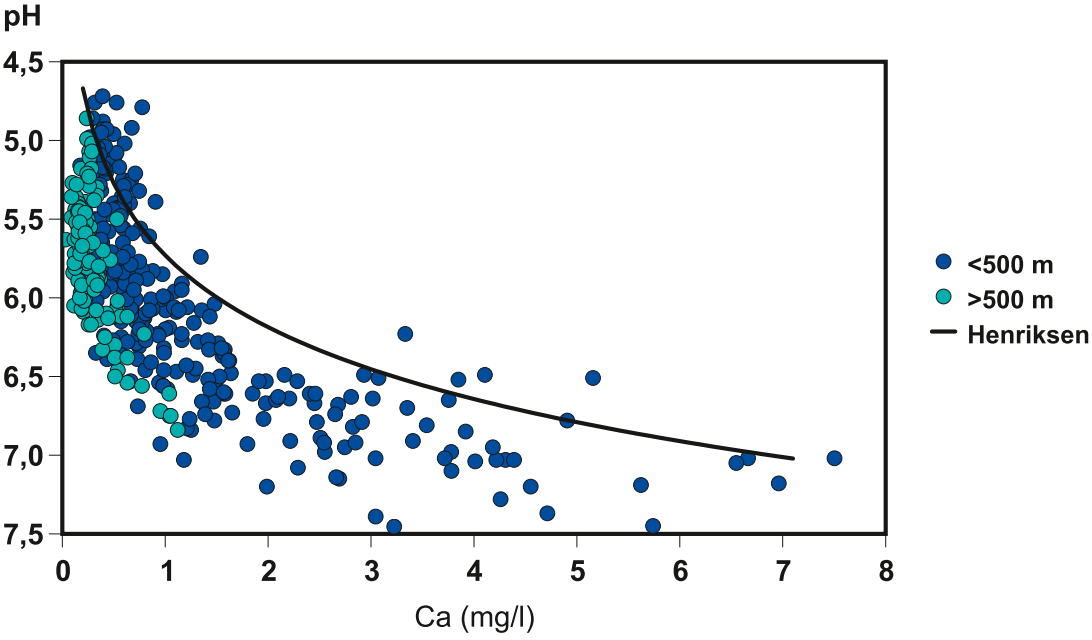

Figure 3. Plot of current pH&Ca values (2012) in the "Henriksen diagram" (18 lakes having Ca beyond the figure upper limit were not shown).

Lake & altitude (m)	pH_0	pH (measured)		
		2002	2007	2012
Erlandsdalsvatn (56)	6.8	7.18	7.00	6.81
Åsvatn (215)	6.8	7.01	6.48	6.79
Gautevatn (964)	5.9	6.15	6.08	6.72
Eikelandsvatn (150)	6.6	6.04	6.44	6.66
Tengesdalsvatn (166)	6.3	6.36	6.30	6.66
Nordvatn (40)	5.8	6.68	6.63	6.61
Natlandv. (293)	6.4	6.42	6.13	6.43
Svinstølvatn (705)	6.0	6.48	6.32	6.30
Liavatn (121)	6.4	6.12	5.88	6.13
Gamlastølsvatn (816)	6.1	6.22	6.01	6.13
Mosvatn (600)	5.9	6.24	5.85	6.10
Vasstølvatn (753)	6.0	5.90	5.94	6.08
Breilandsvatn (197)	6.4	6.42	5.93	6.06
Fossdalsvatn (622)	5.7	5.69	5.88	6.05
Fjellgardsvatn (154)	6.3	6.05	5.82	6.04
Finnabuvatn (898)	5.9	5.91	5.92	5.95
Svåvatn (10)	6.2	5.56	5.74	5.87
Svartavatn (749)	5.7	5.94	5.74	5.66
Giljastølsvatn (406)	5.7	5.44	5.39	5.65
Fundingslandsvatn (342)	5.8	5.91	5.26	5.58
Røyravatn (230)	6.0	5.42	5.42	5.58
Revsvatn (234)	5.7	5.51	5.60	5.44
Kvitlavatn (350)	5.9	5.94	6.08	5.43
Hagavatn (210)	6.1	5.44	5.41	5.32
Spjotevatn (162)	5.9	4.91	4.98	5.10
Sandvatn (194)	5.9	5.01	5.10	5.07
Dypingsvatn (343)	5.9	5.18	5.10	5.05
Fjellavatn (277)	5.8	4.85	4.93	5.04
Homsvatn (142)	5.5	5.00	4.89	5.02
Gjuvatn (389)	5.1	5.10	4.89	5.01
Førlandsvatn (244)	5.5	5.01	5.03	4.96
Holevatn (337)	5.2	4.92	4.95	4.93
Difference ($pH-pH_0$)	Average	-0.16	-0.25	-0.16
	SD	0.44	0.39	0.43

Table 2. Estimated pre-acidification pH-values (pH_0), and observed pH for the years 2002, 2007 and 2012.

lack of documentation of accuracy; i.e. testing against the "real" pre-acidification water chemistry. However, if similar results are obtained by different models, using different data sets, this is a strong indication that these estimates are in the range of the "true" values.

All the three applied models agreed that the present acidification in Rogaland is limited. The acidification indicator by Henriksen (1978) indicated that most of the lakes were not acidified, and both the ALK_0- and pH_0-approaches suggested limited acidification, except for low altitude lakes and lakes in the south-eastern parts of Rogaland, figure 2. Strictly, the estimates of ALK_0 should have been adjusted for possible increased mobilization of Ca due to the acidification (Henriksen 1982b). However, at the current low levels of sulfate in mountain areas of Rogaland (Enge 2012), and relatively low "F"-factors, the ALK_0 adjustment becomes negligible.

The "acidification map", figure 2, based on the ALK_0 approach, suggests that the lakes along the Ryfylke "basin" and Jæren were apparently subject to extensive acidification. It is possible, at least for some of the lakes, that this apparent acidification was caused by limitations in the relatively simple model applied. The model does not consider the dynamic properties of the acidification recovery and the estimates of ALK_0 were based on Ca^* only. Additionally, the applied background acidification (Henriksen 1978) is provided for granitic bedrock while the low altitude geology in Rogaland also includes metamorphosed rocks. This apparent acidification has, however, limited significance due to the relatively high alkalinity associated with the low altitude lakes.

The acidification recovery during the last 2-3 decades was considerable, figure 1. In the mid 1980s, the pH values of the lakes in south-eastern parts of Rogaland were generally <5.0, while the pH ranges of 5.0-5.5 and even 5.5-6.0 were most frequent throughout the 2000s. Only small changes in pH values were registered throughout the 2000s, suggesting that the acidification recovery has slowed down throughout the last decade. This observation is supported by other recent studies too (Skjelkvåle 2011).

The limited water chemical variations observed throughout the 2000s were probably caused by direct and/or indirect effects of meteorological conditions. The very low conductivity values in the mountain lakes (>500 m) in 2012 (3/4 of the 2002- and 2007-values), were probably caused by dilution effects due to large snow accumulation and prolonged snowmelt. The precipitation during the first 6 months of 2012 was 122% (Lysebotn) and 135% (Nesflaten), referred to the normal period 1961-1990 (eKlima.no 2013). Slightly higher conductivity in 2007 than in 2002 was possibly caused by generally higher sea salt deposition (Conductivity vs. Cl: r^2=0.92).

The mountain lakes (>500 m) had generally very low levels of color (organic matter), suggesting that a large fraction of the Al is uncomplexed (toxic). However, other studies have established that the Al values in these areas are relatively low, and generally below toxic limit for brown trout (Enge and Kroglund 2010).

An apparent discrepancy between chemical recovery and biological recovery is currently arising: The present study establishes that the water chemistry already has responded considerably to reduced deposition of acid components, while other studies have suggested that the biological recovery still is restricted by unsuitable water chemistry (Skjelkvåle 2011). Thus, we cannot reject the possibility of the existence of areas having marginal water quality even without any acidification.

In very dilute lakes with TOC<2 mg/l, Wright and Cosby (2012) suggested pre-acidification pH and ANC values as low as 5.0 and -10 µeq/l respectively, possibly detrimental to brown trout (Lien et al. 1996, Sevaldrud and Muniz 1980). In many humic lakes, pre-acidification pH may have been <5.0 (Hindar 2011, Hindar and Wright 2002). Due to poor geology and limited marine ion contribution, many high altitude lakes in Rogaland have extremely low ionic strength. Median conductivity (H^+ adjusted) and Ca for the lakes located above an altitude of 500 m (n=128) was 8.7 µS/cm and 0.23 mg/l

respectively (2012-data). The corresponding minimum values were 3.1 µS/cm and <0.05 mg/l. Enge and Kroglund (2010) suggested that conductivity <5 µS/cm is detrimental to early life stages of brown trout, and Dale et al. (1975) found 3 µS/cm to be lethal for trout fry. Wathne and Rosseland (2000) suggested that Ca>0.38 mg/l is required to support healthy trout populations in mountain lakes. In Lake Vetratjørn, one of the most dilute lakes in the current study (Conductivity=3.7 µS/cm, Ca=0.12 mg/l), earlier attempts of stocking brown trout have failed (Berg 1972).

Thus, we cannot reject the possibility of extremely dilute water as an important restrictor for brown trout in mountain lakes in these parts of Norway. This effect has probably existed in the past too, but has been less apparent during the acidification period.

Conclusion

The water chemistry of the lakes in Rogaland has recovered considerably during the last 2-3 decades, and is currently close to "pre-acidification" conditions. Data from 2002, 2007 and 2012 suggested that the observed water chemical variations in the lakes during the last decade were natural variations, directly or indirectly caused by meteorological conditions. The water quality in many mountain lakes in Rogaland is extremely dilute and is probably restricting the distribution of brown trout.

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