When soft waters becomes softer; drivers of critically low levels of Ca in Norwegian lakes

D. O. Hessen,[1]* T. Andersen,[1] K. Tominaga,[1] A. G. Finstad[2,3]
[1]Department of Biosciences, Aquatic Ecology and Toxicology (AQUA), University of Oslo, Oslo, Norway
[2]Department of Natural History, NTNU University Museum, Norwegian University of Science and Technology, Trondheim, Norway
[3]Norwegian Institute for Nature Research, Trondheim, Norway

Abstract

In this study, we analyzed long-term trends in calcium (Ca) concentrations for 70 Norwegian lakes spanning a broad geographical range and for a wide range of ambient drivers. A striking decline in Ca concentrations was observed during the past 30 yr. The trend was most pronounced in southern, previously acidified localities, while there were weaker and less consistent patterns for central and northern regions. For previously acidified areas, this observed decline in Ca is likely related to base cation depletion or reduced Ca mobilization as a consequence of reduced H_2SO_4-deposition. However, lower chloride concentrations, increased runoff and forest density (represented by Normalized Difference Vegetation Index, NDVI) also exerted strong negative impacts on Ca-concentrations. For lakes with a significant negative trend ($n = 34$), the average decline was -11.05% per decade, corresponding to -0.023 mg Ca L^{-1} yr^{-1}. This trend may pull levels of Ca in these already Ca-deficient poor lakes toward, or below, critically low levels for a range of organisms. A somewhat paradoxical effect of the successful battle against anthropogenic acidification is thus that it may impose an increased Ca-deficiency of freshwater ecosystems.

A somewhat paradoxical response to recovery from acidification is a striking decline of base cations, notably Ca and Mg, following decreased H_2SO_4-deposition (hereafter S-deposition) in previously acidified areas (Likens et al. 1996; Jeziorski et al. 2008; Futter et al. 2014). The downward trend for Ca and Mg in rivers and lakes in these areas primarily reflects the combined effect of reduced mobilization and depletion of base cation reservoirs in watersheds since export rates exceed weathering rates (Likens 1996; Likens and Buso 2012; Futter et al. 2014). This decline has received much attention as a potential threat to the biota of Canadian and North-American lakes, but the trend is also prominent in Europe, notably over southern Scandinavia and UK as assessed from the ICP Waters program (Garmo et al. 2014).

Adding to the post-acidification effects, land use changes and repeated logging and timber removal could also contribute

to depletion of Ca from catchments (Likens et al. 1966; Watmough and Dillon 2003; Akselsson et al. 2007). On the other hand, vegetation and root activity may boost weathering (Beerling and Berner 2005; Archer 2010). For catchments with low human impacts, weathering is the prime source of key elements like calcium (Ca), silicate (Si) and phosphorus (P) (Humborg et al. 2014). CO_2 reacting with silicate minerals or apatite yields Ca, Si, and P in various forms and proportions (Archer 2010). While increased runoff could increase the annual export flux of Ca and other elements from catchment, it will also cause a dilution, yielding lower concentrations in surface waters.

Ca is an essential element to invertebrates with a calcified exoskeleton as well as for bony structures in vertebrates. Ca deficiency in soft-water localities may be a major determinant to distribution and growth of gastropods (Økland 1990), benthic crustaceans (Alstad et al. 1999), and likely other invertebrates. There is also evidence that Ca deficiency could act as an important determinant of zooplankton community structure (Tessier and Horwitz 1990; Hessen et al. 1995). There are also striking differences in Ca-content and Ca-requirements of various species (Wærvågen et al. 2002), such that Ca-concentrations may be an important driver of community structure (Jeziorski et al. 2008; Cairns and Yan 2009).

*Correspondence: d.o.hessen@ibv.uio.no

mean Ca

delta Ca

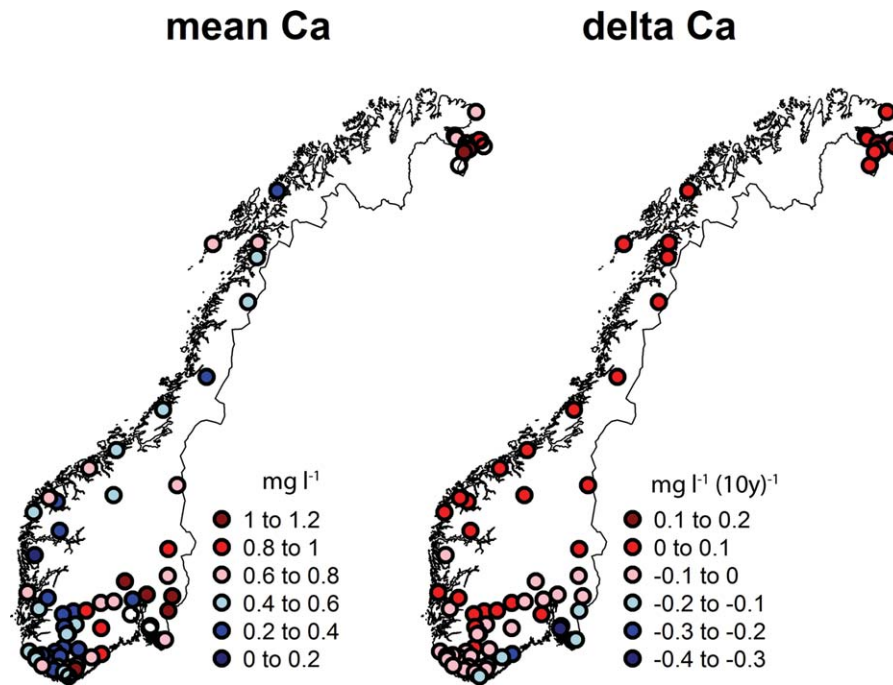**Fig. 1.** Mean Ca (mg L^{-1}) during the study period (1986–2013 and absolute change in Ca per decade (mg L^{-1} 10 yr^{-1}).

The potential role of Ca deficiency will surely be most relevant for soft-water localities. Such lakes may show a scattered distribution depending on local geology, but are particularly abundant in geographical areas like the Canadian Shield (Neary and Dillon 1988) and major parts of Scandinavia. A national survey of 1500 Norwegian lakes revealed that median Ca concentration was 0.9 mg Ca L^{-1}, 90% of the localities had less than 2.5 mg Ca L^{-1} and almost no localities exceeded 5 mg Ca L^{-1} (Skjelkvåle et al. 2001). Ca-deficiency is thus likely already an important stressor and determinant of community structure and productivity, while any further reduction of Ca will pose additional constraints to the biota of these soft-water systems.

To assess the spatial trends in Ca-concentrations and the drivers towards lower Ca, we analyzed a 30-yr time series for 70 Norwegian lakes for which there has been regular, annual monitoring of post-acidification recovery. These lakes span a wide latitudinal and longitudinal gradient and also cover a tenfold variation in past and present S (and N)–deposition in their respective catchments. For all catchments we compiled time-series of vegetation density (as the Normalized Difference Vegetation Index, NDVI), as well as runoff and temperature in addition to the major anions chloride and sulphate (both S-deposition and S-concentration).

Materials and methods

The data in this analysis were obtained from a Norwegian lake monitoring program from 70 lakes covering the mainland Norway that have annual samples from 1986 to 2013 (Fig 1). These lakes are a subset of the 1000-lake acidification survey of 1995 (Henriksen et al. 1998), and represent acid sensitive, headwater lakes on granitic or gneissic bedrock with negligible local pollution sources. Water samples are collected annually at the outlet after the autumn circulation period and analyzed at the Norwegian Institute of Water Research (NIVA) (Skjelkvåle et al. 2001). The data are available from http://vannmiljo.miljodirektoratet.no.

We first conducted an analysis of temporal trends in the time series of Ca concentrations in these lakes, using a regional Kendall trend test where each individual lake were used as a block (Helsel and Frans 2006). The analysis was run using the rkt library (Marchetto 2015) in R v. 3.2.1 (R Core Team 2015).

Lake catchments area were delineated from digital maps (*see* Finstad et al. 2014; Finstad and Hein 2012). Time series of annual mean catchment vegetation (NDVI), runoff, temperature, and S-deposition were extracted using the raster library (Hijmans 2015) in R v. 3.2.1 (R Core Team 2015).

Catchment vegetation density was represented by the Normalized Difference Vegetation Index (NDVI; GIMMS NDVI3g) (Fensholt and Proud 2012). The NDVI represents a radiometric measurement of the fraction of photosynthetically active radiation ($\sim$400 to 700 nm) absorbed by chlorophyll in a vegetation canopy (Myneni et al. 1995). While changes in NDVI cannot directly be assessed as changes in net primary production, it is a good proxy of changes in photosynthetic activity and vegetation density (Myneni

et al. 1995; Forkel et al. 2013). The NDVI3g data set has a pixel resolution of 8 x 8 km. The maximum NDVI value over a 15-day period is used to represent each 15-day interval to minimize bias due to cloud contamination and variations in atmospheric turbidity, scan angle, and solar zenith angle (Holben 1986). This provides two maximum-value NDVI composites per month.

Temperature and runoff time-series for individual catchments were obtained using gridded data from 1980 to 2013 (1 km^2 resolution from the Norwegian Meteorological Institute and the Norwegian Water Resources and Energy Directorate (Mohr 2008)). S-deposition at the catchment level was interpolated from EMEP deposition raster temporal composite (1980, 1985, 1990, and annually from 1995 to 2011) (Schulz et al. 2013), using recalculations if available. In the present study we used total S-deposition, comprising both dry and wet deposition. Each type of deposition was modelled using regression kriging with EMEP precipitation as a linear regression model predictor, and with residuals fitted using a linear variogram model with no nugget. Using the variogram model and linear model, deposition flux over the catchment area was predicted using block kriging with the target geometry being the catchment polygon. Missing values from 1985 to 1990 and 1990 to 1995 were estimated within each catchment using loess regression.

The catchment time-series occurring on daily (temperature and precipitation) and bi-monthly (NDVI) scale were collapsed to yearly means using June to August averages, and standardized to zero mean and unit standard deviation prior to the analyses. Other aggregates of NDVI, such as maximum yearly NDVI and June–August maxima gave similar results in terms of temporal dynamics, and were therefore not included in further analyses. Theil-Sen's slopes for individual lake and catchment variables, estimated by the rkt library (Marchetto 2015) in R v. 3.2.1 (R Core Team 2015) were used for visualizing spatial trend patterns on maps.

The statistical analyses were conducted using linear mixed effect models, fitted using the nlme library (Pinheiro et al. 2012). We used log transformed Ca as dependent variable and catchment drivers (S-deposition, catchment NDVI, runoff, and air temperature) as explanatory variables. In addition, year was entered as a controlling variable (Freckleton 2002) to account for spurious correlations with unobserved environmental drivers.

Judgment of cross correlation function between the time-lags of different catchment variables and Ca revealed that the best correlation between NDVI, S-deposition, and Ca measured in lake water involved a time lag (4 and 5 yr, respectively). Hence, we used 4 and 5 yr lagged values of NDVI and S-deposition as input to the models. However, rerunning the analyses using different time lags confirmed that the major model predictions were robust with regard to the particular time lags used in the analyses.

The full model had a random structure with random intercept and slopes nested within lake. In addition, an AR1 autocorrelation structure was added, nested within lake. The random intercept effect represents between-lake variation in baseline Ca load caused by static catchment variables that not expected to change on the investigated time scale (e.g., geology). Model selection was based on comparing models using AIC (following the protocol in Zuur et al. 2009). We first selected the random structure by comparing models containing all explanatory variables as fixed structure, but with different random structure: M1; without random structure, M2; with random intercept (lake); M3: with random intercept (lake) and random slope (year) nested within lake; M4: with random intercept (lake) and AR1 autocorrelation (year) nested within lake; M5: with random intercept (lake), random slope (year) nested within lake, and AR1 autocorrelation (year) nested within lake.

A random intercept with random slope (year) nested within lake (model M3) provided the best fit to the model (ΔAIC 186.64 compared with next best model) and were used for selecting the fixed structure of the model. Selection of fixed structure were done by model comparison, re-running the model using the selected random structure and compare models using AIC weights with the MuMin library (Barton 2015). While the selection of random structure used the robust but biased REML (restricted maximum likelihood) method, the fixed effect model selection used re-fitting by the less biased ML (maximum likelihood) method (Zuur et al. 2009). Residuals from the final model did not reveal any deviations of normality or heteroscedasticity, nor autocorrelations.

As an example of how the declining levels of Ca potentially could affect crustaceans, we use established thresholds for survival of *Daphnia magna* and *D. pulex* (0.5–1.0 mg Ca L^{-1}) based on both laboratory experiments and field studies (Hessen et al. 2000; Jeziorski et al. 2015; Prater et al. 2015).

Results

A pronounced decline in Ca was observed in most southern localities, while there were weaker and less consistent patterns for central and northern regions (Fig. 1). For lakes with a significant negative trend (at 5% level, $n = 34$), the average decline was −11% per decade, which in absolute terms implied an average decline of −0.023 mg Ca L^{-1} yr^{-1}, yet with a pronounced variability between localities.

There was a close spatial accordance between trends in Ca concentration and S-deposition, especially in the southern regions most impacted by acid deposition (Fig. 2), but decreases in Cl and increases in NDVI were also accompanied with reduced levels of Ca (Fig. 2). A model containing main effects of all predictor variables provided the best fit to the data with Akaike weights at 0.83 (representing the relative likelihood) in the full model comparison set (Table 1).
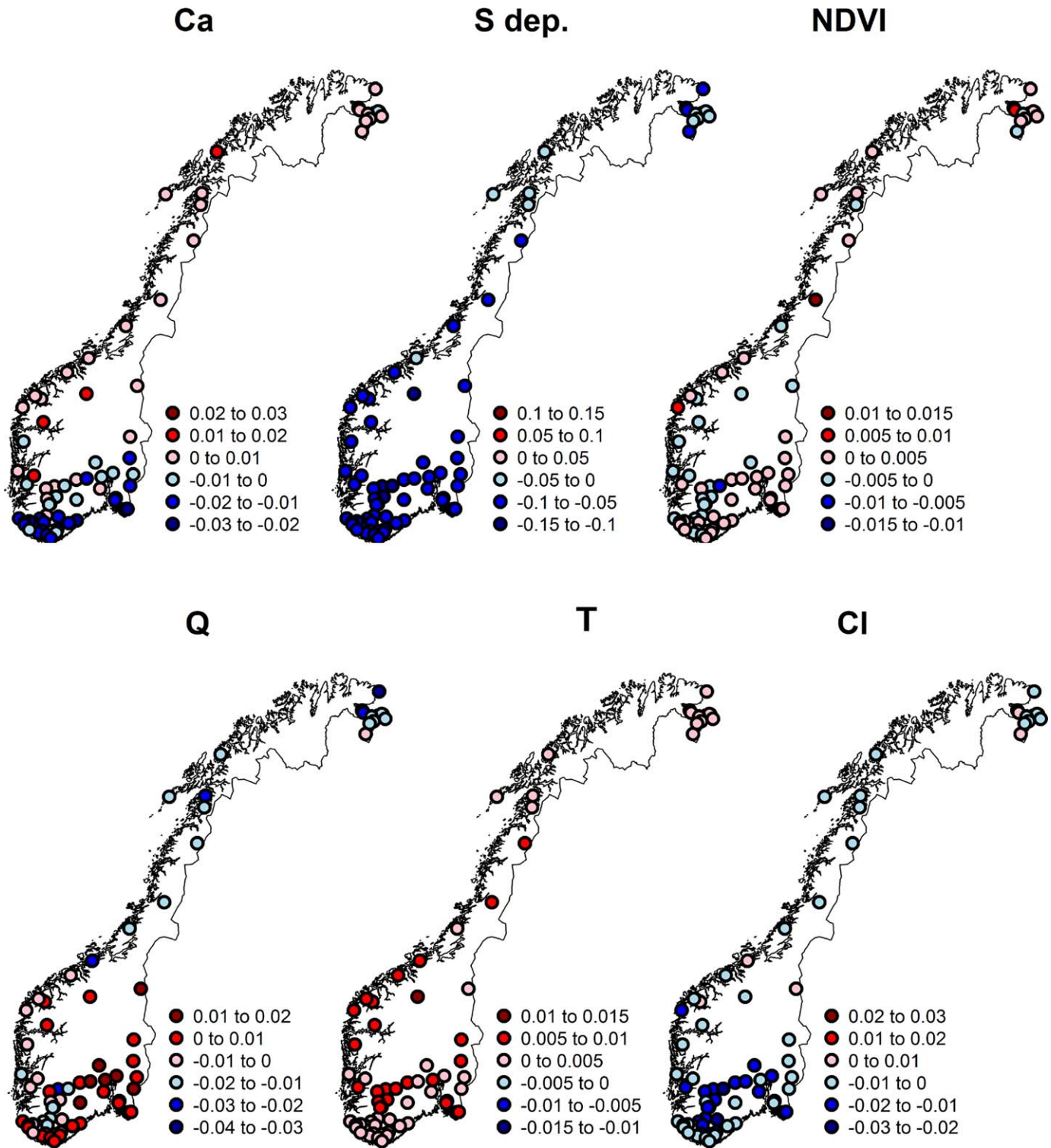
Fig. 2. Time trends (parametric slope estimates) for calcium concentrations (Ca), sulphur deposition (S-dep), Normalized Difference Vegetation Index (NDVI), runoff (Q), air temperature (T), and chloride concentration (Cl). Ca and Cl measured in water samples, and S-dep, NDVI, T and Q in catchments.

Judged by the standardized coefficients from the best model (Table 2) the most important predictors of Ca-concentration were runoff, S-deposition, NDVI and temperature, respectively, with negative contributions from NDVI (likely attributed to forest uptake), runoff (dilution), and positive contributions from S-deposition and temperature. Note that

Table 1. Model selection tables of Ca against NDVI, run-off (Q), S-deposition, temperature and year.

Intercept	Cl	NDVI	Q	S-dep.	Temp	Year	df	LogLik	AIC	Delta	Weight
−0.01	0.31	−0.06	−0.17	0.06	0.05		10	−15.15	50.31	0.00	0.65
−0.03	0.31	−0.06	−0.17	0.06	0.06	0.03	11	−14.92	51.84	1.54	0.30
−0.15	0.32	−0.06	−0.17		0.05	−0.06	10	−18.24	56.48	6.17	0.03
−0.14	0.32	−0.07	−0.16		0.05		9	−19.59	57.18	6.87	0.02
−0.01	0.30		−0.17	0.07	0.05		9	−22.43	62.85	12.55	0.00
−0.01	0.30	−0.06	−0.17	0.05			9	−22.89	63.77	13.47	0.00
0.00	0.30		−0.17	0.08	0.05	0.03	10	−22.18	64.37	14.06	0.00
0.00	0.30	−0.06	−0.17	0.07		0.03	10	−22.34	64.69	14.38	0.00

The tables show parameter estimates for model terms included in the models, log likelihood (LogLik), AIC, AIC difference from best model (delta), and Akaike weights (weights). Top eight models displayed.

Table 2. Summary result for best model (*see* Table 1) for Ca against NDVI, run-off (Q), S-deposition, chloride, temperature, and year.

	Estimate	Sd. error	df	t	p
Intercept	−0.01	0.10	1609	−0.04	0.963
NDVI	−0.06	0.02	1609	−3.84	<0.001
Q	−0.16	0.02	1609	−9.26	<0.001
S-dep.	0.06	0.02	1609	3.29	0.001
Temp.	0.05	0.01	1609	3.35	<0.001
Cl	0.31	0.02	1609	16.39	<0.001

the positive contribution from S-deposition implies reduced Ca with reduced S-deposition.

There were strong spatial patterns with most prominent effects in the southern regions, and also the increasing trend in NDVI is most prominent for the southern localities (Fig. 2). Also runoff turned out to be a significant negative contributor to Ca concentrations. However, while there are trends of increasing runoff, the spatial pattern is by far overriding the temporal trends, with a striking longitudinal difference with higher runoff at the coastal sites (Fig 2). Runoff serves a dual role by both mediating Ca export fluxes as well as diluting Ca concentrations. The negative net effect of runoff demonstrates an overriding impact of dilution. Finally, also temperature was a significant predictor (positive) of Ca.

The smoothed, temporal trajectories of Ca in relation to S-deposition, runoff, temperature and NDVI are given in Figure 3. For S-deposition, there was a striking decrease at all sites, but the relative effect size on Ca was modest, and only prominent for lakes with initially high S-deposition (i.e., the southern localities most affected by acidification). For some of the low Ca localities, there was in fact an increase. The overall negative relationship between runoff and Ca is quite evident (Fig 3, upper right panel), while a strong trend is seen primarily in sites with low runoff. For temperature (Fig. 3, lower left panel), there was a somewhat similar overall

positive relation, but with the strongest trends is seen in the high temperature range (i.e., southern localities). Some of the low temperature sites (mostly high latitude or altitude) had an opposite trend. For NDVI, the increase is quite consistent all over, while the downward trend in Ca is again most prominent in southern sites (cf. Fig. 2).

The biotic responses to declining Ca will differ between taxa and probably also depend on local adaptations. With the proviso that it is impossible to arrive at exact thresholds for optimal performance or extinction in natural systems due to a wide range of confounding factors, tentative critical thresholds of Ca-concentrations for the crustaceans *D. magna* and *D. pulex* can nevertheless be used an example of the potential Ca-limited distribution of such Ca-demanding species. These species should perform poorly in the majority of Norwegian lakes and be unable to establish in the major fraction of western lakes (Fig 4). Also, the lower threshold has been reached in almost 10% of the surveyed localities over the past 3 decades whether using 0.5 or 1.0 mg Ca L^{-1} as the lower bound for existence (Fig. 4).

Discussion

Our study confirms a striking decline in Ca-concentrations in the lakes that has been most affected by high S-deposition. This trend has been explicitly linked to depletion in base cations in Canadian and North-American (Keller et al. 2001; Jeziorski et al. 2008, 2015; Likens and Buso 2012) as well as European rivers and lakes (Futter et al. 2014; Garmo et al. 2014). Also, concentrations and trends for Cl displayed strong regional patterns, but generally with negative trends in the coastal and southern sites (cf. also Skjelkvåle et al. 2007), and were associated with reduced Ca. The apparently strong impact from Cl was somewhat surprising, although it can be attributed an ion-balance effect, where Cl$^-$ serves as a mobile anion affecting the catchment export of base cations (Wiklander 1975). While Cl did not contribute to Ca mobility at Hubbard Brook, this was accredited to its low contribution of strong anions in this system (9%) (Likens et al. 1998). In the oceanic influenced Norwegian catchments it can
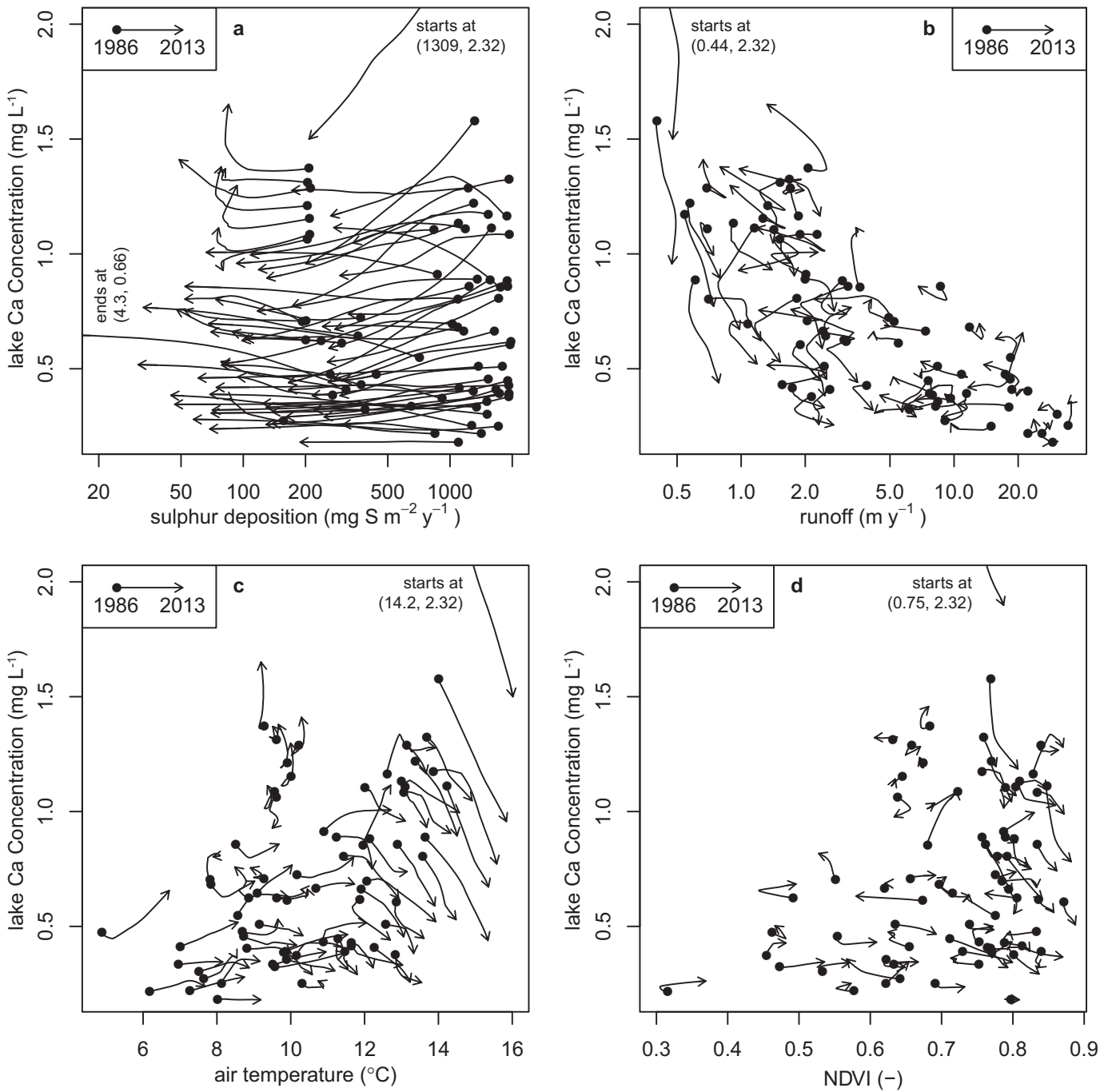
Fig. 3. Smoothed time trend trajectories for annuals means of sulphur deposition (**a**), runoff (**b**), air temperature (**c**) and NDVI (**d**) vs lake concentrations of calcium over the period 1986–2013. The two-dimensional cross-smoothed time-series curves (solid lines) starts 1986 (dots) and ends 2013 (arrow heads) and are given for individual lakes. Coordinates were obtained by smoothing lake-specific time-series using lowess (R Core Team 2015) with a smooth span of 0.8. Only years with a complete pair of observations are used in this figure.

make a substantial contribution, and especially during "seasalt episodes" (Skjelkvåle et al. 2007). While NO_3 clearly also may contribute to Ca dynamics in catchments, it was not included in our analysis due to its complete spatial overlap with S-deposition, and the absence of significant time-trends over this 30-yr period (Garmo et al. 2014).

Time-series of satellite-derived NDVI serve as a good proxy of vegetation changes that may affect Ca. Our results support that forest cover and forestry may contribute to the observed trends in Ca in addition to the negative cation balance following initial mobilization by H_2SO_4-promoted weathering and subsequent post-acidification cation
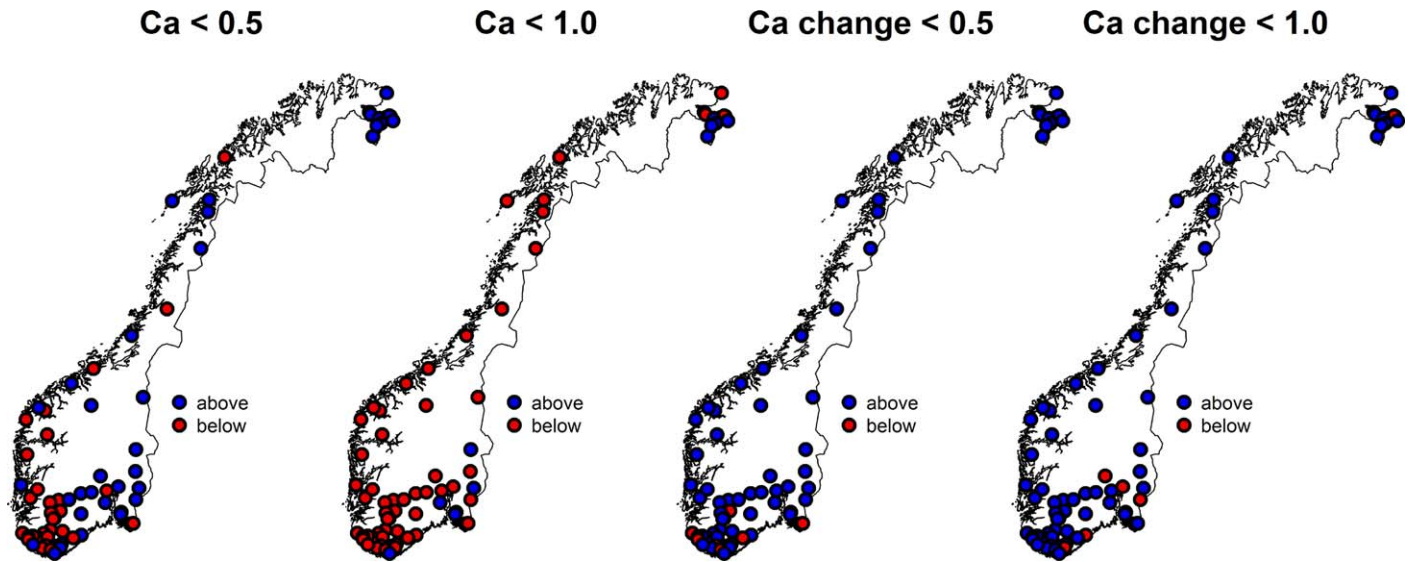
Fig. 4. Lakes that in 2013 are below the tentative critical range of Ca based on published data for *D. magna* and *D. pulex* (red symbols: <0.5 mg L^{-1}, $N = 29$ and <1 mg L^{-1}, $N = 57$) and lakes that during the study period 1986–2013 has entered the critical levels (<0.5 mg L^{-1}, $N = 7$ and <1 mg L^{-1}, $N = 8$).

depletion (Likens and Buso 2012). Logging practices and timber removal deplete Ca from catchments, simply due to Ca in plant tissue being removed from the catchment (Likens et al. 1998; Watmough and Dillon 2003; Akselsson et al. 2007). Vegetation also promotes weathering (Beerling and Berner 2005), however, the net balance between uptake and root weathering and efflux of standing forest may depend on several factorsin addtion to net forest growth. Our analysis did suggest a net negative impact of forest density (as inferred from NDVI), which might reflect the recent strong increase in net forest biomass. Increased forest volume will by itself imply an increased demand and storage of Ca in forest biomass, and reduced catchment export. Only over the past decade there has been a fully 26% increase in forest volume (http://www.ssb.no/jord-skog-jakt-og-fiskeri/statistikker/lst/aar/2015-08-27) in Norway, mainly as a consequence of past logging practices and recent afforestation. An increased biomass of standing forest has occurred over most of Europe since the 1950s (Gold et al. 2006). While this increase constitutes an important sink of atmospheric CO_2, it is also a sink of Ca and other elements. For some regions, notably alpine and subalpine areas, climate change and reduced grazing will continue to increase forest cover and elevate treelines (Bryn et al. 2013).

Runoff was identified as a major predictor of Ca change in our analyses. This likely illustrates the net negative effect caused by dilution. It is important to stress that the spatial distribution in runoff strongly adds to the temporal patterns in the mixed model output, i.e. low Ca concentrations in south-western areas could to some extent be attributed the high precipitation in these areas, causing a dilution of Ca.

For Fennoscandia, a general increase in precipitation has been seen over the past few decades, with a further projected increase for many areas (Wilson et al. 2010). This will likely pose further stress on Ca-deprived biota.

The spatial patterns of N-deposition (with almost equal contributions from NO_3 and NH_4) closely match those of SO_4-deposition, where southern regions have experienced a strong increase over past decades and with a more than 20-fold span in deposition (Mylona 2002; Tørseth et al. 2012; Vet et al. 2014). Since the peak around 1980, there have been only modest reductions in N-deposition, while S-deposition has declined by 70–90% (Tørseth et al. 2012; Vet et al. 2014), and in southern regions the contribution to catchment acidity from N is becoming increasingly important (Henriksen et al. 1997). This means that N-deposition plays a twofold role, both by maintaining catchment acidity and hence likely counteracting the impacts of reduced S-deposition on Ca, and by promoting forest growth. Hence not only the decline in S-deposition, but hydrology and forest volume and logging practices will have profound effects on future levels of Ca in soft water areas.

The biotic responses to low and declining levels of Ca is likely to be profound, yet not really scrutinized in Nordic areas. Since the areas with the strongest decline are also in the process of post-acidification recovery, it is difficult to disentangle the effects of reduced deposition of strong acids and increased pH with the impacts of decreased base cations. The same regions also show a pronounced increased in the export and concentrations of dissolved organic carbon ("browning"), partly driven by the same factors (Monteith et al. 2007; Weyhenmeyer and Karlsson 2009; Larsen et al.

2011). This browning is contributing organic acidity to surface water as well as reducing productivity at all trophic levels simply due to increased light attenuation and reduced visibility (Karlsson et al. 2009; Thrane et al. 2014). Ca-depletion will pose an additional stress to the biota of these systems.

Many organisms, and especially invertebrates with a calcified exoskeleton, will to some extent likely suffer from Ca-limitation in soft waters. This is most strikingly seen in gastropods where the diversity is strictly coupled to levels of Ca (and Mg) (Økland 1990), but also insect larvae, crustaceans and other taxa are may be affected either in terms of reduced fitness or local extinctions. Different susceptibility and optima will also promote distinctive community changes like the observed "jellification" in Canadian lakes where Ca-demanding *Daphnia* are replaced by *Holopedium* (Jeziorski et al. 2015), reflecting a strikingly different Ca demand among different species (Hessen et al. 1995; Wærvågen et al. 2002).

For most organisms there will be a wide gap between Ca optima and critically low levels, e.g. laboratory studies with *D. magna*, suggest that this species has a lower threshold of 0.5–1.0 mg Ca for survival, and that growth in juveniles, which have the highest specific Ca requirements, may be sub-saturated <5 mg Ca L^{-1} (Alstad et al. 1999; Hessen et al. 2000), which would comprise 95% of Norwegian lakes. This corresponds well with the observed responses for *D. pulex* (Prater et al. 2015). By using these species as examples of potential impacts of low—and declining—levels of Ca, it is evident that the effects even over a decadal time-span may be profound. As real indicators, these species are not well suited however, since their distribution will be more restricted by predatory fish and other water quality parameters.

Other zooplankton species may have lower thresholds as judged from their specific Ca content (Wærvågen et al. 2002), while a number of benthic invertebrates no doubt have much higher demands. Edwards et al. (2009) observed reduced Ca-acquisition in the crayfish *Orconectes virilis* <8 mg Ca L^{-1}. For the crayfish *Astacus astacus* and the amphipod *Gammarus loricatus*, Rukke (2002) reported reduced growth and survival <10 mg L^{-1} and molting failure <5 mg L^{-1}. This means that many organisms may be naturally Ca-deficient in these soft water lakes, and the present communities reflect some "ghost of Ca-deficiency past" (cf. Connell 1980) by being devoid of Ca-demanding species.

Also nutritional status, growth rate, ontogeny, local adaptations, temperature, pH, UV-radiation, and a suite of other ambient parameters could affect the lower bound of Ca-tolerance (Hessen and Rukke 2000; Ashforth and Yan 2008; Prater et al. 2015). Exact limits can therefore not be given. Reduced calcification is typically a result of suboptimal Ca-concentrations, and this may increase predation risk (Jeziorski and Yan 2006; Riessen et al. 2012), as well as posing a change in size structure (towards smaller individuals). A correlation between body size and specific Ca contents found, e.g. in

Daphnia species (Wærvågen et al. 2002) suggest that large-bodied species are replaced by smaller ones in soft water localities because of reduced fitness at low ambient Ca (cf. Tessier and Horwitz 1990; Hessen et al. 1995).

It is a somewhat paradoxical effect of the successful battle against anthropogenic acidification that it may impose an increased Ca-deficiency of freshwater ecosystems. This effect can become even more severe due to changes in hydrology, forest cover and climate. Given the potential widespread consequences of this for vast regions with low ionic content of surface waters, it is indeed a field that deserved increased attention both with regard to drivers and consequences.

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Acknowledgments

Conflict of Interest

None declared.