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LETTER

From legacy effects of acid deposition in boreal streams to future environmental threats

Hjalmar Laudon[1], **Ryan A Sponseller**[2] **and Kevin Bishop**[3]

[1] Department of Forest Ecology and Management, Swedish University of Agricultural Sciences, Umeå SE-901 83, Sweden
[2] Department of Ecology and Environmental Sciences, Umeå University, Umeå 901 87, Sweden
[3] Department of Aquatic Sciences and Assessment, Swedish University of Agricultural Sciences, Uppsala SE-750 07, Sweden

E-mail: Hjalmar.Laudon@slu.se

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Abstract

Few environmental issues have resulted in such a heated policy-science controversy in Sweden as the 1990s acidification debate in the north of the country. The belief that exceptionally high stream acidity levels during hydrological events was caused by anthropogenic deposition resulted in a governmentally funded, multi-million dollar surface-water liming program. This program was heavily criticized by a large part of the scientific community arguing that the acidity of northern streams was primarily caused by naturally occurring organic acids. Here, we revisit the acid deposition legacy in northern Sweden two decades after the culmination of the controversy by examining the long-term water chemistry trends in the Svartberget/Krycklan research catchment that became a nexus for the Swedish debate. In this reference stream, trends in acidic episodes do show a modest recovery that matches declines in acid deposition to pre-industrial levels, although stream acidity continues to be overwhelmingly driven by organic acidity. Yet there are legacies of acid deposition related to calcium losses from soils, which are more pronounced than anticipated. Finally, assessment of these trends are becoming increasingly complicated by new changes and threats to water resources that must be recognized to avoid unnecessary, expensive, and potentially counterproductive measures to adapt and mitigate human influences. Here we make the argument that while the acidification era is ending, climate change, land-use transitions, and long-range transport of other contaminants warrant close monitoring in the decades to come.

1. Introduction

Recognizing the damage caused by acid deposition, and then dramatically reducing the atmospheric emissions that caused this problem stands as one of the great success stories in evidence-based environmental policy (Grennfelt *et al* 2020). Separating natural processes from the effects of acid deposition was key to this achievement. Several important studies showing the role of anthropogenic acidification appeared to have resolved these issues by 1990, partly thanks to the National Acid Precipitation Assessment Program in the U.S. (NAPAP 1990), and the Surface Water Acidification Program in Europe (Mason 1990). What is less recognized is that in Sweden the role of natural sources of acidity remained contested for at least another decade (Bishop *et al* 2001), even though the issue of organic acidity had been recognized in other areas long before then (Gorham *et al* 1986). Indeed, it was the belief that stream pH below ~6.0 was caused by anthropogenic deposition that became the basis for a governmentally funded, surface water liming program that started in the 1980s and is still partly continuing 40 years and more than 350 million US dollars later. The expansion of this program into the humic waters of northern Sweden in the 1990s led to a heated policy-science controversy fueled by arguments about whether the biogeochemical properties of northern streams included a large component of natural (organic) acidity related to the high levels of dissolved organic carbon (DOC) common to the region.

While chronic acidification in the northern part of Sweden never was a major issue because of relatively low acid deposition levels (Korsman 1999), low pH episodes (often to pH well below 5.0)

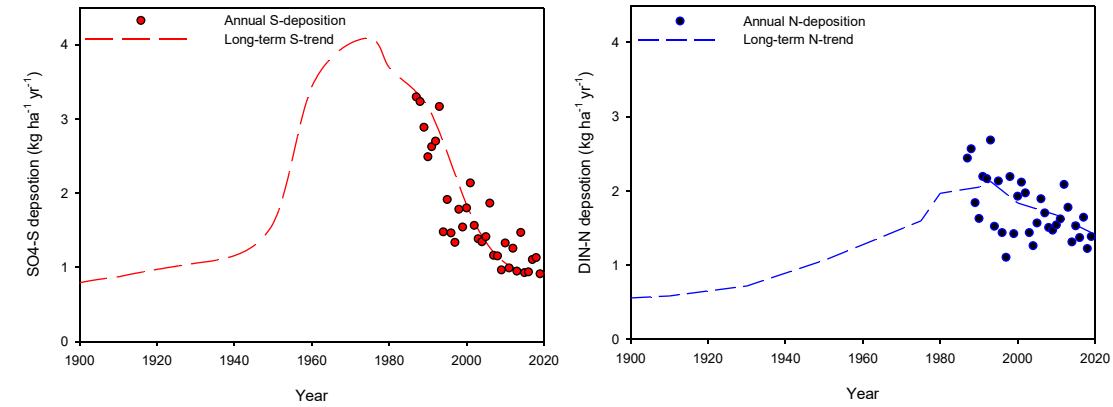

Figure 1. Annual sulfur (SO_4-S; panel (a)) and nitrogen (dissolved inorganic N; panel (b)) deposition measured in Krycklan, together with the long-term trend scaled to local conditions based on Engardt *et al* (2017). Every analysis of annual average is based on volume weighted daily samples.

associated with hydrological events, especially spring snowmelt, became the target for the controversy. Episodic pH declines are typically associated with changes in the concentration of most major dissolved solutes due to a shift in the sources and pathways of water feeding streams (Buffam *et al* 2007). During baseflow conditions, water is primarily derived from deeper, mineral soil horizons and groundwater sources with longer residence times, where the effect of mineral weathering renders water relatively circumneutral. During hydrological events, rain or snowmelt input generates a rising water table that activates water flow through more shallow layers of riparian soils that alters stream chemistry. These upper soil horizons are organic-rich and thus more acidic due to higher concentrations of natural organic acids (Ledesma *et al* 2017). In the former glaciated landscapes of the north, upper horizons also typically have a higher hydraulic conductivity allowing a more rapid transfer of water through the soil to the stream (Rodhe 1987). Thus, the pH of the surface water originating from episode-activated hydrological reservoirs is often depressed considerably relative to baseflow because large amounts of rain and snowmelt water can be transported through organic rich soil horizons (Bishop 1991, Laudon *et al* 2001). Any additional acidity originating from acid deposition can hence be superimposed on this natural pH decline and cause detrimental water quality effects if the combined effect exceeds critical thresholds.

Despite the threat to aquatic biodiversity, a large governmental liming subsidy, and growing recognition of the acidity problem in northern landscapes globally (see review by Eshleman *et al* 1992), surprisingly few scientific studies on episodic acidification were conducted in northern Sweden during this time. Early, isolated studies suggested that both anthropogenic (Jacks *et al* 1986) and natural factors (Bishop *et al* 1990, Jansson and Ivarsson 1994) could be the source of the increased acidity during spring flood and rainfall-driven flow events. At the same time, trends in acid deposition underwent a large transition with a peak in sulfate (SO_4) concentration in precipitation occurring in the late 1970s, followed by a decline to almost pre-industrial levels 20 years later when the policy debate in Sweden about natural acidity reached its peak. It was not until the initiation of the Swedish EPA *Episode Project* in the late 1990s that scientists from various disciplines came together to resolve this conflict. By combining a time-series analysis tool developed by Laudon and Bishop (1999) with an organic acid dissociation model to calculate pH (Köhler *et al* 2000) a conceptual model was developed which enabled the natural and anthropogenic components in acid episodes to be separated for the first time (Bishop *et al* 2000, Laudon *et al* 2000). With this work the conflict was partially resolved by showing that the majority of the pH decline during episodes was indeed of natural origin, but also that there was an anthropogenic component during snowmelt ranging between 0.1 and 0.2 pH units across northern Sweden in the late 1990s (Laudon *et al* 2001).

Since the peak of the acidification debate, sulfur (S) and nitrogen (N) deposition in snow and rain across northern Sweden have continued to decline to levels on par with pre-industrial conditions (figure 1). During this period only a few studies investigated the long-term effects of acidification, demonstrating that the anthropogenic component during snowmelt is strongly regulated by the winter S deposition levels (Laudon and Hemond 2002, Lawrence *et al* 2008), and revealing interactions between acidification and long-term trends of DOC levels in streams (Erlandsson *et al* 2011). Further, while earlier studies based on water chemistry trends predicted that episodic acidification would soon recover (Laudon and Bishop 2002), other, ongoing environmental changes

in the region (e.g. warming, increased drought frequency, and browning; Laudon and Sponseller 2018) have the potential to alter or mask such recovery trajectories. Finally, while stream pH was the focal point of the policy debate, other chemical legacies of acid deposition (e.g. trends in base cations) received less attention at the time, but are nonetheless important from soil and water quality perspectives.

Here, we revisit these legacies in northern Sweden, two decades after the culmination of the controversy, by examining water chemistry trends in the Svartberget/Krycklan research catchment. Research at this *flagship* site was central to resolving some of the scientific issues in this national debate. In the decades that followed, continued monitoring and research at the Krycklan catchment now provide a unique opportunity to revisit the original arguments related to the causes of acidity and test for trends in acidic episodes. Leveraging insight from numerous past studies at the site, the purpose of our retrospective analysis was to test the prediction that spring flood minimum pH has largely followed the recovery of acid deposition. At the same time, we expected that no such trend would be observable for rain driven episodes during summer and autumn, when the chemistry of event water is more strongly influenced by contact with organic soils. Finally, we also evaluated other large-scale acid recovery trends, asking whether this history has left legacies in the stream chemistry record, and placing these responses in the context of contemporary and future environmental changes in the region.

2. Methods

Our analysis focuses on the Svartberget catchment, also called C7 in several studies, which is one of the long-term monitoring sites within the Krycklan Catchment Study (Laudon *et al* 2013), and the site with the most extensive data record. Monitoring of runoff and precipitation at Svartberget began in 1980, providing an almost 40 year record of concentration and flux estimates of various solutes and compounds collected at an approximately weekly basis. Research in this reference catchment (no lime treatment) began with a focus on forest hydrology and nutrient balances, but soon became a focal site for studies of anthropogenic acidification, natural acidity, and DOC research in general (Bishop 1991). The Svartberget catchment is located approximately 50 km northwest of the city of Umeå in northern Sweden (64° 14′ N, 19° 46′ E). The 50 ha catchment is primarily forested by 100 year old Scots pine (*Pinus sylvestris*) and Norway Spruce (*Picea abies*), while 15% of the area is covered by minerogenic mires. The climate is characterized as boreal to sub-arctic with persistent snow cover during the winter season, which on average lasts 167 day (Laudon and Ottosson Löfvenius 2016). The 30 year mean annual temperature (1981–2010) was 1.8 °C with a long-term mean annual precipitation and runoff of 614 and 311 mm, respectively. The 10 year running average air temperature in the catchment has increased from 0.4 °C in 1985 to 2.9 °C in 2016. Deposition of SO_4 and dissolved inorganic nitrogen (DIN) peaked around 4 kg ha^{-1} yr^{-1} for SO_4-S and 2.5 kg ha^{-1} yr^{-1} for DIN-N around 1980 and 1990, respectively, before declining to almost pre-industrial levels by 2020 (figure 1).

Field and analytical methods used here have been described in detail by Buffam (2007) and Ledesma *et al* (2016) for major an- and cations, as well as by Tiwari *et al* (2019) and Fork *et al* (2020) for DOC. Briefly, our analysis is based on stream samples collected ca. weekly for a total of 1480 occasions between 1986 and 2019. We first used the observed minimum snowmelt (April–May) and rain driven (June–October) pH each year to ask whether the severity of acid episodes has changed over this sampling record during spring and summer/fall, respectively. Second, we explored time series of SO_4, base cations (BC, including Ca, Mg, Na and K), pH, and acid neutralizing capacity (ANC calculated as the sum of BC minus strong anions measured in chemical equivalents (eq)) to assess whether other water quality parameters reveal long-term responses linked to acid deposition recovery. For all solutes, we evaluated changes in concentrations over time using Mann–Kendall trend tests to determine the Theil–Sen estimate of slopes (using the package 'rkt'; Marchetto 2017). Finally, we also isolated important climate events during this period to explore how these relate to trends in SO_4 and other water quality parameters (e.g. pH and DOC). As part of this assessment, we calculated the Palmer drought severity index (Palmer 1965) for each summer (July–August) based on precipitation and temperature records from the site. We used this as a metric for summer drought conditions and tested the extent to which such episodes affected the autumn stream chemistry as well as altered the trends in chemical variables that may otherwise be linked to deposition recovery.

3. Results

Minimum pH during the spring flood has increased from 1987 to present by 0.12 units decade^{-1} ($p < 0.001$; figure 2(a)). Specifically, average minimum pH during spring flood between 1986 and 1995 was 4.35, and as low as 4.18, but has remained above 4.5 from 2010 to 2019, with an average of 4.63. This recovery of snowmelt related pH decline is concurrent with the acid deposition trajectory that has occurred over this 30+ year period (figure 1). In contrast to our expectations, we also observed an increase in episodic pH of 0.05 units decade^{-1} associated with precipitation-driven events during summer

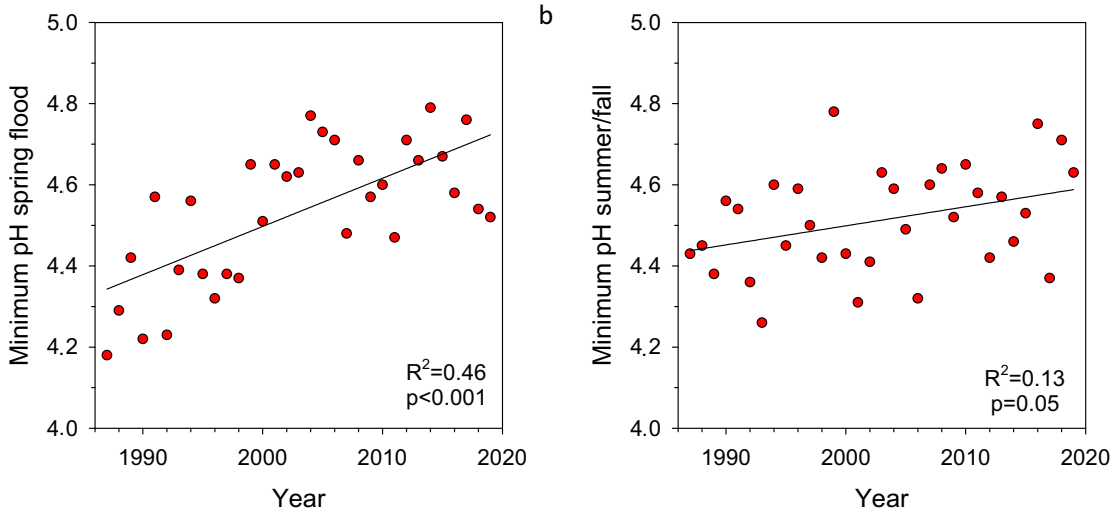

Figure 2. Episodic acidification recovery between 1986 and 2020. Panel (a): long-term annual minimum pH at spring flood (April and May). Panel (b): long term changes in rain driven episodes (June–October).

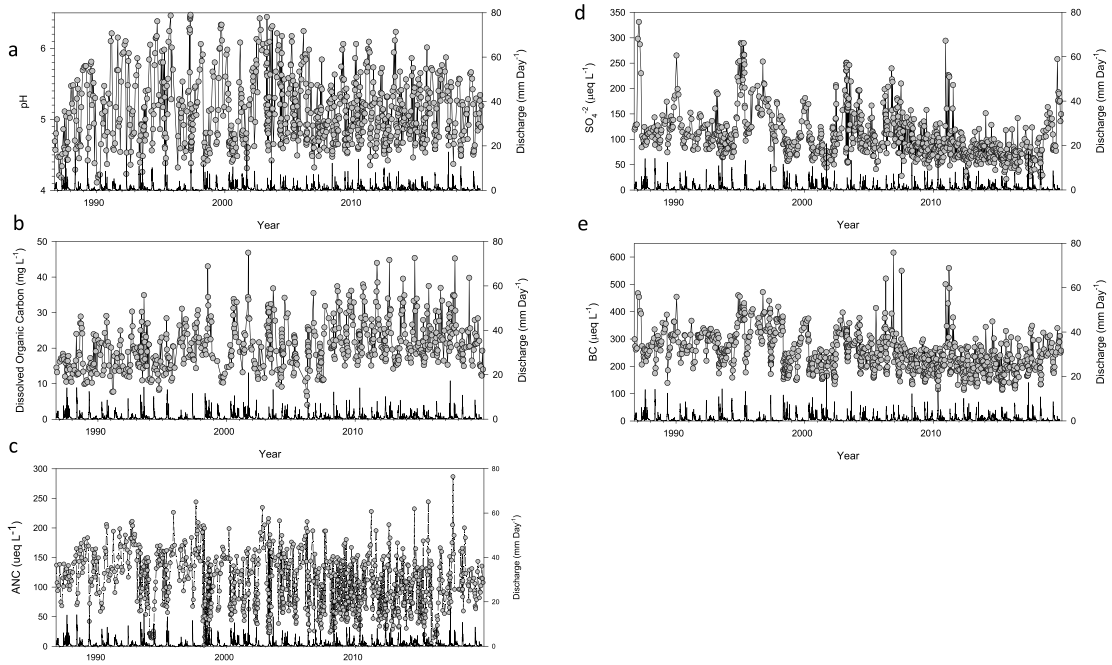

Figure 3. Long-term monitoring data of pH (panel (a)), dissolved organic carbon (DOC; panel (b)), acid neutralization capacity (ANC; panel (c)), sulfate (SO_4; panel (d)) and base cations (BC; panel (e)).

and autumn ($p = 0.05$; figure 2(b)). However, compared to the trends during snowmelt, recovery of acid episodes during summer and autumn was modest, translating into an increase in rain driven pH events of approximately 0.15 pH units over the entire period of monitoring.

While recovery in episodic acidification has been significant, we observed no statistically significant change in the average acid–base chemistry during the monitoring period, despite declines in acid deposition. If anything, this stream has become slightly more acidic over time (by ~0.1 pH units; figure 3(a)). This trend is most strongly linked to changes in DOC concentration, which increased by 0.35 mg l^{-1} yr^{-1} ($p \ll 0.001$) until 2005, and thereafter by an additional 0.15 mg l^{-1} yr^{-1} ($p = 0.05$; figure 3(b)). While the nature of this relationship has changed over time, DOC remains an important predictor of pH in this stream (figure 4(a)). Finally, ANC has also declined by approximately 35 μeq l^{-1} over this entire period (-1.1 μeq l^{-1} yr^{-1}, $p \ll 0.001$; figure 3(c)), equal to a loss of approximately one-fourth of the total acid neutralization capacity.

Two of the most striking patterns in the long-term stream chemistry record are the changes in SO_4 and base cation (BC) concentrations. Here,

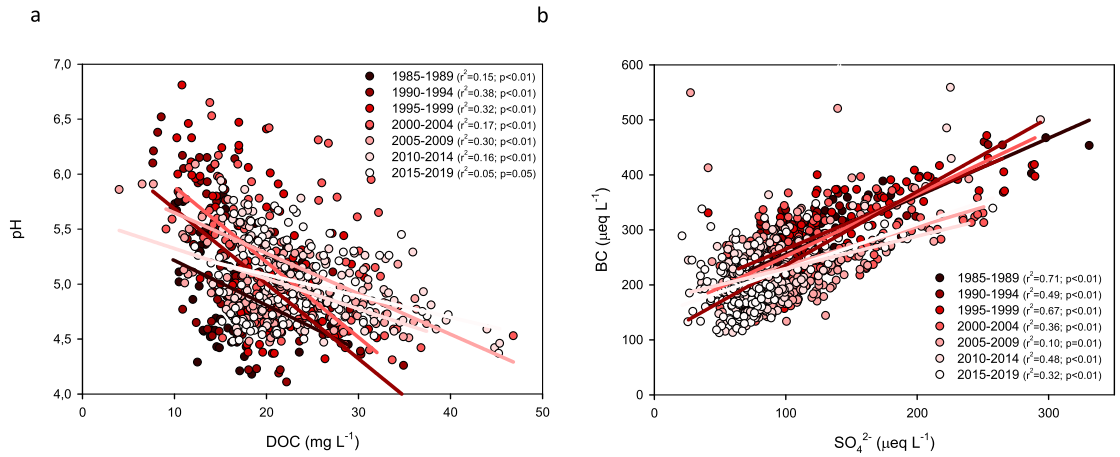

Figure 4. Correlation between DOC and pH (panel (a)) and SO_4 and base cations (BC; panel (b)). Data in both panels are divided into 5 year intervals to illustrate temporal changes in these bivariate relationships.

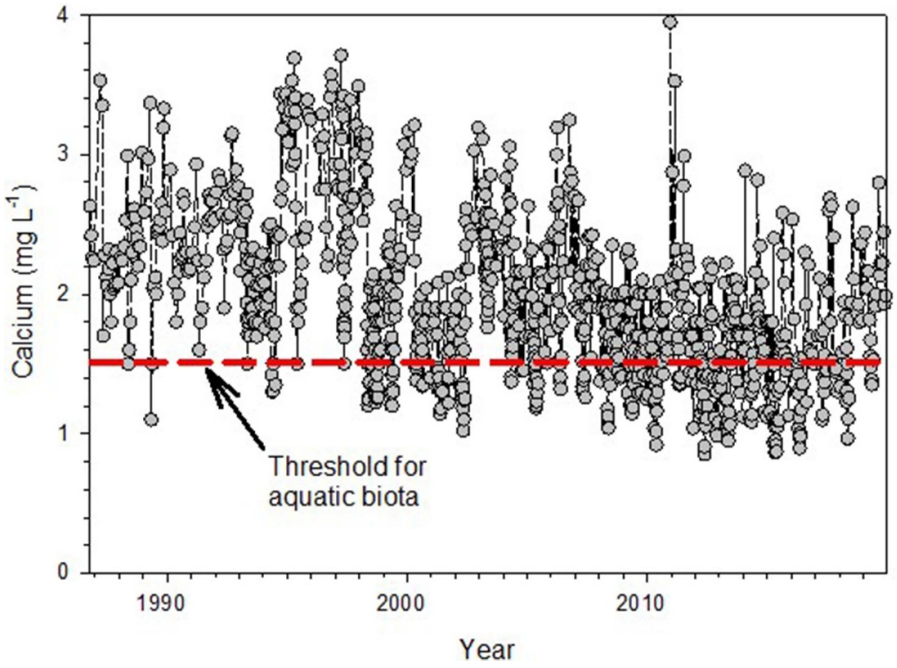

Figure 5. Long-term calcium (Ca) trend for site 7 in the Krycklan Catchment. The line at 1.5 mg l^{-1} represent a critical threshold for aquatic organisms (Jeziorski *et al* 2008).

SO_4 has declined by approximately 43% on average (-1.7 μeq l^{-1} yr^{-1}, $p \ll 0.001$; figure 3(d)), while the sum of BCs has declined by 37% (-2.3 μeq l^{-1} yr^{-1}, $p \ll 0.001$). Of the individual base cations, Ca has declined by 44% (-1.6 μeq l^{-1} yr^{-1}, $p \ll 0.001$; figure 5), Mg by 32% (-0.4 μeq l^{-1} yr^{-1}, $p \ll 0.001$), K by 23% (-0.1 μeq l^{-1} yr^{-1}, $p \ll 0.001$), and Na by 5% (-0.1 μeq l^{-1} yr^{-1}, $p = 0.04$). Although the percentage decline in SO_4 has been somewhat larger, the decrease in total BC eq has surpassed that of SO_4 by approximately 30 μeq l^{-1}. This pattern suggests that the absolute loss in BC has been greater than that from SO_4, which corroborates the long-term decline in ANC reported above, despite decreasing acid deposition. Worth noting is a strong linear correlation between the concentrations of SO_4 and BC across the entire monitoring period (figure 4(b)). Yet, dividing these data into 5 year increments reveals two different populations of correlations, one with higher SO_4 per BC prior to year 2005 and one with a lower ratio in the years that follow. By comparison, we observed no clear correlation between SO_4 and stream DOC on either episodic or intermediate (5 year) time perspectives.

Superimposed on these long-term trends, drought-related changes in water chemistry were also evident in the record. The most striking of these responses was observed for SO_4 concentrations in autumn and winter, which were positively correlated with the Palmer drought severity index in the

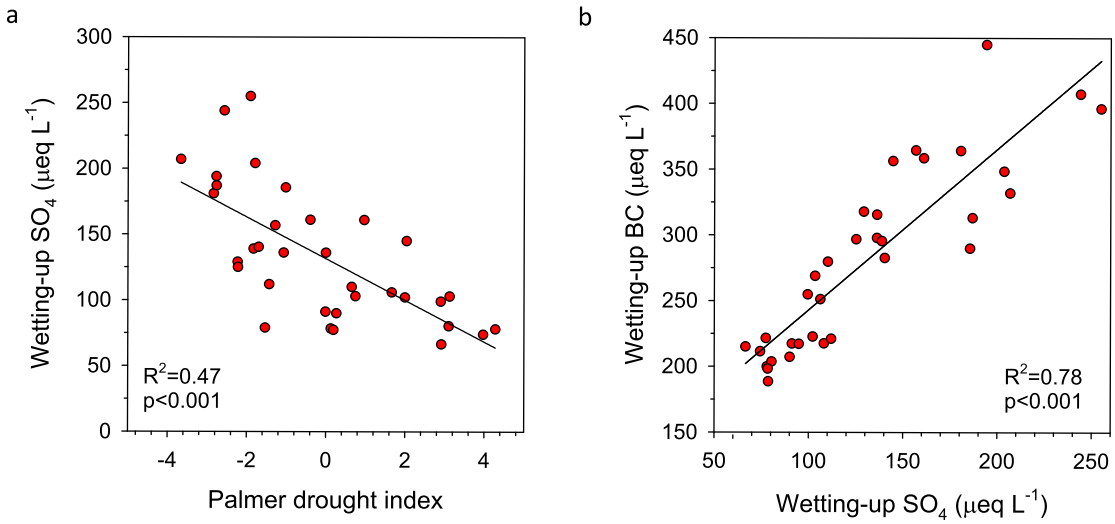

Figure 6. Panel (a): correlation between sulfate (SO_4) concentration during the autumn wetting-up phase and the severity of drought in the preceeding summer, represented by the Palmer drought index. Panel (b): the relationship between SO_4 and base cations (BC) release following these same summer–autumn transitions.

preceding summer ($p < 0.01$; $R^2 = 0.47$; figure 6(a)). In turn, this SO_4 response to drought strongly affected BC concentrations ($p < 0.01$; $R^2 = 0.78$), with a close to 1:1 response in the equivalent concentration of the two solutes (figure 6(b)). Here, the correlation with individual base cations was especially strong for Ca ($p < 0.01$; $R^2 = 0.83$) and Mg ($p < 0.01$; $R^2 = 0.77$), but also evident for Na ($p < 0.01$; $R^2 = 0.35$) and K ($p = 0.05$; $R^2 = 0.10$). As a result, neither pH nor ANC were affected by the drought-induced release of SO_4 ($p > 0.1$; $R^2 = 0.01$ and $p > 0.1$; $R^2 = 0.00$, respectively).

4. Discussion

In light of conflicting views of the role of acid deposition in northern Sweden, we look back at the historical time-series to disentangle anthropogenic influences from natural variability in acid-base chemistry, yet we also discover long-term changes not anticipated at the time. For example, the recovery of episodic acidification during snowmelt is in line with earlier predictions (Laudon and Bishop 2002). The rainfall induced episode recovery, however was less expected. Furthermore, despite recovery in atmospheric inputs and acidity during episodes, average pH has actually *decreased* slightly, which likely has to do with concurrent increases in organic acidity and DOC. Interestingly, while large scale studies show changes in both DOC (Monteith *et al* 2007, Erlandsson *et al* 2008), and more recently Ca (Weyhenmeyer *et al* 2019), these trends were not expected to be so dramatic in this northern latitude stream because of the relatively low level of historic acid deposition.

Many northern aquatic environments have been acidified by natural processes ongoing since the last glaciation (Renberg *et al* 1993). The long-term accumulation of soil organic matter and production of organic acids, together with the aging of mineral surfaces have slowly decreased pH and reduced base cation concentrations in soils and groundwater, while increasing DOC concentrations across recently glaciated landscapes (Rosén *et al* 2011). Superimposed on this long-term chemical trajectory are transient, hydrologically driven changes in stream water chemistry associated with rain and snowmelt events that induce episodic declines in pH caused by a dilution of ANC and increase in DOC (Erlandsson *et al* 2011). Despite the anthropogenic contribution to acid pulses evident from the long-term record (e.g. figure 2), these episodes are, and will continue to be driven by organic acids in this northern region, with large pH declines based on natural processes alone (Laudon *et al* 2001).

The recovery in spring flood episodic acidification is well in line with what was predicted, because the anthropogenic contribution to ANC and pH decline during these events is strongly related to the winter deposition level of SO_4 (Laudon and Bishop 2002). The direct connection between the acid deposition load and spring flood chemistry is made possible by the strong hydrological connection between melting snow and adjacent streams via shallow groundwater flowpaths (Laudon *et al* 2007). Given that atmospheric SO_4 deposition has decreased by over 80% since the peak in early 1980s, it is not surprising that there has been a concurrent recovery in these acid episodes during spring flood. By comparison, the recovery observable for rain induced acid episodes was less expected because the direct hydrological connections are considerably weaker during summer and autumn rainfall events. However, we know now that rain induced episodes also can result in the rapid transfer of event water during summer and

autumn episodes due to shallow groundwater tables, especially in wetlands (Peralta-Tapia *et al* 2015b), potentially reconciling this surprising finding. Further, it should be noted that this recovery is equal to 0.15 pH unit recovery over 35 years, and would not have been detectable without the long and detailed stream water records available.

One factor that complicates interpretation of the recovery from acidification is the concurrent trend in annual average air temperature, which in this region has increased by 2.5 °C the last 30 years (Laudon *et al* 2021). While annual precipitation has not varied directionally, the amount of rain-on-snow (Kozzi *et al* 2017), winter runoff (Laudon and Sponseller 2018) and evapotranspiration rates have all increased because of the warming trend, making the drivers of long-term change in stream water quality more difficult to disentangle. This is especially true for the long-term increase in DOC, which was most rapid prior to 2010 and thereafter has slowed. A combination of drivers that include recovery from acid deposition, climate and hydrologic change and, perhaps increases in forest growth all may contribute to this ongoing brownification trend (see Kritzberg *et al* 2020). Further, while SO_4 can drive DOC trends (Monteith *et al* 2007), it is difficult to isolate the importance of past anthropogenic deposition from the effects of generating SO_4 internally through the drying/rewetting of organic-rich riparian soils that characterize many Krycklan streams (Fork *et al* 2020). Regardless of the mechanism, this increase in DOC exerts powerful influences on stream pH that overwhelm most changes directly connected to the trends in acid deposition in the region.

In contrast to DOC, the over 37% decline in BC concentration has a clear mechanistic link to the change in acid deposition. Results suggest that the decline in SO_4 deposition has reduced the mass of mobile anions that co-transport positively charged ions, primarily base cations, through the groundwater system into the stream (Reuss and Johnson 1986). Dividing the time-series into 5 year intervals (figure 4(b)) illustrates a switch in the regression slopes between those samples collected prior to 2005, and those after. Whether this switch in BC per SO_4 is related to changes in acid deposition or more to internal soil processes is not currently known, but the overall pattern points to a strong connection between SO_4 and BC in stream water over time. Of the base cations, Ca is the cation most affected by this change in acid deposition and SO_4 inputs, which is line with other studies (Lawrence *et al* 1999). During recent years, Ca has decreased below 1.5 mg l^{-1} for several sampling occasions, which is seen as a critical level for many aquatic species (Jeziorski *et al* 2008). In fact, during the last 5 years, over 50% of the observations have been below this critical threshold, whereas 10% of observations have been 1.0 mg l^{-1} or lower. If the downward trajectory of Ca in stream water continues, it could become a serious concern for the aquatic biota of small streams and headwater lakes in this region (Weyhenmeyer *et al* 2019).

The long-term declining Ca trend raises a series of new questions regarding what the natural pre-industrial background concentration were, and if other external drivers—both natural and anthropogenic—can affect this trajectory in the future. Under natural conditions, chemical weathering of soils and bedrock is the only major original source of Ca to most inland waters. For regions dominated by slowly weathered bedrock, as is most of Sweden, this process is very slow, resulting in low natural Ca concentrations, especially in small streams dominated by relatively short groundwater travel times (Peralta-Tapia *et al* 2015a). In retrospect, large-scale liming in northern Sweden, in the past intended to elevate stream pH, could be perhaps be a future management approach aimed at maintaining Ca levels. Because lime primarily consists of calcium carbonate, its application could theoretically function as a remedy for streams with too low Ca concentrations. However, similar to the debate regarding the usefulness of liming to increase pH of northern streams, such an intervention would likely be controversial, misguided, and cause larger problems than it would solve. Based on studies in Sweden and elsewhere, applying lime or similar compounds to soils can cause delayed and unpredictable responses (Rosi Marshall *et al* 2016) and can have potentially negative effects on terrestrial ecosystems (Löfgren *et al* 2009). Similarly, liming in naturally acid streams can alter the composition of aquatic communities, with consequences for the ecosystem processes they support (e.g. litter decomposition; Mckie *et al* 2006). Hence, without a proper understanding of the natural background levels or the long-term consequences of massive Ca additions, such a remedy would require much more scientific evaluation before it could be considered.

Periodic drought events also complicate the assessment of recovery from acid deposition. Droughts are known to cause large changes in stream water quality, including extreme SO_4-related acid surges during the wetting-up phase that have been shown to delay the recovery from episodic acidification both in Sweden (Laudon 2008), and elsewhere (Mitchell *et al* 2008). Consistent with this, SO_4 concentrations were several times higher during the wetting-up phase that followed severe summer droughts in our long-term record (figure 6(a)). However, in contrast to other studies this response induced no corresponding change in either pH or ANC, but instead caused an almost 1:1 response in the BC concentrations, especially Ca (figure 6(b)). One interesting observation is that the drought related release of SO_4 (and BC) is similar today as it was almost four decades ago. In fact, while the long-term trend in SO_4 clearly is decreasing, there is no

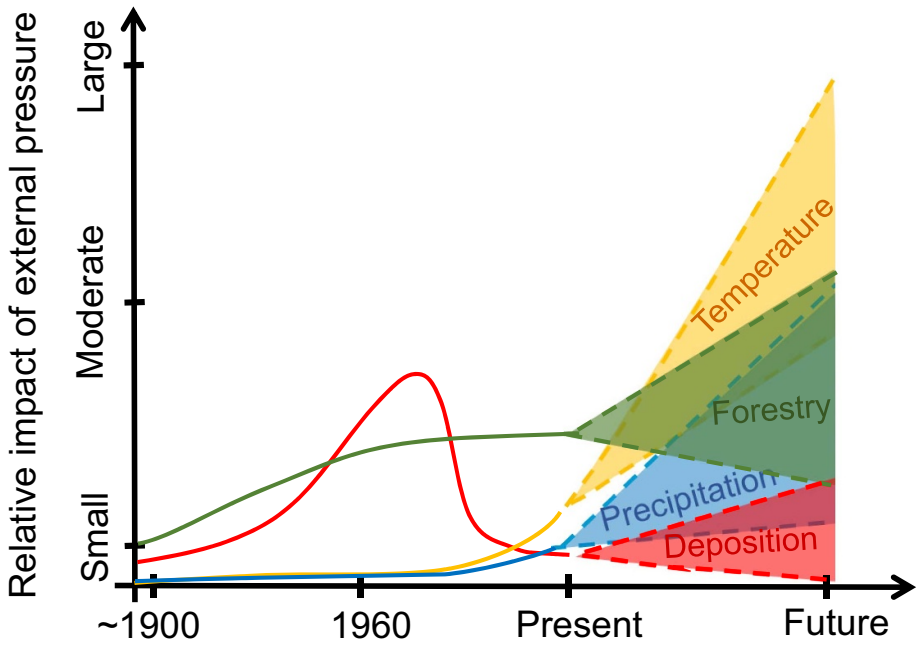

Figure 7. Schematic overview of past, present, and future potential external pressures on aquatic ecosystems in the boreal forest region based on work by Laudon *et al* (2011), Schelker *et al* (2013), Lidman *et al* (2014), Oni *et al* (2015), Teutschbein *et al* (2018), Gustavsson *et al* (2019) and Gomez-Gener *et al* (2020).

trend in the drought related peak SO_4 concentrations across the 35 year monitoring period. Since we could expect that the drought released SO_4 derived from legacy deposition would decrease over time, this result suggests that the time-series is too short or that the main source of the released SO_4 instead originates from weathering. In either case, extreme droughts are predicted to become more frequent and severe in northern Fennoscandia (Spinoni *et al* 2018), with potentially far reaching consequences for stream water quality in the future (Gomez-Gener *et al* 2020). Our results suggest that these effects may include a mechanism that generates further depletion of BC from catchment soils.

Finally, forests of the boreal region, especially in Fennoscandia, have been exploited for timber and biomass for several hundred years. Recent intensification of forest harvesting, in combination with new forest management practices, could have large biogeochemical consequences for both terrestrial and aquatic ecosystems, including effects on the storage and availability of carbon, N, and base cations (e.g. Laudon *et al* 2011, Lucas *et al* 2016). It is unclear to what extent weathering can maintain the store of base cations in these soils, much less replenish available BCs depleted by acid deposition in northern Sweden (Akselsson *et al* 2019). Failure to compensate for these losses could have consequences for forest growth, but since forests still have access to considerable stores of BC in the soil (Rosenstock *et al* 2019), it is the streams that that may be most susceptible to these effects (de Jong *et al* 2017). Of greatest concern is if the declining Ca concentrations in streams will continue in a unidirectional manner from a combination of pressures that include the recovery from acidification, forestry, and climate change.

5. Summary and implications

It is remarkable how clear the picture of natural acidification driven by DOC during hydrological episodes now seems. However, in the 1990s, this connection was much less obvious and became obscured by inappropriate models and lack of proper field data. Further, while aspects of this story have played out as expected (e.g. episodic acid recovery), other trends are more surprising, and others still reflect the emergence of new environmental changes that influence some of the same solutes (e.g. brownification). One lesson here is that piecing this complexity together would not have been possible without the combination of long-term monitoring and basic research, which together clarified the hydrological and biogeochemical processes underpinning acidic episodes and catchment responses to atmospheric deposition. A similar marriage between research and monitoring will be critical as new threats to water quality emerge (figure 7). In this sense, our contention is that these high latitude landscapes currently sit at a cross-roads where recovery from a past stressor (e.g. deposition) is giving way to new external pressures linked to climate change (e.g. extreme drought and flooding), forestry intensification, and long-range transport of new contaminants. To understand the new emerging pressures the value of long-term hydrochemical and hydrological time-series cannot

be overstated (Tetzlaff *et al* 2017). Unfortunately, recent years have seen the closure of a large number of long-term research sites, especially in the north (Laudon *et al* 2017), which is perhaps the greatest threat to our ability as a society to identify, understand, and remediate negative effects on water quality.

Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: www.slu.se/Krycklan.

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ORCID iD

Hjalmar Laudon https://orcid.org/0000-0001-6058-1466

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