

Article

Climate Effects on Water Chemistry in Acid-Sensitive Catchments

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Abstract

Substantial declines in acidifying emissions across Europe have led to pronounced chemical recovery of Norwegian surface waters. In recent decades, however, changes in water chemistry have increasingly coincided with climate change, complicating the attribution of observed trends to individual drivers. This study assesses whether ongoing climate change has produced detectable effects on freshwater chemistry in Norway and how these effects vary among catchments with differing sensitivities to acidification. In this study, the Model of Acidification of Groundwater In Catchments (MAGIC), which is based on current understanding of the processes governing acid–base chemistry in soils and waters, was used to simulate the effects of declining acid deposition. Deviations between observed and modelled water chemistry were provisionally interpreted as climate-related effects. However, these residuals may also reflect model or parameter uncertainty and other unaccounted-for processes. The analysis draws on long-term monitoring data (1986–2022) from 59 acid-sensitive Trend Lakes distributed across Norway, together with four Field Research Stations (1986–2020) representing contrasting hydroclimatic and biogeochemical conditions. Temporal trends were evaluated using the Mann–Kendall test and Sen’s slope estimator, while relationships between inferred climate effects and climatic variables were examined using Pearson’s correlation analysis. Across the Trend Lakes, inferred climate effects were predominantly positive for acid-neutralising capacity (ANC) and weathering-derived cations, suggesting that climate change may contribute to accelerated chemical recovery, particularly in catchments less sensitive to acidification. The inferred climate effects varied substantially among the Field Research Stations. Higher temperatures were generally associated with enhanced recovery, possibly through intensified silicate weathering, whereas increased precipitation and runoff appeared to dampen recovery. Overall, the results suggest that climate change exerts a measurable influence on freshwater chemistry in Norway, although the magnitude and direction of the response are strongly modulated by catchment-specific characteristics. While previous studies have identified climate-related influences on individual chemical variables, quantitative attempts to separate climate- and acid-deposition-related effects across a large number of acid-sensitive catchments remain rare. Here, we use deviations between observed water chemistry and MAGIC simulations of acid deposition recovery as a screening approach to investigate whether climate-related signals can be detected at the national scale and whether these signals vary among catchments with differing sensitivities to acidification.



Academic Editor: Paula M. Carreira

Received: 30 April 2026

Revised: 14 July 2026

Accepted: 16 July 2026

Published: 17 July 2026

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Keywords: climate change effects; freshwater chemistry; acid deposition recovery; acid-neutralizing capacity (ANC); MAGIC model; long-term monitoring; catchment processes; Norway

1. Introduction

Regional pollution levels in Norway exhibit a pronounced south–north gradient, largely driven by the deposition of long-range transported air pollutants from continental Europe. Substantial reductions in emissions of acidifying gases across Europe have, however, led to a marked decline in acid deposition in Norway [1]. Nationally, concentrations of sea salt-corrected sulphate (SO_4^{2-*}) in freshwaters have decreased by approximately 75% [2]. This reduction in acid rain has decreased the input of strong acid anions to catchments, resulting in substantial chemical recovery of surface waters and improved conditions for aquatic ecosystems. Recovery has been characterised by increases in acid-neutralising capacity (ANC), dissolved organic matter (DOM), and pH, together with declines in concentrations of toxic inorganic aluminium. In this study, ANC is defined according to the convention commonly used in acidification research as the charge balance between major cations and strong acid anions. Although ANC is often similar to alkalinity in carbonate-rich waters, this equivalence does not necessarily hold in acid-sensitive Norwegian surface waters, where bicarbonate concentrations are low and organic acid anions may contribute substantially to charge balance. Under such conditions, ANC cannot be assumed to be equivalent to bicarbonate concentration, while changes in weathering-derived cations, strong acid anions and organic acids may all influence ANC. Nitrogen deposition has also declined during the study period, potentially influencing nitrate export. However, nitrate concentrations are additionally regulated by processes such as mineralisation, nitrification and plant uptake, and denitrification, resulting in a long-term accumulation of reactive nitrogen within the catchments.

Over the past two decades, declines in acid deposition and the associated chemical recovery trends have begun to level off [2]. Acid deposition previously enhanced the leaching of divalent base cations from soils through charge balance requirements associated with mobile sulphate anions. Here, the term base cations refers to calcium, magnesium, sodium, and potassium. As sulphate deposition declined, this cation depletion gradually diminished. Consequently, the long-term decline in non-marine calcium and magnesium ($\text{Ca}^{2+} + \text{Mg}^{2+}$)* has slowed and, in some regions of mid- and northern Norway, reversed to increasing trends [2]. The term non-marine calcium and magnesium refers to the fraction derived from catchment weathering after correction for sea salt inputs originating from marine aerosols.

Water chemistry in rivers and lakes is also influenced, both in the short and long term, by climate change and climate variability. Warming has been particularly pronounced since the 1980s. Relative to the reference period 1961–1990, mean temperatures during 1991–2020 were approximately 1.0 °C higher, corresponding to a total increase of about 1.4 °C since the beginning of the 19th century. The period 1991–2020 represents the warmest climatological normal on record in Norway, both nationally and across all regions. This warming has resulted in longer growing seasons and milder winters. Concurrently, annual precipitation has increased by approximately 20–21% since the beginning of the 20th century, with the most pronounced increases occurring after 1980. Much of this increase is associated with more intense precipitation events, which have important implications for flood risk [3] and catchment hydrology. More frequent and intense weather events can directly influence

water chemistry by altering flow paths, residence times, and the relative contribution of soil water and groundwater to streams and lakes [4].

The extended growing season has also contributed to increased terrestrial biomass production. The forest biomass in Norway increased by 29% between 1971 and 2000 [5], driven by climate change as well as other environmental and land use factors [6]. Increased biomass production may enhance the generation of DOM, including root exudates, and increase root respiration. These processes can elevate concentrations of weak organic acids and carbonic acid, which, together with higher temperatures, may enhance mineral weathering. In some catchments, such climate-driven effects may accelerate recovery from acidification, whereas in others they may adversely affect water quality. The influence of climate change on water chemistry is expected to intensify as warming continues [7]. If greenhouse gas emissions proceed at current rates, temperatures in Norway could increase by as much as 4.2 °C by the end of this century [3], accompanied by further increases in precipitation and more frequent extreme weather events.

Several studies based on long-term monitoring of Norwegian rivers and lakes have linked observed chemical changes to climate variability and climate change. For example, de Wit et al. [8] attributed differences in nitrate export from small experimental catchments to reduced snowmelt and lower nitrogen deposition. Vogt et al. [9] demonstrated that, in catchments less affected by acid deposition, climate change may increase alkalinity and ANC, through enhanced silicate weathering of predominantly Precambrian granitic and gneissic rocks, driven by organic and carbonic acids and resulting in increased export of organic acid anions (Org^-), bicarbonate (HCO_3^-), and base cations such as calcium. Higher temperatures may enhance silicate weathering by increasing reaction kinetics, biological activity, and carbonic acid production, thereby increasing the release of weathering-derived cations and bicarbonate to surface waters [10,11], while increased precipitation reduces water residence time and thereby the effect of weathering [12]. De Wit et al. [13] analysed DOM trends in 426 minimally disturbed headwater systems across Europe and North America and found that climatic and chemical drivers were of comparable importance during 2002–2016. Increased summer precipitation enhanced DOM export, whereas warming appeared to dampen this response. Similar climate effects on DOM export have previously been reported by several other publications [14–17].

This study aims to assess whether climate change has altered freshwater chemistry in Norway and to improve our understanding of how climatic variability influences water quality. Although previous studies have identified climate-related influences on individual chemical variables, quantitative attempts to separate climate-related effects from those of declining acid deposition across a large number of acid-sensitive catchments remain rare. Here, we use deviations between observed water chemistry and MAGIC simulations of acid deposition recovery as a screening approach to investigate whether climate-related signals can be detected at the national scale and whether these signals vary among catchments differing in sensitivity to acidification, with four Field Research Stations providing detailed case studies of potential mechanisms.

2. Materials and Methods

2.1. Monitoring Data

The data used in this study originate from the national monitoring programme ØkoForsk Trend [2]. Since 1986, the programme has conducted annual water chemistry surveys following autumn turnover in 78 Trend Lakes, complemented by high-frequency monitoring at six Field Research Stations. All sites are small (median catchment area: 6.21 km²), relatively acid-sensitive headwater lakes distributed across Norway. Their catchments are predominantly composed of boreal forest, peatland, and alpine ecosystems

developed on thin till deposits overlying Precambrian crystalline bedrock, including granites, gneisses, and related metamorphic rocks, although local geological variation occurs. Carbonate-rich lithologies are uncommon, and pyrite-bearing sedimentary rocks are of limited extent. Consequently, silicate weathering is generally the dominant long-term source of base cations in solution. In this study, 59 of the 78 Trend Lakes and four of the six Field Research Stations were included. Nineteen Trend Lakes were excluded because of poor model performance.

The Field Research Stations Kårvatn, Storgama, Øygardsbekken, and Langtjern were selected because they represent contrasting combinations of climate, acid deposition history, geology, vegetation cover, soil development, and DOM concentrations (Table 1), thereby encompassing much of the environmental variability represented within the national monitoring network. Weekly water chemistry monitoring of small lake outlet streams began in 1974 at Langtjern, in 1975 at Storgama, and in 1993 at Øygardsbekken, while biweekly sampling has been conducted at Kårvatn since 1978.

Table 1. Characteristics of the Field Research Stations. Elevation indicates the minimum and maximum catchment elevations (m above sea level, a.s.l.). The catchment area includes lakes. Bedrock composition is listed in order of dominance. Mean annual temperature (Temp.) and mean annual precipitation (Precip.) are based on long-term climatic averages.

Field Research Station	Elevation (m a.s.l.)	Area (km ²)	Dominant Bedrock Lithology	Temp. (°C)	Precip. (mm)
Kårvatn	200–1375	24.4	Granitic gneiss/Meta-arkose	0.5	1844
Storgama	580–690	0.6	Granite	2.9	1037
Øygardsbekken	185–544	2.0	Granite/Norite/Migmatite	4.8	2063
Langtjern	510–750	4.8	Dioritic gneiss	1.2	791

Kårvatn (62°46′58″ N, 8°53′36″ E), a comparably large catchment located in central Norway, is characterised by relatively low sensitivity to acidification and minimal influence from acid deposition, resulting in very low concentrations of SO_4^{2-*} and NO_3^- . The catchment is sparsely vegetated and consists of thin peat and till deposits interspersed with scree and exposed bedrock. Despite the limited soil cover, both ANC and $(\text{Ca}^{2+} + \text{Mg}^{2+})^*$ have increased steadily since 1986. pH increased from 6.0 to 6.3 until 2010, followed by a slight decline to 6.1 by 2020. Concentrations of sodium (Na^+), chloride (Cl^-), and total organic carbon (TOC) remained relatively stable throughout the monitoring period [2].

Storgama (59°03′08″ N, 8°39′13″ E), located in a mountainous region of southern Norway, is characterised by shallow soils, a thin mineral soil layer, extensive exposed bedrock, and sparse vegetation. Concentrations of SO_4^{2-*} and NO_3^- declined markedly between the 1980s and 2020, from approximately 75 to 12 $\mu\text{eq L}^{-1}$ and from 15 to 2 $\mu\text{eq L}^{-1}$, respectively. Over the same period, ANC increased from −40 to +19 $\mu\text{eq L}^{-1}$, while $(\text{Ca}^{2+} + \text{Mg}^{2+})^*$ declined from approximately 40 to 19 $\mu\text{eq L}^{-1}$. pH increased from about 4.6 to 5.0. Concentrations of Na^+ and TOC increased, whereas Cl^- remained relatively stable [2].

Øygardsbekken (58°37′18″ N, 6°06′23″ E) is located in a humid coastal region of southwestern Norway and receives a mean annual precipitation of 2063 mm. The catchment lies within the coastal subalpine birch forest zone, with a field layer dominated by tall grasses and juniper growing on thin till deposits and exposed bedrock. Water chemistry is moderately affected by acid deposition. Concentrations of SO_4^{2-*} and NO_3^- declined from approximately 40 and 12 $\mu\text{eq L}^{-1}$, respectively, in the early 1990s to around 10 $\mu\text{eq L}^{-1}$ by 2020. During the same period, ANC increased from −14 to approximately +10 $\mu\text{eq L}^{-1}$, while $(\text{Ca}^{2+} + \text{Mg}^{2+})^*$ remained relatively stable at around 25 $\mu\text{eq L}^{-1}$. pH increased from

just below 5.0 to 5.5, and TOC increased from 1.2 to 2.1 mg C L⁻¹. Concentrations of Na⁺ and Cl⁻ remained relatively stable throughout the study period [2].

Langtjern (60°22'20" N, 9°43'36" E) is a dystrophic site in eastern Norway, with a catchment dominated by peatlands and low-productivity coniferous forest growing on thin till and peat deposits. The concentration of SO₄²⁻* declined similarly to those at Storgama, from approximately 71 to 14 µeq L⁻¹ between the 1980s and 2020, while NO₃⁻ concentrations remained low (<3 µeq L⁻¹). ANC increased from slightly above 0 to 47 µeq L⁻¹, despite a decline in (Ca²⁺ + Mg²⁺)* from approximately 74 to 40 µeq L⁻¹. This apparent decoupling between ANC and (Ca²⁺ + Mg²⁺)* reflects the importance of changing strong acid anion concentrations and organic acid chemistry in regulating ANC in humic waters. pH increased only slightly, from 4.7 to approximately 5.0. Sea-salt-corrected Na⁺ (Na⁺*) increased from approximately 5 to 15 µeq L⁻¹, while Cl⁻ concentrations declined. TOC concentrations were high and increased slightly, from approximately 9 to 11 mg C L⁻¹ [2].

All data are available through the national water database Vannmiljø [18]. Data from the Trend Lakes were used to assess national-scale patterns in inferred climate effects, whereas the Field Research Stations were used as mechanistic case studies to examine the relationships between interannual variability in the inferred climate-related effects on water chemistry and climatic variables.

Daily estimates of air temperature (°C), precipitation (mm), and runoff (mm) for the period 1957–2024 were obtained for each sampling site using the GridTimeSeries (GTS) service provided by the Norwegian Water Resources and Energy Directorate (NVE) [19].

2.2. Model Application

The Model of Acidification of Groundwater In Catchments (MAGIC v8; Ref. [20]) was used to simulate the effects of declining acid deposition. Several dynamic acidification models have been developed, including MAGIC, SAFE, and SMART. MAGIC was selected because it has been extensively applied and validated for Norwegian surface waters and because historical calibrations were already available for the Trend Lake network. MAGIC provides a robust process-based framework for reconstructing historical acidification and predicting future recovery by integrating the key biogeochemical processes governing soil and water chemistry over decadal timescales. Its strength lies in linking acid deposition to catchment processes and surface water responses within a mass balance framework. However, MAGIC operates at an annual time step and relies on simplified process representations and calibrated parameters, limiting its ability to resolve climate effects, short-term variability, seasonal dynamics, episodic events, and complex interactions involving organic matter. Consequently, climatic influences operating at a shorter temporal scale may contribute to residual variability that cannot be explicitly resolved by the model.

The model was calibrated for each monitoring site using observational data from 1986 to 1995 and subsequently applied to simulate water chemistry for the period 1882–2020/22. Assuming that MAGIC adequately reproduces the effects of declining acid deposition (cf. [21]), deviations between observed and simulated water chemistry (Δ) during 1986–2022 (1986–2020 for the Field Research Stations) were interpreted as residual variation potentially arising from climate-related processes not represented in the model. However, these residuals may also reflect model structural limitations, parameter uncertainty, and other unaccounted-for environmental drivers.

Deposition measurements from nearby monitoring stations do not always accurately represent conditions within individual catchments. To improve agreement between simulated and observed water chemistry, sulphate and chloride deposition were estimated using mass balance calculations during the calibration period. Base cation deposition was derived

from sea-salt ratios and further refined through optimisation of soil parameters, including organic acid content, weathering rates, base cation saturation, and Al–OH dissociation constants. These parameters are associated with substantial uncertainty owing to spatial heterogeneity, limited empirical data, and assumptions regarding pre-industrial (1882) soil conditions. This is particularly relevant for simulations of Na^+ . In addition, MAGIC assumes that temporal changes in organic acids are identical in soil water and surface waters, introducing a further source of uncertainty.

Deviations between observed and simulated values may reflect stochastic variability and model limitations in addition to potential climate-related influences. Model uncertainty was evaluated using Monte Carlo simulations, which generate 95% confidence intervals around model predictions and thereby provide a basis for assessing whether observed deviations can reasonably be explained by parameter uncertainty alone. Confidence in the interpretation of residuals is strengthened when deviations exceed model uncertainty bounds and exhibit coherent relationships with independent climatic variables.

The median percent root mean square error (%RMSE) across all sites during the calibration period was 16.6%, 10.3%, 7.6%, 7.5%, and 13.5% for ANC, Ca^{2+} , Mg^{2+} , Na^+ , and potassium (K^+), respectively, indicating generally good model performance. Performance was strongest for Mg^{2+} and Na^+ and somewhat weaker for ANC and K^+ . These values are comparable to those reported in previous regional MAGIC applications [16]. RMSE values below 20% are generally considered acceptable for regional-scale applications of MAGIC.

Because MAGIC operates at an annual time step, short-term variability in climate and hydrology is not resolved. Annual median values were calculated for each parameter at each Field Research Station before trend analysis, meaning that seasonal variability influences the median values but not the strength of the statistical tests. Since annual runoff variability is included in the model calibration, direct climate effects of increased precipitation—such as dilution and enhanced fluxes—are not explicitly attributed to climate change in this framework.

The MAGIC simulations for the Trend Lakes were originally performed as part of a study by Austnes et al. [22]. More information regarding the modelling of Trend Lakes and Field Research Stations is provided in [22].

Simulations for 19 of the 78 Trend Lakes were excluded because of unrealistic parameter estimates, the inability to reproduce observed calibration chemistry, excessive uncertainty (high RMSE), or because site characteristics fell outside the model's valid calibration range. These lakes were primarily located in northern Norway and in alpine regions. Compared with the included lakes, they were generally smaller (median 0.5 vs. 0.7 km^2), colder (median 0.87 vs. 4.03 $^{\circ}\text{C}$), drier (1037 vs. 1896 mm yr^{-1}), more alkaline (51 vs. 36 $\mu\text{mol L}^{-1}$), and lower in DOM (TOC: 1.9 vs. 4.2 mg C L^{-1}).

Inferred climate effects (Δ) were calculated for Ca^{2+} , Mg^{2+} , Na^+ , K^+ , NO_3^- , Al, pH, organic acid anions (Org^-), and acid-neutralising capacity (ANC) for the period 1986–2022 for the remaining 59 Trend Lakes. However, pH was excluded from subsequent analyses of the Trend Lakes due to poor calibration performance at several sites. Since Org^- is calibrated to reproduce pH, and its deviations depend on uncertainties in multiple parameters, these results should be interpreted with caution.

Significant ($p < 0.05$) monotonic trends in climatic and chemical variables were assessed using the Mann–Kendall test (MKT) [23], based on the Z statistics. The MKT is robust to outliers and missing data and does not require normally distributed variables. Temporal trends in inferred climate effects (Δ) were evaluated by correlating Δ with time (year) over 1986–2022 (Figure 1), hereafter referred to as “Time-trends”. Trend magnitudes were quantified among the Trend Lakes using Sen's slope estimator [24]. Pearson's correlation coefficient (r) was used to quantify the strength and direction of the Time-trends (Figure 1).

All MKT and Sen's slope analyses were conducted in Python 3.10 [25], while regression analyses were performed using Minitab 22.5.1 (64-bit) [26].

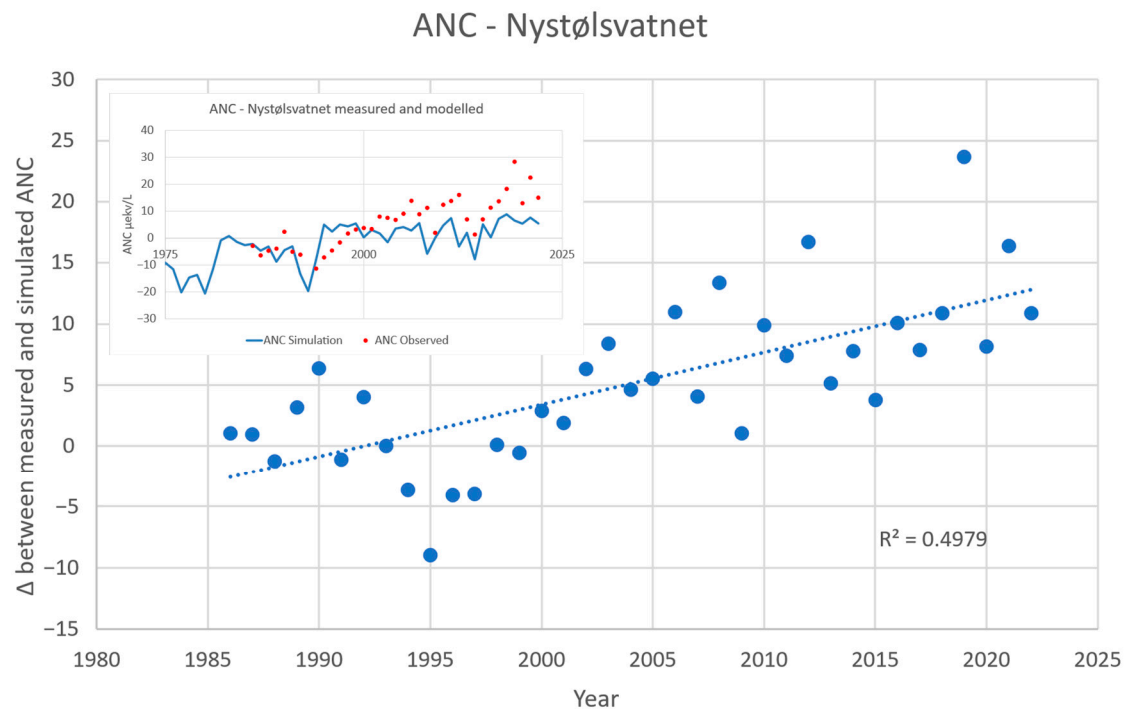


Figure 1. Pearson's correlation coefficient (r) for the Time-trend lake Nystølsvatnet—one of the Trend Lakes—as a measure of the strength and direction of the trend. The actual deviation between annual measured and simulated values of ANC is shown inserted.

3. Results

3.1. Inferred Climate Effects in Trend Lakes

All lakes exhibited significant (MKT) negative Sen's slopes for SO_4^{2-} concentrations, reflecting the decline in sulphur deposition. The largest reductions occurred in southern Norway, where historical sulphur deposition was greatest. Chloride (Cl^-) either showed no significant trend (66% of sites) or declined significantly. Base cations generally exhibited negative Sen's slopes, except for sodium (Na^+). This pattern was most pronounced for magnesium (Mg^{2+} ; 61% of sites) and potassium (K^+ ; 68%). In contrast, acid-neutralising capacity (ANC) exhibited positive Sen's slopes in all but two lakes, indicating increased buffering capacity and ongoing chemical recovery from acidification.

The largest number of significant correlations between inferred climate effects (Δ) and time (Δ vs. year; i.e., Time-trends), explaining more than 10% of the variance ($R^2 > 0.1$), was observed for Na^+ (42 of 59 lakes), followed by ANC (38), K^+ (30), NO_3^- (29), Al (28), and Ca^{2+} (27). Mg^{2+} exhibited the fewest significant Time-trends (21 lakes). All reported correlations were statistically significant ($p < 0.05$). For relationships exceeding the $R^2 > 0.1$ threshold, observed values frequently fell outside the model uncertainty intervals (0.025–0.975 quantiles), suggesting that the inferred climate effects are unlikely to be explained solely by model uncertainty. This is illustrated in Figure 2, which compares MAGIC simulations of acid deposition recovery with observed water chemistry at the Field Research Stations.

Among the significant Time-trends ($R^2 > 0.1$), positive trends predominated for Na^+ (97%), ANC (88%), Ca^{2+} (71%), and Mg^{2+} (63%), whereas fewer than 36% of the trends for K^+ , NO_3^- , and Al were positive. Collectively, these patterns suggest that climate-related processes may have contributed to accelerated chemical recovery from acidification across the Trend Lakes, expressed as increasing ANC and base cation concentrations.

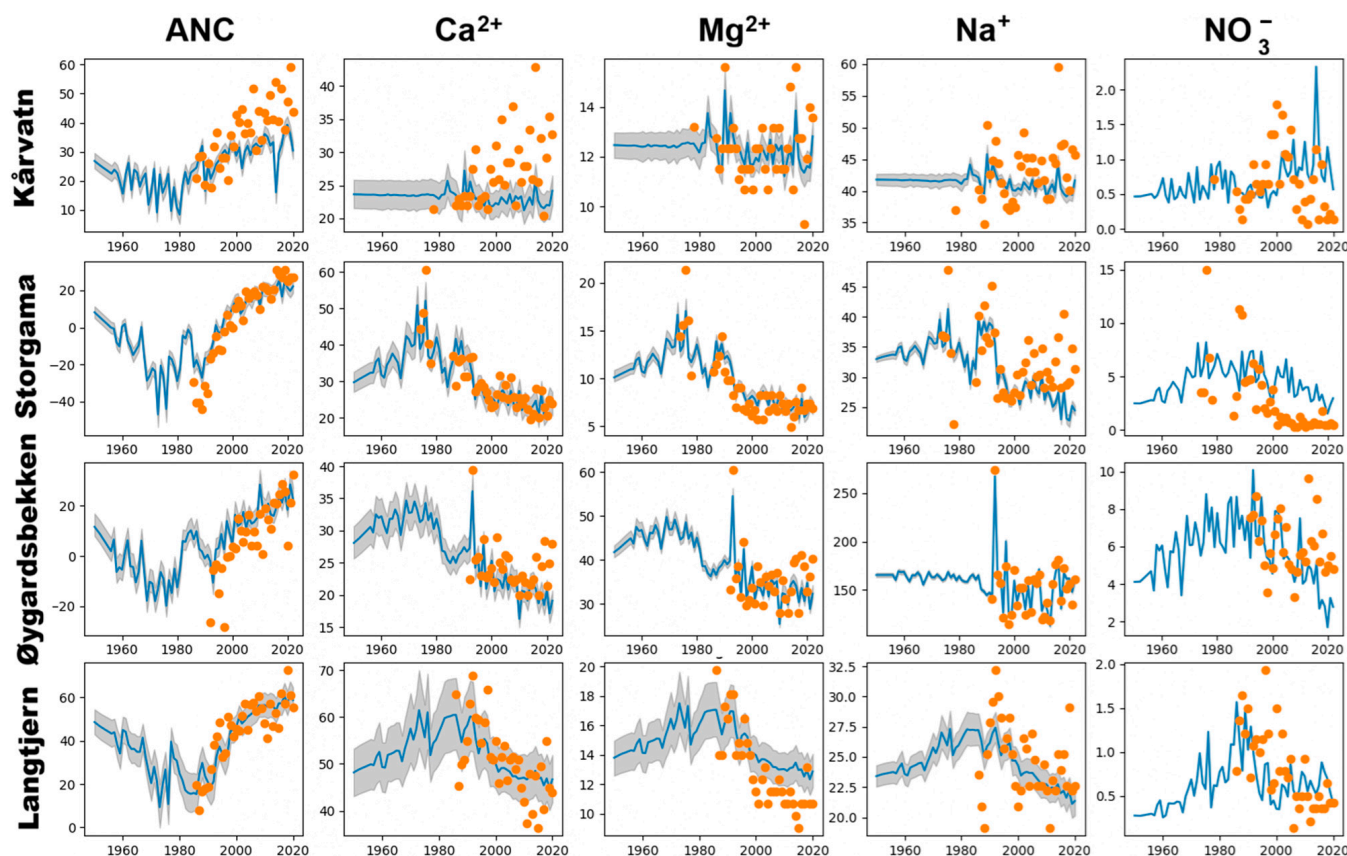


Figure 2. Results from MAGIC simulations of water chemistry ($\mu\text{eq L}^{-1}$) changes attributed to the declining acid deposition at the Field Research Stations. Orange points represent observations, the blue line indicates modelled values, and grey shading denotes model uncertainty.

Despite the use of MAGIC to account for declining acid deposition, Time-trends in the deviation (Δ) between the observed and modelled Ca^{2+} concentrations were positively correlated with Sen's slopes for SO_4^{2-} ($R^2 = 0.23$). The inferred climate effect on Ca^{2+} shifted from negative to positive as declines in SO_4^{2-} became weaker (Figure 3a). Because reductions in mobile sulphate anions would normally be expected to reduce Ca^{2+} concentrations through charge balance constraints, this pattern suggests that MAGIC may slightly overestimate the effect of declining acid deposition on Ca^{2+} reductions in strongly acidified regions while underestimating it in less affected regions. This is consistent with previous findings [2,13,27]. Interpretation is complicated by the co-variation between SO_4^{2-} declines and mean temperature ($R^2 = 0.27$), as the largest reductions in sulphate occurred in the warmer regions of southern Norway.

For NO_3^- , the opposite pattern was observed. Inferred climate effects shifted from positive Time-trends in regions with large reductions in acid deposition to negative trends in less affected areas (Figure 3b). This suggests that climate-related reductions in NO_3^- may be more pronounced in catchments historically less affected by acid deposition. Co-variation between acid deposition recovery and climate trends may contribute to this pattern, as temperature increases have been greatest in northern Norway. Similar mechanisms were proposed by de Wit et al. [8], who demonstrated that changes in winter climate, together with declining nitrogen deposition, influenced long-term nitrate trends in stream water.

For ANC, Mg^{2+} , and Na^+ , spatial variation in SO_4^{2-} Sen's slopes explained less than 9% of the variation in inferred climate effects, suggesting that MAGIC largely captures the effects of declining acid deposition for these variables.

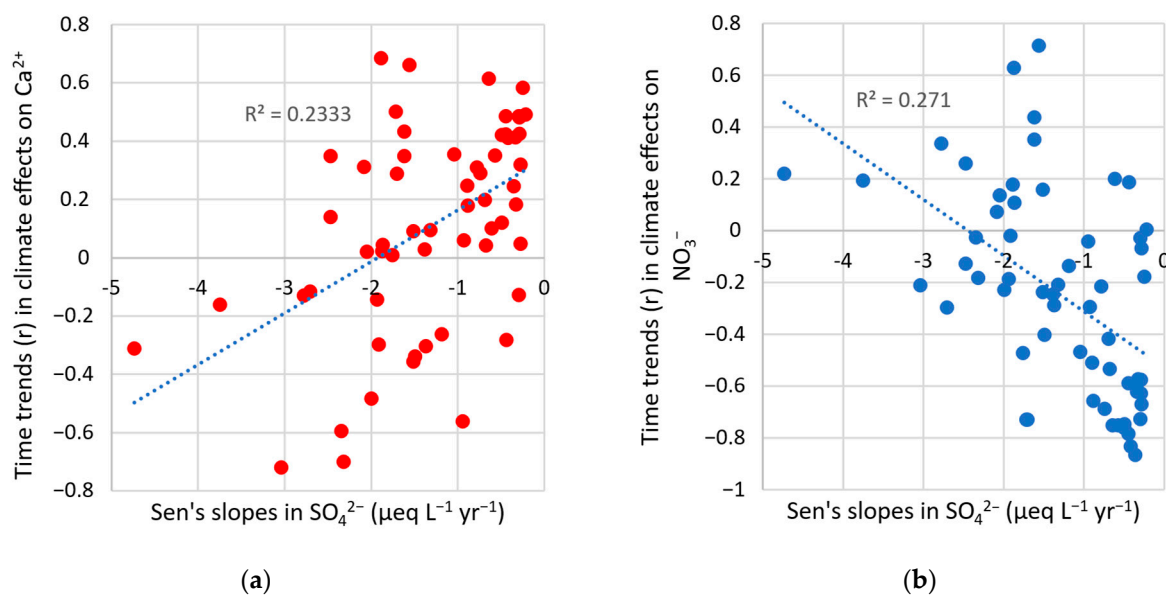


Figure 3. Spatial relationships between the strength and direction (r) of Time-trends in inferred climate effects (Δ/yr) for Ca^{2+} (a) and NO_3^- (b), and Sen's slopes for SO_4^{2-} (1990–2020) across the 59 Trend Lakes.

The model appears to overestimate the response of Ca^{2+} to declining acid deposition in strongly acidified regions and underestimate it in less-affected areas (Figure 3a). This interpretation is supported by the relationship between Ca^{2+} deviations and pH (Figure 4a), which shows predominantly negative Time-trends at low pH and positive trends at higher pH.

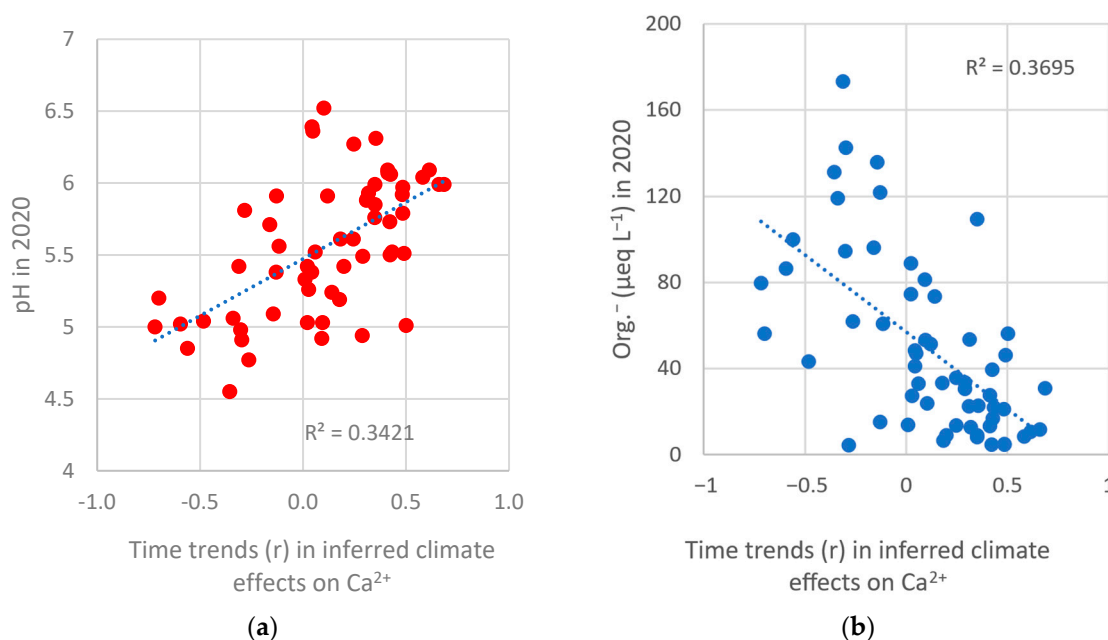


Figure 4. The relationship between measured pH (a) and organic anion charge (Org^-) in 2020 (b), and the strength and direction (r) of Time-trends in inferred climate effects (Δ/yr) on Ca^{2+} (1986–2022) across the 59 Trend Lakes.

One possible explanation lies in the representation of organic anions (Org^-) within MAGIC. Increasing Org^- charge enhances metal complexation and influences both weathering processes and solution equilibria. The model assumes a constant density of functional groups (site density, SD) on DOM. However, declining acid deposition has increased DOM

concentrations and altered DOM quality, with a shift towards more humic and coloured material [28–31]. This likely implies a decline in SD over time [32,33]. If SD is overestimated, the model will overestimate Org^- charge at a given pH and DOC concentration, leading to increasingly negative model bias through time (i.e., more negative r).

This interpretation is supported by the transition from positive to negative Ca^{2+} deviations with increasing modelled Org^- charge (Figure 4b). However, substantial uncertainty remains in the representation of Org^- within MAGIC, and these results should therefore be interpreted cautiously. The regression analysis identified a threshold near pH 5.5 (Figure 4a), above which Ca^{2+} deviations became increasingly positive. We interpret this threshold as reflecting a shift in the relative importance of carbonate-system buffering and silicate weathering, rather than a universal geochemical threshold [34,35].

3.2. Inferred Climate Effects at Field Research Stations

Annual inferred climate effects (Δ) on ANC, Ca^{2+} , Mg^{2+} , Na^+ , K^+ , NO_3^- , Org^- , Al, and pH at the four Field Research Stations were analysed using Pearson correlations with time (i.e., Time-trends) and with the following explanatory climate parameters: annual mean, median, and maximum values of temperature, precipitation, and runoff.

Only statistically significant relationships ($p < 0.05$) explaining more than 20% of the variance ($R^2 > 0.20$) were considered. Time-trends in the inferred climate effects (Δ/yr) were predominantly positive (Table 2), and the strongest and most consistent Time-trends were observed at Kårvatn and Storgama, whereas Langtjern exhibited only a weak negative Time-trend for Mg^{2+} . No significant Time-trends were identified for K^+ at any station.

Table 2. Significant ($p < 0.05$) Pearson’s correlation coefficients (r values; shown only for $R^2 > 0.20$) between inferred climate effects and year (Δ/yr , i.e., Time-trends) for the period 1986–2020 ($n = 35$ years).

Field Research Station	ANC	Ca^{2+}	Mg^{2+}	Na^+	K^+	NO_3^-	Org^-	Al	pH
Kårvatn	+0.571	+0.571		+0.490		−0.488	+0.796		
Storgama	+0.836		+0.522	+0.753			+0.883	+0.452	−0.843
Øygardsbekken	+0.456	+0.494	+0.481			+0.613	+0.744		
Langtjern			−0.576						

Climatic variables showed limited explanatory power for K^+ , and no variable explained more than 20% of its interannual variability at any station. Similarly, neither annual median nor maximum precipitation exceeded the explanatory threshold for any response variable. These predictors were therefore excluded from further analyses.

3.2.1. Kårvatn

At Kårvatn, the inferred climate effect (Δ) was positive for Org^- and pH throughout the study period, although only Org^- exhibited a significant Time-trend (Table 2). Positive inferred climate effects occurred in 78%, 70%, and 68% of the years for Ca^{2+} , ANC, and Na^+ , respectively, and all three variables exhibited significant positive Time-trends (Table 2; Figure 2). A positive Δ indicates that observed concentrations exceeded those predicted from declining acid deposition alone, suggesting that climate-related processes may have contributed to accelerated chemical recovery.

For Mg^{2+} , positive inferred climate effects were only slightly more common than negative effects (59% of the years), whereas NO_3^- exhibited more frequent decreases than increases and a significant negative Time-trend (Table 2). This pattern is consistent with enhanced nitrate retention, possibly driven by increased biological uptake and denitrification under warmer conditions and longer growing seasons.

Climatic variables explained a substantial proportion of the interannual variability in Δ at Kårvatn, with 20 relationships exceeding the $R^2 > 0.20$ threshold (Table 3). Higher temperatures were associated with increased ANC (Figure 5a), whereas runoff was negatively correlated with Δ for Ca^{2+} , Na^+ , Org^- , and ANC (Figure 5b). A similar pattern was observed for precipitation, reflecting its strong influence on runoff. Collectively, these results suggest that warming promotes chemical recovery, whereas increased precipitation and runoff exert a dampening influence.

Table 3. Significant ($p < 0.05$) coefficients of determination (R^2 ; shown only for $R^2 > 0.20$) between annual inferred climate effects (Δ) on water chemistry (Ca^{2+} , Na^+ , NO_3^- , Org^- , and ANC) and climatic variables for the period 1986–2020 at Kårvatn. No climatic variables explained more than 20% of the variation in Mg^{2+} , Al, or pH. Likewise, median temperature did not exceed this explanatory threshold. Red and blue coefficients indicate positive and negative correlations, respectively.

Kårvatn		Ca^{2+}	Na^+	NO_3^-	Org^-	ANC
Precipitation	Average	0.266			0.219	0.263
Temperature	Average		0.306			0.244
	Maximum		0.200	0.257		
Runoff	Average	0.310	0.247	0.203	0.352	0.340
	Median	0.324	0.208		0.328	0.333
	Maximum	0.245		0.293	0.327	0.258

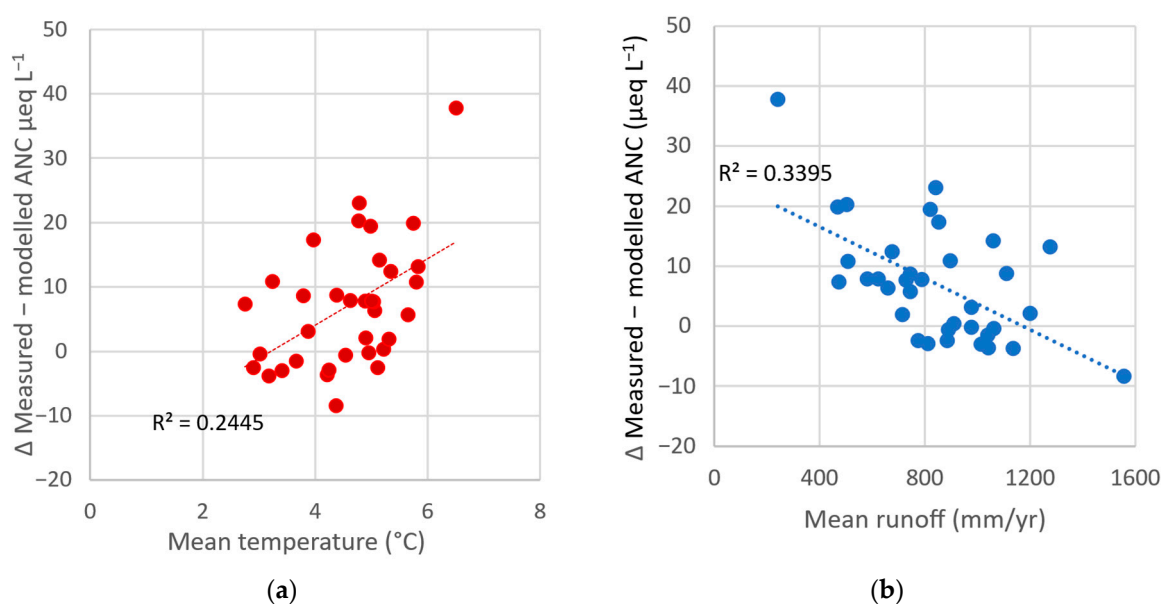


Figure 5. Significant ($p < 0.05$) correlation between annual inferred climate effects (Δ) on ANC (r) and mean annual (1986–2020) temperature (a) and mean annual runoff at Kårvatn (b).

At Kårvatn, where pH exceeds 5.5, water chemistry—and particularly the ANC—is strongly influenced by bicarbonate (HCO_3^-) generated through carbonic acid-driven weathering of Precambrian bedrock. During wet years, shorter water residence times may reduce weathering intensity, resulting in lower HCO_3^- production and reduced mobilisation of Ca^{2+} and Na^+ .

The inferred climate effect on NO_3^- became increasingly negative with increasing maximum temperature ($R^2 = 0.257$), supporting the hypothesis that warmer conditions enhance biological assimilation and nitrate retention.

3.2.2. Storgama

At Storgama, the inferred climate effects were consistently negative for K^+ and pH, although only pH exhibited a significant negative Time-trend (Table 2). Negative Δ values were also common for NO_3^- , Mg^{2+} , and ANC, occurring in 89%, 70%, and 62% of the years, respectively. Despite this, Mg^{2+} and, in particular, ANC exhibited positive Time-trends, reflecting a shift from negative Δ during the early part of the record to positive inferred climate effects in later years (Figure 2).

Overall, these patterns suggest that climate-related processes have increasingly contributed to chemical recovery at Storgama. Org^- exhibited positive inferred climate effects throughout the study period and showed a particularly strong Time-trend ($R^2 = 0.883$; Table 2). Positive Δ values were also common for Al (92% of years) and Na^+ (76%), whereas Ca^{2+} showed a more balanced distribution, with positive effects in 54% of years and no significant Time-trend.

Climatic parameters explained a substantial proportion of the interannual variability in Δ at Storgama, with 11 correlations exceeding the $R^2 > 0.20$ threshold (Table 4). Maximum runoff was negatively correlated with Ca^{2+} , although this relationship was strongly influenced by an extreme event in 1987 and should therefore be interpreted with caution.

Table 4. Significant ($p < 0.05$) coefficients of determination (R^2 , shown only for $R^2 > 0.2$) between annual inferred climate effects (Δ) on water chemistry (Ca^{2+} , Na^+ , NO_3^- , Org^- , Al, pH, and ANC) and climatic parameters for the period 1986–2020 at Storgama. No climatic variables explained more than 20% of the variation in Mg^{2+} and K^+ . Likewise, precipitation, as well as maximum and minimum temperature and minimum runoff, did not exceed this explanatory threshold. Red and blue coefficients indicate positive and negative correlations, respectively.

Storgama		Ca^{2+}	Na^+	NO_3^-	Org^-	Al	pH	ANC
Temperature	Average		0.299		0.371		0.305	0.252
	Median		0.228		0.348		0.497	0.202
Runoff	Average			0.207				
	Median						0.258	
	Maximum	0.271 *						

Note: * Poorly robust due to an outlier.

Higher temperatures were associated with positive inferred climate effects on Na^+ (Figure 6a), consistent with enhanced biological activity and, consequently, increased weathering under warmer conditions, leading to increasing Na^+ mobilisation from the albite-rich granite and gneiss-derived till material. The negative Δ on NO_3^- weakened with increasing runoff (Figure 6b), possibly reflecting reduced residence time and limited nitrate processing. Positive inferred climate effects on Org^- increased with temperature (Figure 6c), potentially reflecting enhanced biological activity and increased DOM production. Conversely, the negative inferred climate effect on pH became stronger with increasing temperature (Figure 6d), consistent with increased production of organic acids.

At Storgama, where pH remains below 5.5, and TOC concentrations can reach 7 mg C L^{-1} , DOM dynamics exert strong control on ANC. Indeed, variations in Org^- explained 81% of the variation in ANC, with ANC shifting from negative Δ in cooler years to positive Δ in warmer years characterised by enhanced DOM production.

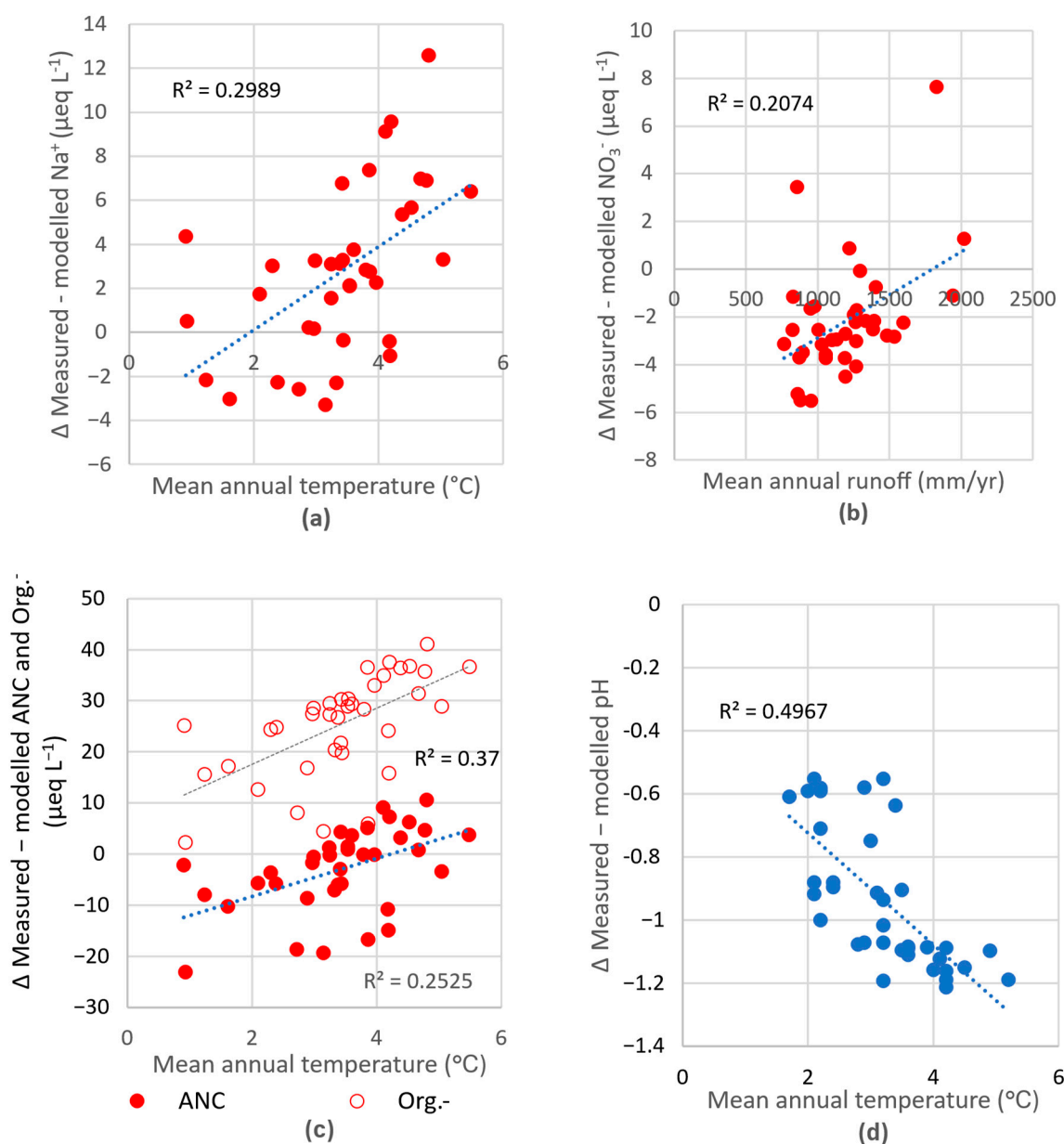


Figure 6. Significant ($p < 0.05$) correlation between inferred climate effects (Δ) on Na^+ and mean annual temperature (a), NO_3^- and mean annual runoff (b), ANC and Org^- and mean annual temperature (c), and pH and mean annual temperature (d) at Storgama (1986–2020).

3.2.3. Øygardsbekken

At Øygardsbekken, inferred climate effects (Δ) were predominantly positive, particularly for Org^- , Al, and Ca^{2+} , which exhibited positive Δ values in 94%, 87%, and 84% of the years, respectively. For Org^- and Ca^{2+} , this pattern was also reflected in significant positive Time-trends (Table 2). In contrast, pH exhibited a weak but persistent negative inferred climate effect without a significant Time-trend. Although ANC showed negative Δ values in 71% of the years, its Time-trend was weakly positive, indicating a gradual increase in recovery over time.

Climatic variables generally exhibited relatively limited explanatory power at this site, with only seven correlations exceeding the $R^2 > 0.20$ threshold (Table 5). As at Kårvatn, higher runoff was associated with reduced inferred climate effects on Ca^{2+} and Mg^{2+} (Figure 7a), consistent with shorter water residence times and reduced weathering intensity.

Table 5. Significant ($p < 0.05$) coefficients of determination (R^2 , shown only for $R^2 > 0.2$) between annual inferred climate effects (Δ) on water chemistry (Ca^{2+} , Mg^{2+} , and NO_3^-) and climatic parameters for the period 1986–2020 at Øygardsbekken. No climatic variables explained more than 20% of the variation in Na^+ , Org^- , Al, pH, or ANC. Likewise, median and maximum precipitation and temperature and maximum runoff did not exceed this explanatory threshold. Red and blue coefficients indicate positive and negative correlations, respectively.

Øygardsbekken		Ca^{2+}	Mg^{2+}	NO_3^-
Temperature	Average			0.300
Precipitation	Average			0.251
Runoff	Average	0.237	0.259	0.213
	Maximum	0.220	0.240	

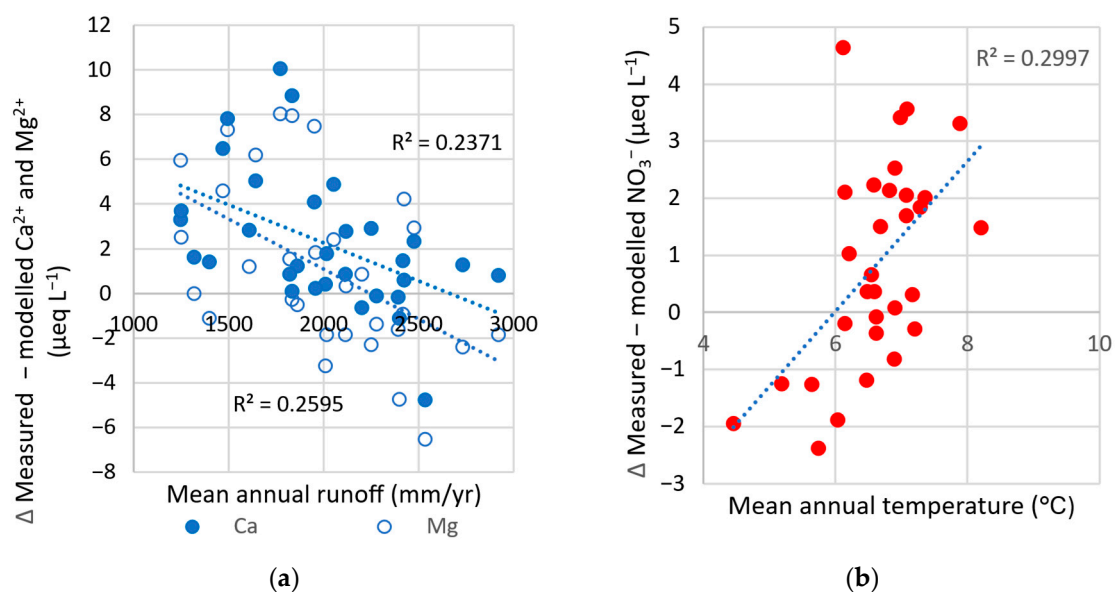


Figure 7. Significant ($p < 0.05$) correlation between inferred climate effects (Δ) on Ca^{2+} and Mg^{2+} and mean annual runoff (a), and between inferred climate effects on NO_3^- and mean annual temperature (b) at Øygardsbekken (1986–2020).

The inferred climate effect on NO_3^- was positively related to temperature, precipitation, and runoff, with temperature showing the strongest relationship ($R^2 = 0.30$; Table 5; Figure 7b). This finding is somewhat unexpected, as higher temperatures would typically enhance biological uptake and thereby reduce NO_3^- concentrations. Several mechanisms may explain the positive relationship between temperature and residual NO_3^- concentrations at Øygardsbekken. Higher temperatures may enhance mineralisation and nitrification rates, thereby increasing nitrate production. Reduced snow accumulation [8] and altered winter hydrology may also increase nitrate export, as reported by de Wit et al. [11]. Because these mechanisms may operate simultaneously, the observed relationship should be interpreted with caution.

3.2.4. Langtjern

At Langtjern, inferred climate effects were generally balanced, with approximately equal numbers of positive and negative years across the analysed variables (Figure 2). Na^+ , Org^- , and Al exhibited the highest proportions of positive Δ (60–66% of years), whereas K^+ and Mg^{2+} were predominantly negative, occurring in 69% and 77% of the years, respectively.

Climatic variables exhibited limited explanatory power for interannual variation in Δ at this site. Only one significant Time-trend was identified, namely a negative trend for Mg^{2+} (Table 2). Likewise, only seven correlations with climate parameters exceeded the $R^2 > 0.20$ threshold (Table 6), all of which were negative. These results suggest that increased precipitation and runoff are associated with negative Δ on Ca^{2+} , Mg^{2+} , Na^+ , and ANC.

Table 6. Significant ($p < 0.05$) coefficients of determination (R^2 , shown only for $R^2 > 0.2$) between annual inferred climate effects (Δ) on water chemistry (Ca^{2+} , Mg^{2+} , Na^+ , and ANC) and climatic parameters for the period 1986–2020 at Langtjern. No climatic parameters explained more than 20% of the variation in NO_3^- , Org^- , Al, or pH. Furthermore, median and maximum precipitation, temperature, and runoff, as well as mean annual temperature, did not exceed this explanatory threshold. Blue coefficients indicate negative correlations.

Langtjern		Ca^{2+}	Mg^{2+}	Na^+	ANC
Precipitation	Average		0.226	0.215	0.270
Runoff	Average	0.213	0.292	0.259	0.354

This pattern is consistent with the findings from the Trend Lakes, where negative inferred climate effects on ANC were primarily observed in dystrophic systems such as Langtjern (Section 3.1). As at Kårvatn, increased runoff may reduce water residence times, thereby limiting weathering of the gneiss-dominated catchment and reducing ANC (Figure 8).

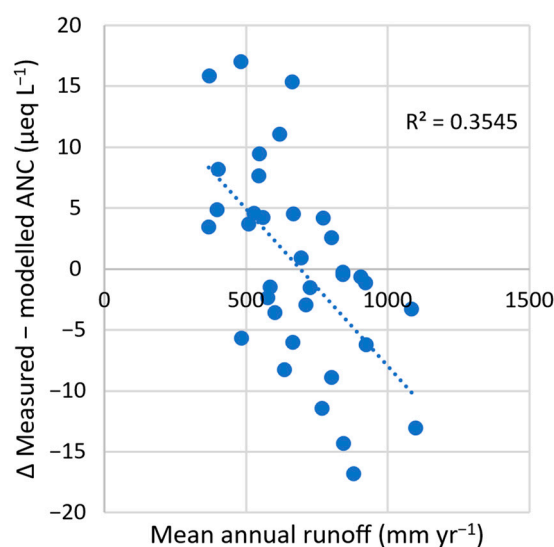


Figure 8. Significant ($p < 0.05$) correlation between modelled climate effects (Δ) on ANC and mean annual (1986–2020) runoff at Langtjern.

4. Discussion

When interpreting correlations among time series, it is important to emphasise that strong correlations do not imply causality, as multiple drivers may change concurrently over time. Moreover, the reported correlations explain only a fraction of the observed variability. In addition to declining acid deposition and climate change, several other unaccounted-for pressures are therefore likely to contribute to the residual variation, including increased biomass production and biological activity associated with warming, elevated atmospheric CO_2 concentrations, reduced grazing pressure, afforestation, and the accumulation of reactive nitrogen in soils [5]. These factors may influence weathering rates, cation dynamics,

and nutrient cycling, and may therefore contribute to deviations between measured and modelled values that are not directly attributable to climate change. Consequently, the observed correlations should be interpreted as evidence of consistency with hypothesised climate-related mechanisms rather than as proof of causation.

Furthermore, the representativeness of measured concentrations may be limited. Although samples from the Trend Lakes are collected shortly after autumn turnover, certain in-lake processes, such as nutrient assimilation and sedimentation, are not explicitly represented in the modelling framework. As a result, part of the observed deviations (Δ) may reflect processes that are not accounted for by the model.

4.1. Trend Lakes

Deviations between measured and modelled water chemistry (Δ) for Ca^{2+} , Mg^{2+} , Na^+ , K^+ , NO_3^- , Al, and ANC generally indicate positive inferred climate effects, consistent with the climate-related acceleration of recovery from acidification. Nevertheless, these relationships should be interpreted cautiously because declines in sulphate deposition and temperature increases exhibit substantial spatial covariance across Norway.

However, the MAGIC model tends to overestimate the recovery of Ca^{2+} in the most strongly acidified and warmest regions, while underestimating recovery in less affected and colder regions (Figure 3a). This pattern likely reflects limitations in model structure rather than climate effects alone. MAGIC does not explicitly account for temperature-dependent biological processes or temporal changes in DOM quality and carbonate system dynamics, both of which may influence weathering, metal complexation, ANC, and cation mobilisation.

In previous studies [13,27,36], we observed increasing Ca^{2+} concentrations in recent decades in lakes that were only weakly affected by acid deposition. We hypothesised that this increase may be driven by enhanced weathering of the Precambrian crystalline rocks associated with climate change, potentially mediated through increased primary production. Enhanced biological activity may increase the production of organic acids (e.g., root exudates) and carbonic acid, both of which promote mineral weathering.

This interpretation is further supported by the generally positive Sen's slopes and the large number of Trend Lakes exhibiting positive Time-trends in Na^+ . Compared to calcium and magnesium, sodium is less influenced by biological uptake, cation exchange, and acidification recovery processes. Consequently, long-term increases in Na^+ may provide a more robust indicator of enhanced silicate weathering than equivalent changes in divalent base cations. Collectively, these observations are consistent with the widespread climate-driven enhancement in silicate weathering processes.

4.2. Field Research Stations

Inferred climate effects on water chemistry differed substantially among the four Field Research Stations. Nevertheless, a broadly consistent pattern emerged across sites: higher temperatures were more frequently associated with positive effects on chemical recovery from acidification, expressed as increases in pH and ANC, whereas increased precipitation and runoff tended to exert negative effects through reductions in pH and ANC. These relationships were most pronounced at Kårvatn and Storgama.

Higher temperatures, together with increased biomass production, may promote enhanced weathering of predominantly silicate rocks. Weathering releases base cations that contribute positively to ANC, while weathering reactions also generate alkalinity through the consumption of weak acidity, producing bicarbonate (HCO_3^-) and organic anions (Org^-). However, increased biological uptake of base cations associated with enhanced biomass production may partly offset these effects. This may explain the weaker and more balanced climate responses observed at Øygardsbekken and particularly Langtjern.

Kårvatn and Storgama exhibit the strongest and most consistent inferred climate effects, with positive Time-trends in Org^- , ANC and Na^+ (Table 2). Positive relationships between temperature and ANC (Tables 3 and 4) suggest that warm, and often dry, years promote enhanced weathering, together with increased production of organic matter and bicarbonate, thereby strengthening ANC.

At Kårvatn, increased precipitation and runoff exerted the opposite effect (Table 3), likely through shorter water residence times that limit weathering intensity. Nitrate (NO_3^-) exhibited a pronounced negative temperature response, particularly during years with high maximum temperatures, supporting the hypothesis of enhanced biological assimilation and nitrate retention under warmer conditions.

At Storgama, Δ values were predominantly negative for pH and, in most years, for NO_3^- . Despite this, Mg^{2+} and especially ANC exhibited clear positive Time-trends (Table 2), suggesting that climate-related processes have increasingly contributed to chemical recovery from acid deposition. Concurrent positive inferred climate effects on Org^- are consistent with enhanced DOM mobilisation during warm and dry years. At this site, ANC is strongly governed by Org^- , with variation in TOC explaining 81% of its variability. Higher temperatures, indicative of longer growing seasons, likely promote biological activity and DOM production, resulting in increased inputs of organic acids and consequently lower pH. Despite the limited extent of mineral soil and associated weathering potential, this interpretation is consistent with the observed positive inferred climate effect on Na^+ under warmer conditions (Table 4).

Øygardsbekken exhibited relatively weak climate-related relationships. Inferred climate effects were predominantly positive for Ca^{2+} , Al, and Org^- , and positive Time-trends were observed for both Ca^{2+} and Org^- (Table 2). Although Δ values were negative for pH in all years and for ANC in 71% of years, the Time-trend for pH was negligible, while ANC exhibited a weak positive Time-trend. This suggests that climate-related processes contribute only modestly to chemical recovery at this hydrologically responsive site. Years with high runoff were generally associated with weaker inferred climate effects on Ca^{2+} and Mg^{2+} (Table 5), consistent with reduced weathering resulting from shorter water residence times. In contrast, positive inferred climate effects on NO_3^- during warm years differ from observations at Kårvatn and from expectations based on enhanced biological uptake. The underlying cause remains uncertain but may involve hydrological or microbial responses associated with reduced snow cover [9], including changes in organic matter decomposition, nitrification, or denitrification.

Langtjern exhibited the weakest inferred climate effects among the four case study catchments, with approximately equal numbers of years showing positive and negative responses across the analysed variables. Consequently, few Time-trends exceeded the explanatory threshold, except for a weak negative trend in Mg^{2+} (Table 2). As observed at the other sites, increased precipitation and runoff were consistently associated with negative inferred climate effects on several chemical variables (Table 6). This may partly reflect reduced mineral weathering resulting from shorter water residence times, but changes in hydrological flow paths, groundwater contributions, biological uptake, and organic matter mobilisation are also likely to contribute. The relative importance of these processes probably varies among chemical species and among catchments.

5. Conclusions

Climate-related hydro-biogeochemical processes are found to influence freshwater chemistry in Norway to varying degrees and, in some cases, in opposing directions. This assessment assumes that MAGIC provides a realistic representation of the effects of declining acid deposition on water chemistry after 1995. Across most Trend Lakes, inferred climate

effects are associated with increases in base cations (except K^+), pH, organic anions (Org^-), and/or bicarbonate (HCO_3^-), resulting in increased acid-neutralising capacity (ANC). The results further indicate that climate-related processes are becoming increasingly important drivers of long-term changes in water chemistry and should therefore receive greater attention in future monitoring programmes and assessments of environmental change.

The Field Research Stations illustrate how these responses differ among catchments. At Kårvatn, a large catchment (24.4 km^2) minimally affected by acid deposition, rising temperatures are consistent with enhanced chemical recovery through intensified weathering of Precambrian crystalline rocks, potentially driven by increased biological activity and the associated production of carbonic acid. In contrast, increased precipitation and runoff are consistent with reduced recovery, likely through dilution and shorter water residence times that limit weathering.

At Storgama, a more acidic mountain catchment characterised by exposed bedrock and shallow mineral soils, higher temperatures appear to promote the mobilisation of organic anions, contributing to lower pH. Nevertheless, ANC increases over time, consistent with enhanced weathering and an overall positive effect on recovery.

At Øygardsbekken, a humid coastal catchment, climate-related responses appear to be governed primarily by hydrological processes. Changes in flow paths and water residence times dominate over weathering and biological uptake, resulting in relatively weak and partly ambiguous responses in water chemistry.

At Langtjern, a peat-rich dystrophic catchment, climate-related responses appear to be strongly governed by biological processes. Increased production of organic acids and biological uptake of base cations tend to reduce ANC during warm and dry years, thereby slowing recovery from acidification.

Overall, the results demonstrate that climate-related influences on water chemistry are strongly modulated by catchment-specific characteristics, including sensitivity to acidification, catchment size, soil properties, climate, vegetation, and baseline water chemistry, particularly pH, TOC, and ANC:

- In less acid-sensitive catchments with long water residence times and active weathering, inferred climate-related effects are predominantly positive, leading to increased ANC and accelerated recovery.
- In catchments with shallow mineral soils, warming primarily enhances organic acid production and lowers pH, while still promoting some silicate weathering, resulting in modest increases in ANC.
- In regions with high precipitation and rapid hydrological response, hydrological processes tend to counteract temperature effects, producing weaker and less consistent chemical responses.
- In organic-rich, dystrophic catchments, increased production of organic acids and biological uptake tend to slow recovery from acidification.

These contrasting responses highlight that climate-related influences on water chemistry are not uniform and cannot be attributed solely to climate drivers. Instead, they emerge from interactions among climatic, pedological, hydrological, and biogeochemical processes.

Finally, this analysis does not explicitly account for changes in DOM because TOC was not included in the modelling framework, and substantial uncertainty remains in the simulation of Org^- . Forest biomass in boreal regions has increased in recent decades but may stabilise or decline towards the middle of this century in response to climate change [37]. Consequently, the implications of future changes in biomass production for DOM dynamics and freshwater chemistry remain uncertain.

Author Contributions: Conceptualisation, R.D.V., M.D.N., K.A., H.A.d.W. and Ø.K.; methodology, R.D.V. and M.D.N.; software, M.S.S. and M.D.N.; validation, M.D.N. and Ø.K.; formal analysis, R.D.V.; investigation, R.D.V.; resources, R.D.V.; data curation, M.S.S.; writing—original draft preparation, R.D.V.; writing—review and editing, R.D.V., K.A., H.A.d.W. and Ø.K.; visualisation, R.D.V.; supervision, R.D.V.; project administration, R.D.V.; funding acquisition, R.D.V. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Norwegian Environment Agency (project number 21087345). This research was also funded by a minor contribution from the Research Council of Norway (contract number 342628/L10).

Data Availability Statement: The data used in this analysis may be sourced from <https://vannmiljo.miljodirektoratet.no/> and <https://api.nve.no/doc/gridtimeseries-data-gts/> (both accessed on 18 December 2025).

Acknowledgments: Numerous individuals, too many to list here, have dedicated considerable time and effort over two decades to carry out water sampling, meticulously manage laboratory procedures and analyses, and oversee database management. Their collective contributions have been instrumental in generating the high-quality monitoring data upon which this work depends. During the preparation of this manuscript, the authors used ChatGTP (OpenAI; GPT-5.2) for language editing, translation, and assistance with formatting. The authors have reviewed and edited the output and take full responsibility for the content of this publication.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

ANC	acid-neutralising capacity
DOM	dissolved organic matter
MAGIC	The Model of Acidification of Groundwater In Catchments
MKT	Mann–Kendall test
Org [−]	organic anions
TOC	total organic carbon

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