

Spatial and temporal variation in Arctic freshwater chemistry—Reflecting climate-induced landscape alterations and a changing template for biodiversity

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Abstract

1. Freshwater chemistry across the circumpolar region was characterised using a pan-Arctic data set from 1,032 lake and 482 river stations. Temporal trends were estimated for Early (1970–1985), Middle (1986–2000), and Late (2001–2015) periods. Spatial patterns were assessed using data collected since 2001.
2. Alkalinity, pH, conductivity, sulfate, chloride, sodium, calcium, and magnesium (major ions) were generally higher in the northern-most Arctic regions than in the Near Arctic (southern-most) region. In particular, spatial patterns in pH, alkalinity, calcium, and magnesium appeared to reflect underlying geology, with more alkaline waters in the High Arctic and Sub Arctic, where sedimentary bedrock dominated.
3. Carbon and nutrients displayed latitudinal trends, with lower levels of dissolved organic carbon (DOC), total nitrogen, and (to a lesser extent) total phosphorus (TP) in the High and Low Arctic than at lower latitudes. Significantly higher nutrient levels were observed in systems impacted by permafrost thaw slumps.
4. Bulk temporal trends indicated that TP was higher during the Late period in the High Arctic, whereas it was lower in the Near Arctic. In contrast, DOC and total nitrogen were both lower during the Late period in the High Arctic sites. Major ion concentrations were higher in the Near, Sub, and Low Arctic during the Late period, but the opposite bulk trend was found in the High Arctic.
5. Significant pan-Arctic temporal trends were detected for all variables, with the most prevalent being negative TP trends in the Near and Sub Arctic, and positive trends in the High and Low Arctic (mean trends ranged from +0.57%/year in the High/Low Arctic to –2.2%/year in the Near Arctic), indicating widespread nutrient enrichment at higher latitudes and oligotrophication at lower latitudes.

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6. The divergent P trends across regions may be explained by changes in deposition and climate, causing decreased catchment transport of P in the south (e.g. increased soil binding and trapping in terrestrial vegetation) and increased P availability in the north (deepening of the active layer of the permafrost and soil/sediment sloughing). Other changes in concentrations of major ions and DOC were consistent with projected effects of ongoing climate change. Given the ongoing warming across the Arctic, these region-specific changes are likely to have even greater effects on Arctic water quality, biota, ecosystem function and services, and human well-being in the future.

KEYWORDS

biogeochemistry, eutrophication, lakes, oligotrophication, rivers

1 | INTRODUCTION

Warmer temperatures, associated with anthropogenic climate change, are having profound effects on Arctic environments (Frey & McClelland, 2009; Hobbie et al., 1999; Kendrick et al., 2018; Roberts et al., 2017). Freshwater ecosystems are intimately linked to their catchments (Hynes, 1975) and diverse and abundant throughout the circumpolar region (Vincent & Laybourn-Parry, 2008; Wrona et al., 2013). Arctic landscapes are affected by a multitude of anthropogenic pressures, such as direct pollution, atmospheric deposition, and climate-induced changes to hydrologic regimes as well as permafrost thaw and increasing terrestrial vegetation growth (Culp et al., 2012 and references therein), all of which are predicted to cause substantial changes to the physical/chemical habitats of both terrestrial and freshwater organisms across the region. Arctic freshwater ecosystems are generally nutrient-poor and relatively pristine (Vincent & Laybourn-Parry, 2008; Wrona et al., 2013), and are thus susceptible to changes in water chemistry due to these anthropogenic pressures, including altered inputs of nutrients and ions (Prowse et al., 2006b).

An important environmental driver of change in Arctic surface waters is the thawing of ground ice as temperature and precipitation increase (Kokelj et al., 2015). Climate change scenarios suggest that by 2100 the degree of warming in the Arctic will be sufficient to cause a large reduction in the areal extent of the upper 2–3 m of permafrost (Schuur et al., 2013). Permafrost thawing can lead to increased sediment and nutrient loading to aquatic environments and concurrent habitat disturbance (Chin et al., 2016; Hobbie et al., 1999; Kendrick et al., 2018; Kokelj, Zajdlik, & Thompson, 2009; Kokelj et al., 2013). Projected increases in precipitation in the Arctic (Bintanja & Andry, 2017) will be likely to increase hydrological connectivity between terrestrial and aquatic environments, which will also lead to greater material fluxes from land to receiving waters (Kokelj et al., 2015).

The consequences of these widespread and profound changes for water quality are uncertain and may show large regional variations. Amon et al. (2012) have suggested that widespread loss of

permafrost may increase dissolved organic carbon (DOC) export to the Arctic Ocean as terrestrial primary productivity and hydrological connectivity increases. However, Striegl et al. (2005) showed that DOC export decreased significantly in the Yukon River (Alaska, U.S.A.), and that these changes were correlated with loss of permafrost in the catchment. It has also been hypothesised that concentrations of major ions (e.g. calcium, magnesium, sulfate) will increase as the active layer (the top layer of the permafrost that thaws annually) deepens and thermokarst processes increase, exposing previously frozen mineral soils (Frey & McClelland, 2009; Vonk et al., 2015). There is already some evidence for the impacts of active layer deepening and thermokarst processes in Alaska (Keller et al., 2010), Canada (Chin et al., 2016; Kokelj et al., 2009, 2013; Levenstein et al., 2018), and Siberia (Frey et al., 2007; Tank et al., 2012). Changes in biogeochemical processes and water quality will ultimately affect the physico-chemical habitat of aquatic flora and fauna and induce changes in their community composition (e.g. Johnson & Goedkoop, 2002).

Climate change may both increase and decrease nutrient concentrations in Arctic freshwaters. Large-scale catchment processes that contribute to reductions in nutrient export to lakes and rivers can be linked to the widespread increase in terrestrial vegetation known as the *greening of the Arctic* (Elmendorf et al., 2012; Hayden et al., 2019; Pouliot et al., 2009). This process is mediated by warmer temperatures, increased nutrient uptake and storage in rooted plants (Elmendorf et al., 2012; Hayden et al., 2019; Jonard et al., 2015), as well as more efficient trapping of phosphorus (P) in soils as a result of increased soil pH induced by declines in acid precipitation (Geelhoed et al., 1997; Huser et al., 2018). In parts of the southern Arctic, declines in sulfate deposition since the 1980s may increase available soil P-binding sites due to less anionic competition (Geelhoed et al., 1997). Declines in acidic deposition in these regions may also result in increased formation of P-binding (oxyhydroxide) metals that can limit P mobility in the catchment and subsequent transport to lakes (Huser et al., 2018; Kopáček et al., 2015). However, changes in catchment hydrology (Crossman et al., 2016), permafrost thawing and soil/sediment sloughing (Kendrick et al., 2018), as well

as increases in internal loading (i.e. release of sediment nutrients; Jensen & Andersen, 1992), may also lead to higher surface water nutrient concentrations and subsequent algal blooms.

Browning related to increasing surface water DOC concentrations has been reported across much of the northern hemisphere and has been ascribed to recovery from acidification (Monteith et al., 2007) and a changing climate (de Wit et al., 2016; Haaland et al., 2010; Hayden et al., 2019; Huser et al., 2011; Kendrick et al., 2018). If its stoichiometry does not change, increasing dissolved organic matter fluxes from catchments should also increase rates of external loading of associated nutrients to surface waters, but this has clearly not been the case in other studies documenting oligotrophication and declining P concentrations (Arvola et al., 2011; Huser et al., 2018).

Water quality of Arctic freshwaters is expected to reflect the impacts of greening and browning as well as permafrost thaw. General predictions for the Arctic suggest that climate change will contribute to increases in nutrient, carbon, and ion levels in freshwaters, with subsequent effects on biological assemblages. Here, we report pan-Arctic patterns in surface water chemistry across a latitudinal gradient from the Near Arctic to High Arctic using data from 1,514 sites (1,032 lakes and 482 rivers). Our objectives were to characterise the chemistry of freshwaters across the latitudinal and longitudinal expanse of the Arctic, to describe the abiotic template for Arctic freshwater communities, and to identify temporal trends in water chemistry that could be indicative of an ecosystem response to a changing climate.

2 | METHODS

2.1 | Study sites

This study is part of the first circumpolar assessment of Arctic freshwater biodiversity conducted by the Freshwater Group of the Circumpolar Biodiversity Monitoring Program, part of the Conservation of Arctic Flora and Fauna (CAFF) working group of the Arctic Council. As part of this effort, scientists from all Arctic countries compiled a database of biotic and abiotic data for freshwaters across the circumpolar region. Data sources included national and regional monitoring programmes, academic research, published reports, and industry monitoring (Lento et al., 2019).

The sites included are located across the circumpolar region in Canada, Greenland, the Faroe Islands, Finland, Norway, Sweden, Russia, and the U.S.A. Water bodies within either the CAFF boundary (politically-derived; www.caff.is) or the boundary of the Arctic Biodiversity Assessment (ABA, based on climate and vegetation; Meltotte, 2013) were considered for analysis (Figure 1). Data from 1,514 monitoring sites (1,032 lake and 482 river sites) were included in the analysis (Table S1). Because of the large number of water bodies included in the analysis, catchment areas were not delineated and were instead estimated by using a global hydrobasin GIS layer (Lehner & Grill, 2013). Hydrobasins are flow-based standardised catchments derived at spatial scales ranging from continental (level 01) to sub-basin (level 12) scales. The mid-range level 07 hydrobasins were used as surrogate catchments for all water bodies.

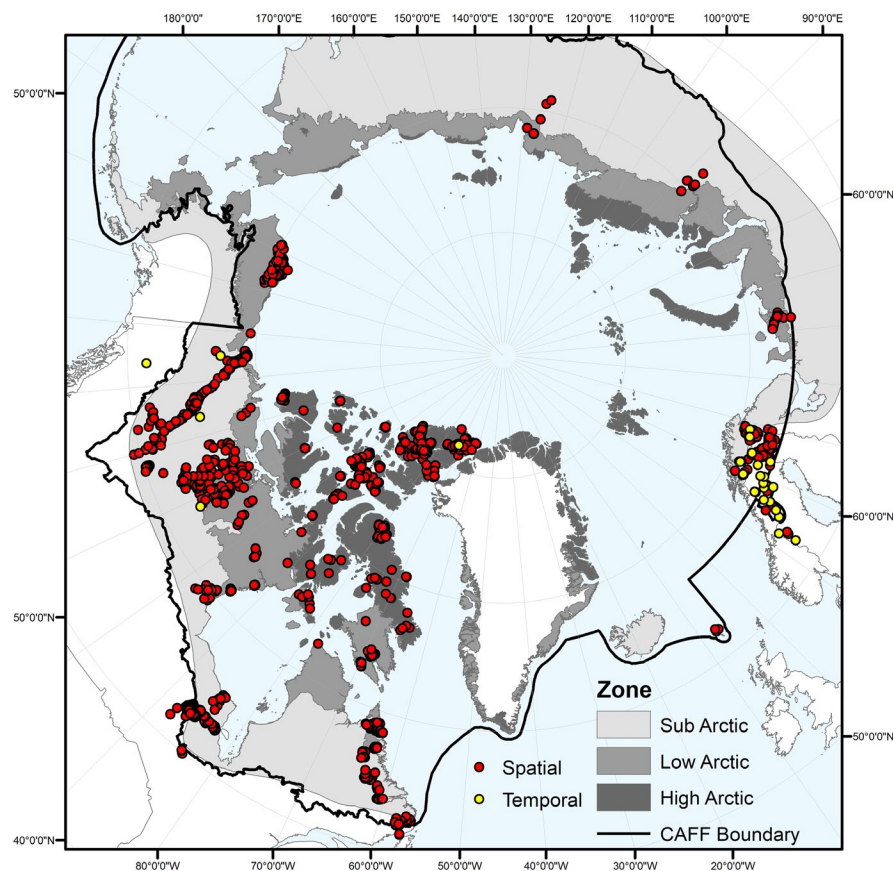


FIGURE 1 Location of monitoring sites included in the study. Arctic Biodiversity Assessment (ABA boundaries) are shaded with light (Sub Arctic), medium (Low Arctic), and dark grey (High Arctic). Sites outside ABA defined boundaries were classified as Near Arctic

Sites were classified, in order from south to north, as belonging to Near Arctic, Sub Arctic, Low Arctic, and High Arctic regions, with the latter three classifications from the Arctic zones defined in the ABA (Meltotte, 2013; Figure 1). We have attempted to avoid bias in the zonal analysis by (1) having evenly distributed and similar number of sites from east to west across the zones (Figure 1), and (2) comparing temporal trends in different regions. Climate and land cover varied substantially across regions (Table 1). Regional classification of water bodies was based on catchment location (as opposed to location of the water body itself). To be categorised as Sub, Low, or High Arctic, the catchment area had to be primarily contained in the respective region of the ABA Arctic zones. Water bodies with <50% of their catchment area within the ABA Arctic zones were categorised as Near Arctic.

2.2 | Study design

Assessments related to data quality were undertaken to produce an internally consistent and comparable data set. Although observations were available from 1960 to 2015, spatial analysis focused on data collected from 2001 to 2015 to document more recent patterns. Temporal analyses were conducted on data collected between 1970 and 2015 due to limited availability of data collected during the 1960s. Time frames for bulk temporal analysis included Early (1970–1985), Middle (1986–2000), and Late (2001–2015) periods. To reduce seasonal variability, data from June through September were used in calculations. Furthermore, only water bodies minimally

affected by urbanisation, mining, agriculture, or other direct anthropogenic pressures were included in the analysis (see Table 1). This determination was made using available land-use data and specific information relating to each site gathered by the groups responsible for the monitoring data. The information set, however, is probably not comprehensive, especially with respect to localised impacts.

Although water column profile data were available for some lakes and rivers, only surface samples (i.e. reported depths between 0 and 2 m) were used in analyses. The majority of surface samples were collected at a depth of 0.5 m; however, if there were multiple samples collected on the same date from a waterbody within the surface sample depth range, the value closest to the 0.5 m depth was chosen. In some studies, the only information available for a sample was a designation of *surface* and these values were included in the analyses as well. Data were excluded if sampling depth was ambiguous.

There were methodological differences in laboratory analysis across data sets, which resulted in differences in detection levels. We chose to retain values below the lower detection limit (LDL) from individual data sets in all analyses. An analysis of mean LDL values across regions and time periods revealed no interpretable patterns (Huser, B.J. and Futter, M.N. unpublished data). It should be noted that variables used in the study had a low percentage of below LDL values. Values below the LDL were divided by 2 and variables that had more than 10% of values below the LDL in the dataset (i.e. including all water bodies) were not analysed further (e.g. phosphate and nitrate–nitrite, Table S2). Variables were only included in the spatial analysis if measurements were available for at least 60% of the available sampling sites. To reduce redundancy in the data set,

TABLE 1 Mean climatic and catchment land cover data for the study sites in each Arctic region

Land cover/climate	High Arctic		Low Arctic		Sub Arctic		Near Arctic	
	Mean	SE	Mean	SE	Mean	SE	Mean	SE
Barren or sparsely vegetated	3.4	0.22	1.0	0.19	0	0	0	0
Closed shrublands	0	0	0	0	0.16	0.16	0	0
Croplands	0	0	0	0	0	0	0	0
Deciduous needleleaf forest	0	0	0.6	0.11	0.02	0.00	0	0
Evergreen needleleaf forest	0	0	0	0	23.1	0.80	1.7	0.24
Grasslands	37.4	0.88	20.4	0.93	14.7	0.36	4.6	0.09
Mixed forest	0	0	0	0	9.0	0.31	0.3	0.14
Open shrublands	5.7	0.53	71.1	0.96	32.3	0.34	26.1	0.44
Permanent wetlands	0	0	2.3	0.27	22.3	1.61	75.0	2.86
Savannas	0	0	4.8	0.20	3.1	0.18	0.33	0.04
Snow and ice	51.3	0.94	0.03	0.02	3.9	0.07	0.43	0.03
Urban and built-up	0	0	0	0	0	0	0	0
Water	9.9	0.28	9.3	0.32	8.5	0.22	2.4	0.09
Woody savannas	0	0	2.4	0.67	36.2	0.49	72.5	0.43
Mean annual precipitation (mm)	14.3	0.29	22.3	0.34	39.9	0.20	49.7	0.16
Mean annual temp (°C)	−16.1	0.10	−8.3	0.07	−2.3	0.04	0	0.01

variables that represented measurement of similar compounds and correlated well with each other (e.g. absorbance and DOC, $r > 0.8$) were compared and the variable with the fewest data was excluded.

2.3 | Data analysis

Throughout, we refer to weak or strong statistical significance depending on whether or not the nominal p -value for a test was above or below 0.05. We did this as in most cases, p -values were not adjusted for multiple comparisons so the nominal p -value is likely to be an over-estimate of the true significance. The p -values were only adjusted for post hoc tests of differences in regional and temporal differences in water chemistry. We focus on presenting and discussing results with strong statistical significance. However, when consistent, weakly significant temporal trends were found, these are also discussed.

2.3.1 | Spatial patterns

There were 917 sites with water chemical data collected between 2001 and 2015 (Late Period), although not all variables were measured at all sites. Spatial analysis of these more recent data focused on samples collected during the ice-free period, and a mean value for June–September was calculated for each variable at each site. Lake and river data were analysed together to maximise sample size for each Arctic region. Spatial analysis focused on comparison of means among Arctic regions (Near, Sub, Low, and High Arctic) for pH, alkalinity, conductivity, major ions (calcium, magnesium, sodium, sulfate, chloride, and silica), carbon (DOC), and major nutrients (total nitrogen [TN], total phosphorus [TP]). Data were transformed (log base 10) prior to analysis. One-way ANOVAs were performed to compare concentrations among regions, with application of the Tukey–Kramer HSD test (Kramer, 1956; Tukey, 1953) at a significance level of 0.05. This test was used because sample size was not consistent between groups and the Tukey–Kramer comparison is conservative in such cases (Hayter, 1984).

A selection of 31 slump-impacted stream sites from the Northwest Territories, Canada (Chin et al., 2016) was used to characterise differences in the water chemistry of slump-impacted and un-impacted systems. Slump-impacted sites were downstream of one or more ongoing permafrost thaw slump disturbances. All sites were compared using data from the Late Period. Using the same suite of variables, means from slump-impacted sites were compared with means from Near, Sub, Low, and High Arctic sites.

2.3.2 | Temporal trends

Temporal analyses included both an assessment of bulk trends for all sites sampled between June and September from 1970 to 2015, and trend analyses of sites with time-series data. Both types of

temporal analyses focused on the same set of variables used in the spatial analysis. Analysis of bulk trends was intended to give a general picture of regional patterns across different time periods (Early, Middle, and Late periods). Time periods were compared separately for each Arctic region (Near, Sub, Low, and High Arctic) using one-way ANOVA with a Tukey–Kramer HSD post hoc test. Due to limited data, only the Middle and Late periods could be compared in the Low and High Arctic region.

Time-series analysis provided detailed evidence of site-specific temporal changes in water chemistry. Twenty-seven sites from Canada, Finland, and Sweden with consistent data for at least 20 years of continuous observations between 1970 and 2015 were analysed for temporal trends. Monthly data were available for all 27 sites included in time-series analyses; however, some stations had multiple observations available during some months. To reduce sampling bias for these stations, a monthly centre analysis was conducted. Using this method, only the sample taken closest to the middle of each month was used in the analysis. Mean or median values for months with multiple sampling events were not used because this could influence the variance (Helsel & Hirsch, 2002). Time-series data were analysed using the Mann–Kendall trend test (Kendall, 1975; Mann, 1945). Nominally significant monotonic trends ($p \leq 0.05$) for surface water variables were determined and Sen's-slope (Sen, 1968) trend estimates (unit/year) were calculated for each water body. Slopes were then used to estimate relative annual changes (trend/mean as %/year) for each site.

3 | RESULTS

3.1 | Spatial patterns

Substantial spatial and temporal variability in surface water chemistry was observed. In the following, only strongly significant spatial patterns (nominal p -value ≤ 0.05) are reported unless otherwise noted. Many variables exhibited similar south to north patterns (Figure 2), with higher values generally detected in the High Arctic (northern-most) and lower values in the Near Arctic (southern-most) region. For example, pH was higher in the High Arctic (mean = 7.9) than in all other regions, and lower in the Near Arctic (mean = 7.1) than in all other regions (Table S3). Values were similar for pH in the Sub and Low Arctic regions (Figure 2). Alkalinity, conductivity, base cations, as well as chloride and sulfate followed similar patterns, with more dilute, lower alkalinity sites generally found in the Near Arctic. Although the region with the highest mean concentrations/values differed by variable (e.g. alkalinity was highest in the Sub Arctic, chloride was highest in the Low Arctic, and conductivity was similarly high across the Sub, Low, and High Arctic regions), values in the Near Arctic were significantly lower than in the other Arctic regions for each variable (Figure 2 and Table S3).

In contrast to many other variables, calcium and silica (silicate-Si) concentrations in the Near Arctic were only weakly lower than in all other regions. Calcium and silicate concentrations were lowest in

the Low Arctic, highest in the Sub and High Arctic, and intermediate in the Near Arctic (Figure 2 and Table S3). These variables reflected patterns in pH, alkalinity, and magnesium, all of which showed generally higher levels in the Sub and High Arctic than in other regions.

Several nutrients had lower values in the Low and High Arctic than in more southerly regions. In particular, DOC was lower in the High Arctic (mean = 2.0 mg/L) compared to the Near Arctic (mean = 7.7 mg/L) region. Mean TN concentrations were lower in the Low and High Arctic regions than Sub Arctic, though only the High Arctic was lower than the Near Arctic region. Mean TP concentrations were lower in the Low and High Arctic (mean = 7.7 and 10.6 µg/L, respectively) than in the Near Arctic (mean = 12.6 µg/L), but only the Low Arctic was notably different (Figure 2 and Table S3).

When un-impacted sites across all Arctic regions were compared with slump-impacted sites (Sub and High Arctic regions), several

variables were found to be elevated at the impacted sites. The strongest difference was found for TP, for which the mean in slump-impacted sites was over an order of magnitude higher than the mean for any Arctic region (Figure 3 and Table S4). Magnesium, sodium, conductivity, and TN were all higher in slump-impacted sites than in unimpacted sites in all Arctic regions.

3.2 | Temporal trends

3.2.1 | Time-series trends

Only strongly significant trends (nominal p -value ≤ 0.05) are reported for time series. Trends were detected for all variables, but to differing degrees and not in all regions (Table 2). Trends in TP, alkalinity, conductivity, and sulfate were observed for more than

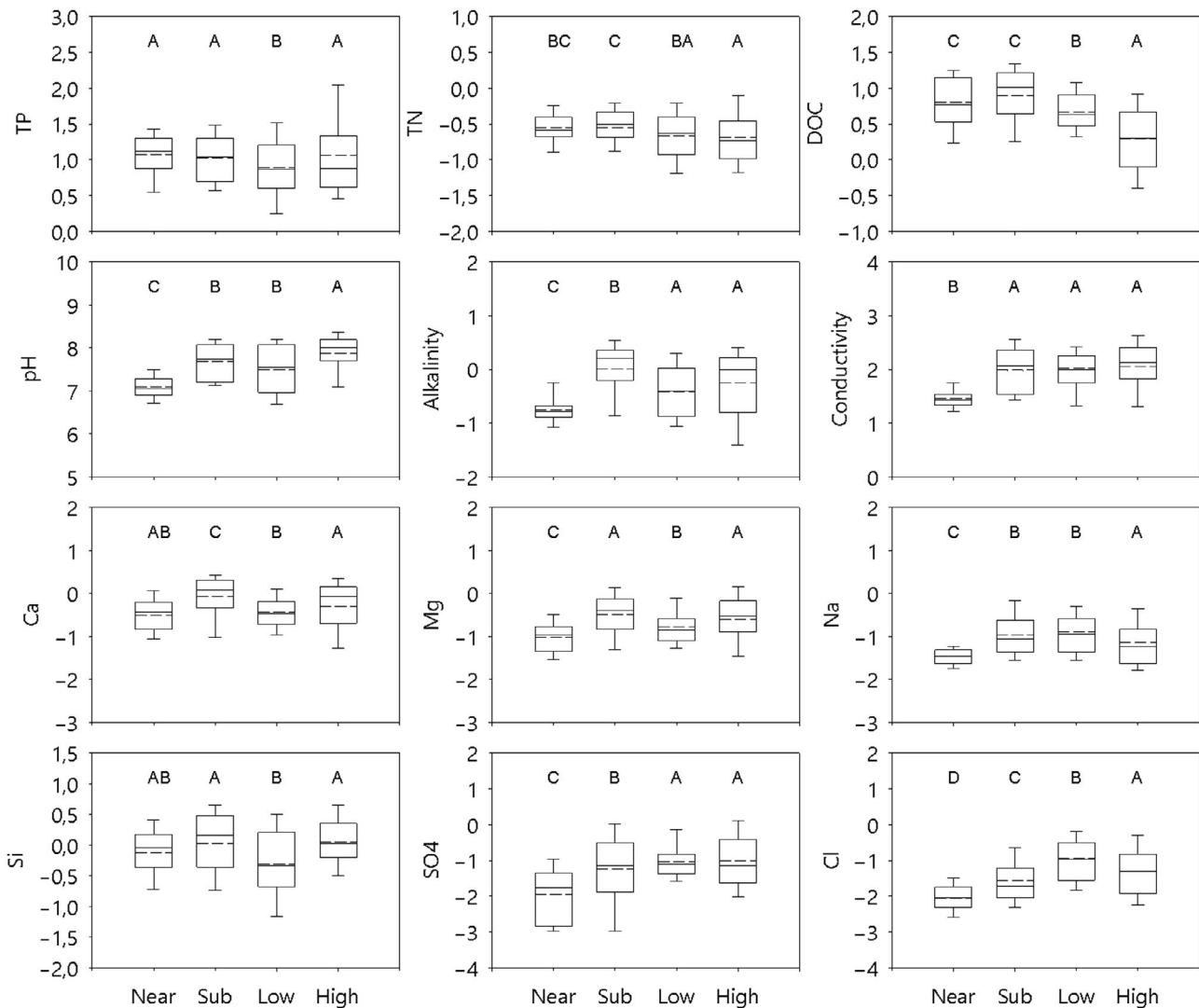


FIGURE 2 Regional boxplot comparison of water chemical variables included in the study during the Late period. Significance between regions ($p < 0.05$) was determined with the Tukey-Kramer HSD test. Different letters indicate significant differences, whereas the same letter for different regions indicates no significant difference. Means and medians are represented by horizontal dashed and solid lines, respectively

half of the sites. Mean trends across the Arctic for TP, chloride, and sulfate were negative (-2.2 and -0.15 , and $-0.32\%/year$, respectively), whereas all other water chemistry trends were positive for all regions across the Arctic. There were substantial differences between regions, however, with TP varying the most, from $+0.6\%/year$ in the Low and High Arctic to -3.6 and $-2.2\%/year$ in the Sub and Near Arctic regions, respectively. Sulfate also varied, with negative trends in the Near Arctic ($-1.9\%/year$) and positive mean trends in the Sub and Low Arctic (2.1 and $0.58\%/year$). Alkalinity, pH, TN, silica, calcium, and magnesium trends were, on average, positive; however, not all Arctic regions had strongly significant trends (e.g. calcium lacked trends in the Low and High Arctic, Table 2). The direction (positive or negative) of sodium, conductivity, and DOC trends varied, depending on the region (Table 2). There was one negative and one positive trend for chloride in the High and Sub Arctic regions, respectively.

Several profound changes in TP concentrations were observed in the time-series data. Long-term monitoring of oligotrophic Near Arctic Swedish sites showed precipitous declines in TP concentrations (nearly $6\%/year$) to ultra-oligotrophic levels (TP $< 4 \mu\text{g/L}$, Figure 4a). These declines are in contrast to TP increases observed at higher Arctic latitudes (Figure 4b).

3.2.2 | Bulk trends

Comparisons of bulk trends between Early (1970–1985), Middle (1986–2000), and Late (2001–2015) periods showed a number of pan-Arctic and regional temporal patterns in water quality that generally followed the time-series results (Figure 5 and Tables S5–S8). Total P was higher in the Late than in the Middle period (Figure 5a) in the High Arctic (11.6 and $8.4 \mu\text{g/L}$, respectively). While differences in TP between time periods were smaller for other Arctic regions, there was a consistent decrease in mean TP from Early, to Middle, to Late periods in the Near Arctic (13.5 , 13.3 , and $12.6 \mu\text{g/L}$, respectively). Concentrations of DOC showed a divergent pattern across Arctic zones, with higher mean values in the Late period compared to the Middle period in the Near Arctic (7.7 and 4.1 mg/L , respectively), and lower DOC in the Late period compared to the Middle period in the High Arctic (2.0 and 2.8 mg/L , respectively). Total N showed a similar pattern with lower concentrations in the Late period than in the Middle period in the High Arctic (0.20 and 0.37 mg/L , respectively) and in the Sub Arctic (0.29 and 0.43 mg/L , respectively).

In the Near Arctic, there was little temporal difference in pH, alkalinity, or conductivity (Figure 5b), whereas concentrations of several major ions varied over time. In particular, calcium, magnesium,

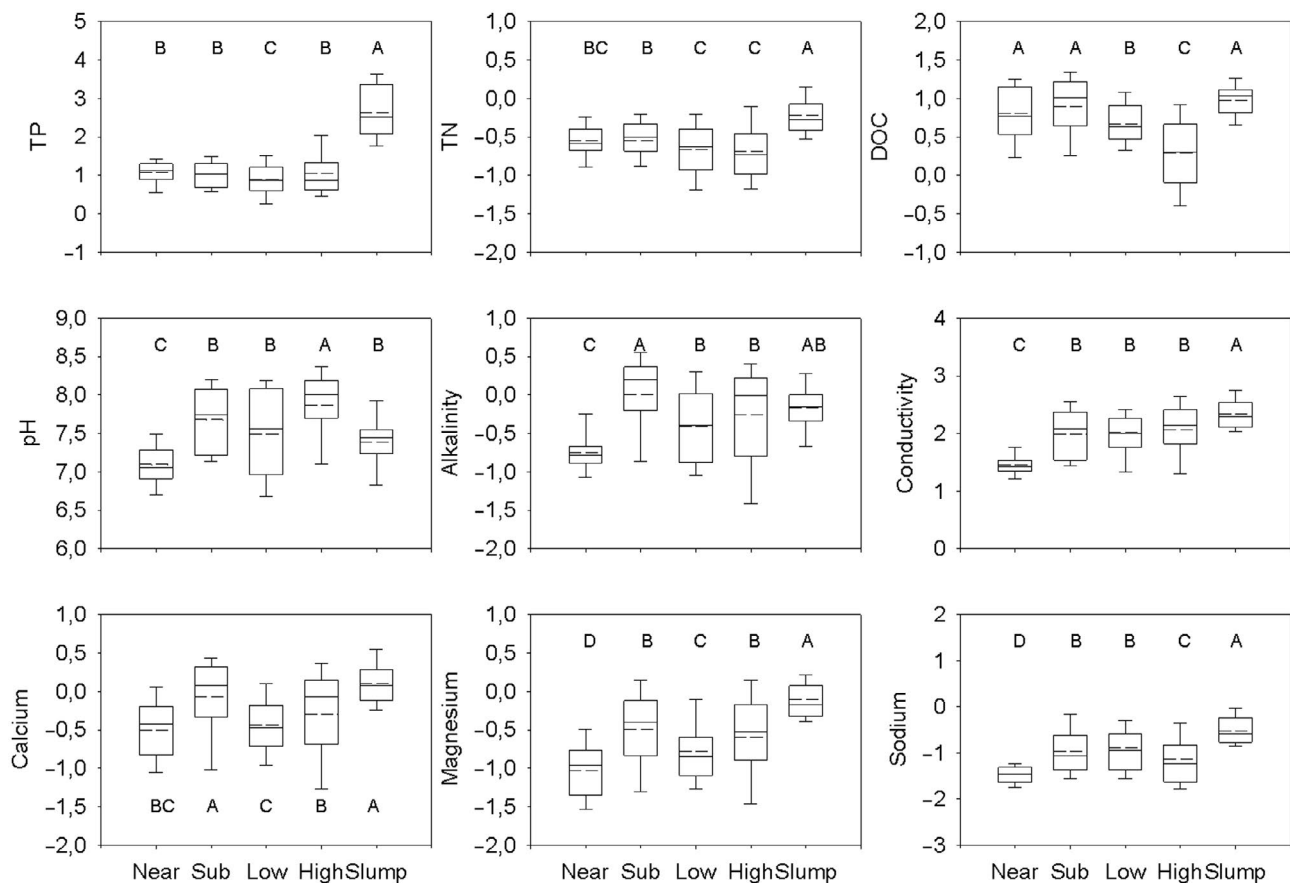


FIGURE 3 Comparison of slump affected sites with all Arctic regions (Late period). Significance between regions ($p < 0.05$) was determined with the Tukey–Kramer HSD test. Different letters indicate significant differences, whereas the same letter for different regions indicates no significant difference. Means and medians are represented by horizontal dashed and solid lines, respectively

TABLE 2 Time-series trends for sites ($n = 27$) in Canada ($n = 5$), Finland ($n = 4$), and Sweden ($n = 18$)

Variable	Units	Sites		Trend direction		All regions		High	Low	Sub	Near
		(n)	(%)	+	-	(units/year)	(%/year)	(%/year)			
pH		13	48	13	0	0.0063 (0.0026)	0.09			0.07 (4)	0.1 (9)
Alkalinity	meq/L	18	67	18	0	0.0012 (0.0006)	0.88		0.93 (1)	0.85 (6)	0.89 (11)
Conductivity	$\mu\text{S}/\text{cm}$	14	52	5	9	0.15 (0.72)	0.11	-0.42 (1)	0.46 (1)	1.16 (4)	-0.39 (8)
DOC	mg/L	8	30	6	2	0.024 (0.03)	0.39		1.12 (1)	-0.19 (4)	0.93 (3)
TN	mg/L	9	33	6	3	0.0029 (0.0031)	1.06			1.12 (4)	1.01 (5)
TP	$\mu\text{g}/\text{L}$	21	78	3	18	-0.071 (0.22)	-2.23	0.57 (1)	0.56 (1)	-3.6 (4)	-2.23 (15)
Silica	mg/L	10	37	10	0	0.016 (0.012)	1.21	0.45 (1)		0.77 (2)	1.44 (7)
Sulfate	meq/L	14	52	7	7	0.0027 (0.0069)	-0.32		0.58 (1)	2.06 (5)	-1.91 (8)
Chloride	meq/L	2	7	1	1	-0.0004 (0.001)	-0.15	-0.5 (1)		0.21 (1)	
Calcium	meq/L	5	19	5	0	0.0069 (0.0069)	0.89			1.12 (3)	0.54 (2)
Magnesium	meq/L	5	19	5	0	0.0042 (0.0056)	0.96		0.57 (1)	1.2 (3)	0.61 (1)
Sodium	meq/L	10	37	3	1	0.0007 (0.0009)	0.17			0.5 (2)	-0.16 (2)

Note: Standard deviation is given in parenthesis for raw trends (units/year) and number of sites is presented in parentheses for trends (%/year) for each region on the right.

Abbreviations: DOC, dissolved organic carbon; TN, total nitrogen; TP, total phosphorous

and sodium were all higher in the Late period than in either of the earlier periods (Figure 5c), with the most dramatic change evident for calcium (which varied from 0.04 mg/L in the Early period to 0.36 mg/L in the Late period). In contrast, sulfate and chloride differed over successive periods with lower concentrations in the Late period than in Early period. Patterns across periods were more consistent for all major ions in the other Arctic regions. In the Sub Arctic, major ion concentrations were higher in the Middle and Late periods than in the Historical (i.e. Early) period (Figure 5b,c). Calcium, magnesium, and sodium were also higher in the Late period than in the Middle period. In the Low Arctic, where historical (Early period) data were not available, all major ion concentrations were higher in the Late period compared to the Middle period. In contrast, all major ion concentrations were lower in the Late period than in the Middle period in the High Arctic.

4 | DISCUSSION

We found notable spatial and temporal differences in surface water chemistry across regions of the Arctic. Spatial analysis revealed regional differences in the chemical habitat of Arctic freshwaters, with more alkaline waters with higher ion content in the High Arctic than in Near Arctic sites. In contrast, TN and DOC were often lower in the High Arctic, while concentrations of TP were generally similar among Arctic regions. However, our temporal analysis suggests that shifts are underway in these systems, with TP increasing in the High Arctic and decreasing in parts of the Sub and Near Arctic.

Furthermore, increasing concentration trends of many major ions in the Near, Sub, and Low Arctic regions, combined with declining trends in more recent years in the High Arctic, indicated that the

chemical habitats of Arctic freshwaters are changing. For example, increases in alkalinity and Ca will favour crustaceans that need Ca for their exoskeleton (Alstad et al., 1999). Increased nutrient inputs to Arctic lakes are predicted to result in increased primary productivity, an increased number of species, as well as a shift in community composition from primary producers to top predators (Heino et al., 2009). By contrast, browning can cause decreased phytoplankton production and increased bacterial production due to reduced water transparency, and may shift Arctic freshwaters to more heterotrophic food webs (Forsström et al., 2005; Wauthy et al., 2018).

4.1 | Major ions, alkalinity, conductivity

The strongest regional differences in water chemistry were between Near Arctic and higher-latitude sites. In general, many variables, including pH, alkalinity, and major ions (calcium, magnesium, sodium, chloride, and sulfate), had higher concentrations/values in the northern-most Arctic regions than in the more southerly Near Arctic region. Several of these variables were also elevated in Sub Arctic sites, particularly pH, alkalinity, calcium, and magnesium. Ion concentrations are driven by a number of spatially variable factors, including underlying geology, weathering and flow rates, and changes to climatic drivers may influence mineral flux in Arctic systems (Petrone et al., 2006; Prowse et al., 2006b). For example, Petrone et al. (2006) noted low concentrations of several ions in areas of Alaska that were dominated by permafrost and suggested that permafrost composition or variation in runoff could have contributed to concentration differences across catchments.

Geologic differences among sampled areas have probably contributed to the higher levels of ions in the some Arctic regions (Hamilton et al., 2001), as sites sampled in the Sub and High Arctic

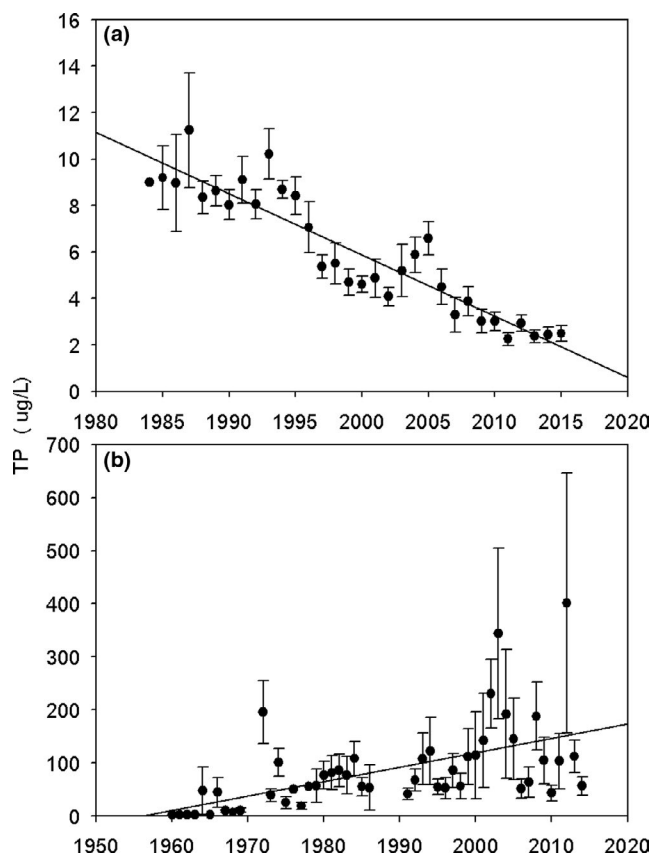


FIGURE 4 Time-series trends for total phosphorous (TP; with SE) in Laxtjærnsbäcken, Near Arctic (a) and the Mackenzie River, Low Arctic (with tributary areas also in lower latitude regions, (b). Regression lines are shown as well

were predominantly underlain by sedimentary bedrock (Harrison et al., 2011; Figure S1), which contributes to more alkaline conditions with higher concentrations of ions. In contrast, sites in the other Arctic regions (Near Arctic in particular, see Figure S1) were predominantly underlain by igneous, metamorphic, or supracrustal bedrock, including Precambrian igneous rock with low buffering capacity in eastern and central Canada (Hamilton et al., 2001; Harrison et al., 2011), which may have contributed to lower levels of ions (calcium and magnesium in particular) and alkalinity. Thus, regional variation in pH, alkalinity, and ions reflected the spatial distribution of bedrock geology types and probably represented natural gradients in the composition of parent material. Regional differences in geology probably influenced, e.g. TP where weak bulk trends followed the same, declining pattern seen in the time-series data in the Near Arctic. Regional differences between the sites across regions, including geology, probably led to natural variability in the dataset that in some cases may have overshadowed long-term changes.

Climate change is likely to lead to profound changes in hydrology and permafrost dynamics in Arctic ecosystems, altering the amount and timing of precipitation and runoff, and causing deepening of the active layer of the permafrost (i.e. the annual thaw depth), all of which will contribute to changes in the water balance, groundwater influence, and geochemical weathering (Haldorsen et al., 2010;

Kokelj et al., 2015; Prowse & Brown, 2010; Prowse et al., 2006a). In general, alkalinity and concentrations of many major ions (e.g. calcium, magnesium, sulfate) are predicted to increase markedly with permafrost degradation and lowering of the water table that will result in less interaction with organic shallow soils and enhanced interaction with deep mineral horizons (Frey & McClelland, 2009; Prowse et al., 2006a). In our study, bulk and time-series trends indicated that concentrations of several ions increased in the Near, Sub, and Low Arctic, consistent with the predicted response to the deepening of the active layer (Prowse et al., 2006a). Kendrick et al. (2018) hypothesised that thawing permafrost would allow groundwater to leach minerals and cations from newly exposed soils. This, in turn, would lead to increasing stream and river alkalinity, and cation concentrations (Frey & McClelland, 2009; Frey et al., 2007; Keller et al., 2010). Studies in Alaska have shown that permafrost degradation can result in increased delivery of major ions (Keller et al., 2007; Petrone et al., 2006; Stottlemeyer, 2001), in addition to nutrients and DOC (Abbott et al., 2014, 2015; Bowden et al., 2008; Larouche et al., 2015). In some cases, permafrost thaw and water table lowering will be likely to cause soil oxidation, which in turn will cause the mobilisation and release of elements accumulated under reducing conditions during previous soil and peatland development (ChagueGoff & Fyfe, 1997).

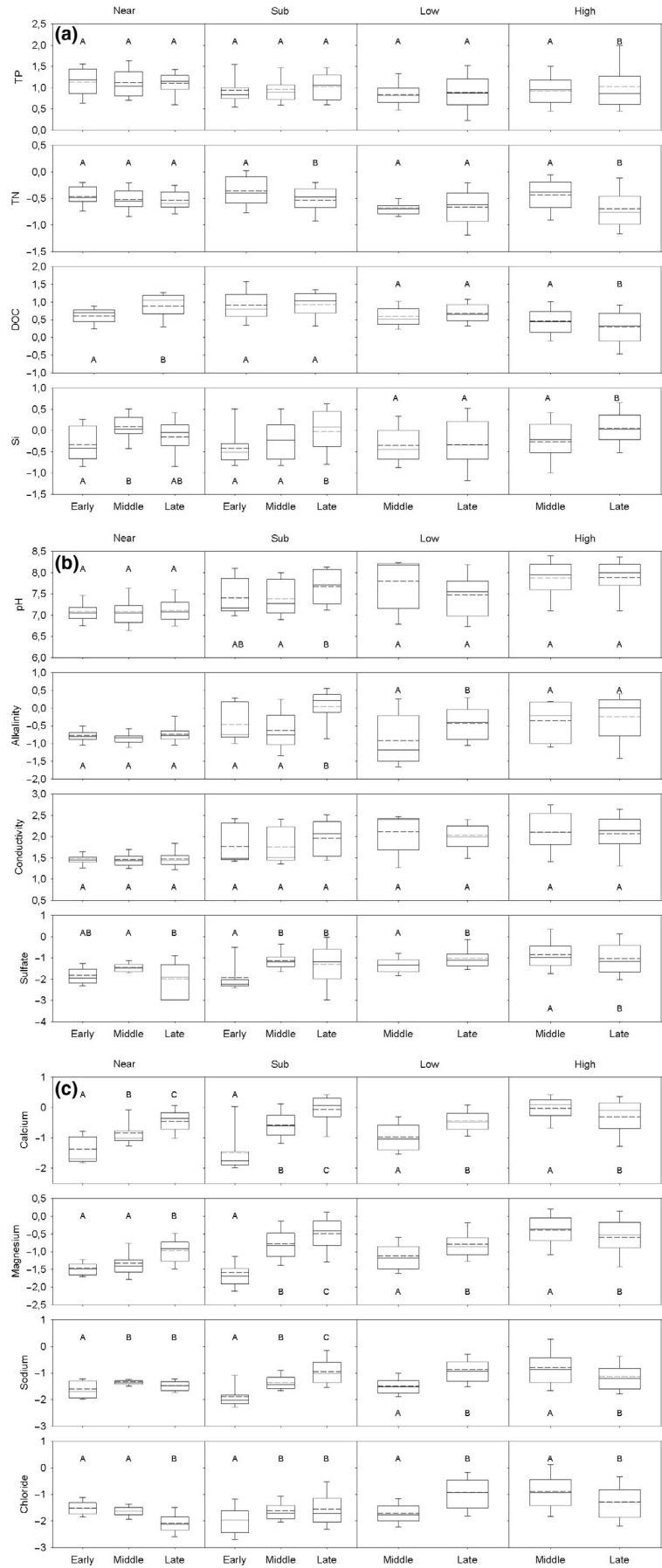
Not all patterns in major ion concentrations reported herein were consistent with the predicted response to climate change. For example, bulk trends indicated that ion concentrations were lower in the Late period in the High Arctic, and time series for conductivity and chloride both showed declining trends over time in this region. Lower sulfate concentrations in the Near Arctic in the Late period, however, were consistent with declines in acid deposition in Scandinavia (Moldan et al., 2013; Monteith et al., 2007).

4.2 | Dissolved organic carbon

As expected, a latitudinal gradient in DOC concentrations, with lower levels in the Low and High Arctic compared to the Near or Sub Arctic, was evident in our data set (Figure 2). Low levels of DOC in the Low and High Arctic regions may reflect low terrestrial inputs of carbon from terrestrial vegetation, particularly above the tree line (Lim et al., 2001). Our results are consistent with comparisons of Arctic lakes from above the tree line to lakes from boreal ecoregions or forested tundra where relatively higher DOC concentrations have been detected where trees are present (Lim et al., 2001; Pienitz et al., 1997; Rühland et al., 2003). However, several studies have found greater DOC concentrations in High Arctic systems (Hamilton et al., 2001; Keatley et al., 2007), particularly where high coverage of grasses and other vegetation has led to elevated inputs of terrestrial carbon (Lim et al., 2001; Michelutti et al., 2002).

Concentrations of DOC may increase in Arctic freshwaters under continued climate change (Prowse et al., 2006a), and our results from Near Arctic bulk trends and time series provide some support for this hypothesis. Weak increases were also detected in the

FIGURE 5 Bulk trend boxplot comparison for water chemical variables included in the study. Significance between regions ($p < 0.05$) was determined with the Tukey–Kramer HSD test. Different letters indicate significant differences, whereas the same letter for different regions indicates no significant difference. Means and medians are represented by horizontal dashed and solid lines, respectively. DOC, dissolved organic carbon; Si, silica; TN, total nitrogen; TP, total phosphorous



Sub and Low Arctic regions. Earlier assessments of how DOC may respond to climate warming in higher-latitude Arctic regions suggest that there may be an overall increase in terrestrial export from soils and peatlands (Prowse et al., 2006c), particularly with increased precipitation and runoff (de Wit et al., 2016; Hayden et al., 2019; Petrone et al., 2006). This is supported by observations from the West Siberian Lowlands where permafrost-influenced catchments have low concentrations of DOC and permafrost-free catchments have markedly higher DOC concentrations that rise sharply as a function of peatland cover (Frey & Smith, 2005). Increases in freshwater organic matter with climate change may also be a result of increasing terrestrial vegetation production associated with a longer growing season (i.e. shrubification; Myers-Smith et al., 2011; Rouse et al., 1997). Studies also indicate indirect effects of permafrost degradation causing increases in vegetation cover (Sturm et al., 2005). These changes can, in turn, impact the amount of DOC in surface waters (Wickland et al., 2007).

By contrast, several factors may instead contribute to a net decrease in DOC in Arctic freshwaters, as was observed in High Arctic sites. For example, adsorption of DOC to mineral soils may lead to declines in stream water DOC discharge (Striegl et al., 2005). Studies from Alaska (Petrone et al., 2007; Petrone et al., 2006; Striegl et al., 2005, 2007), the Yukon (Carey, 2003), and central Siberia (Kawahigashi et al., 2006; Prokushkin et al., 2007) have all suggested that the export of dissolved organic matter (and thus DOC) from Arctic catchments will decline as a result of permafrost thaw, which causes increased organic matter adsorption to the underlying mineral soil. Furthermore, the carbon released from permafrost differs chemically from that derived from vegetation or overland flow (Balcarczyk et al., 2009), which may have implications for food web ecology and ecosystem function (O'Donnell et al., 2019). Permafrost dynamics, however, are not straight-forward; both increases and decreases in organic matter export have been observed or predicted to result from climate warming and permafrost degradation.

4.3 | Nutrients

Like DOC, N and P trends followed a latitudinal gradient, with lower levels of TN in northern Arctic regions and somewhat lower TP concentrations at higher latitudes. Many areas of the Arctic are naturally oligotrophic or ultra-oligotrophic (Hamilton et al., 2001; Wrona et al., 2013), particularly in the Canadian Arctic Archipelago. Levels of C, N, and P are related to underlying geology, but are also driven by surrounding vegetation, local permafrost dynamics, and precipitation, and there is thus great potential for latitudinal variation across the Arctic (Michelutti et al., 2002).

As the limiting nutrient of many aquatic ecosystems in the Arctic, changes in the supply of P to surface waters have the potential to greatly alter the structure and function of aquatic ecosystems (Peterson et al., 1985; Reyes & Loughheed, 2015; Slavik et al., 2004). Climate change may lead to either an increase or a decrease in P concentrations in Arctic freshwaters, both of which were documented

in our study. There were strong and divergent temporal trends for freshwater TP concentrations across the Arctic, with increasing concentrations in the High and Low Arctic and declines in the Sub and Near Arctic regions (Table 2). The deepening of the active layer will probably lead to direct input of sediment and nutrients into surface waters, as can more extreme permafrost thaw slumping (see impacted sites section below), resulting in increased levels of TP, particularly in streams (Chin et al., 2016; Hobbie et al., 1999; Kendrick et al., 2018; Kokelj et al., 2009; Levenstein et al., 2018). These observations support the hypothesis that increased exposure of phosphate-rich mineral soils leads to increases in P supply to surface waters (Frey & McClelland, 2009; Hobbie et al., 1999; Pearce et al., 2015) and subsequent increases in primary production.

Declining TP trends in some areas of the Near and Sub Arctic (particularly Fennoscandian rivers and lakes with time-series data) suggest a combination of warming-induced terrestrial P uptake (Jiang et al., 2016; Jonard et al., 2015) and increased P sorption in catchment soils, leading to lower P concentrations in the southern Arctic (Huser et al., 2018). Modelling suggests that climate warming will lead to tighter cycling of P in terrestrial ecosystems (Jiang et al., 2016), and thus the supply of P to surface waters may actually decrease, resulting in overall declines in primary production and ultimately fish production. In addition, sulfate declines in the Near Arctic (Table 2 and Figure 5b), probably due to declining deposition since the 1980s (Samyn et al., 2012), may increase available soil P-binding sites due to less anionic competition (Geelhoed et al., 1997). Recovery from acidification can reduce catchment P loading because soils recovering from anthropogenic acidification may display increased formation of P-binding (oxyhydroxide) metals that can limit P mobility in the catchment and subsequent transfer to surface waters (Huser et al., 2018; Kopáček et al., 2015). Declining atmospheric P deposition may contribute to oligotrophication (Zhai et al., 2009). Combustion-related sources of atmospheric P deposition have decreased over Europe since the 1980s (Wang et al., 2015); however, few long-term trends for total (natural plus anthropogenic) P deposition have been detected (Tipping et al., 2014) and none were detected at Canadian sites where surface water P declines have occurred (Eimers et al., 2009).

Temporal trends in TN were more variable. Concentrations of TN were lower at high latitudes and increasing trends were seen at low latitudes (Table 2). Experimental and space-for-time studies both suggest that warming-induced increases of N mineralisation in shallow soils and slow uptake of N mobilised in areas of deep subsurface flow should collectively lead to increases in the export of inorganic N from terrestrial ecosystems (Jones et al., 2005; Petrone et al., 2006; Schimel et al., 2004). Although the above studies indicate that warming in the Arctic should lead to greater export of N to surface waters, variations in soil moisture and flow paths can have a strong influence on the loading of N to surface waters from the surrounding landscape. Results have not been consistent among studies, leading to competing hypotheses about the mechanisms controlling N export under varying permafrost conditions. MacLean et al. (1999) hypothesised that longer soil water residence time in areas of less

extensive permafrost facilitates greater dissolved inorganic nitrogen (DIN) removal, whereas later studies in the same region suggested that deeper flow paths facilitate DIN export (Jones et al., 2005; Petrone et al., 2006). This range of possible mechanisms that can either increase or decrease DIN fluxes from terrestrial environments to freshwaters is consistent with results from our study in which we found weak bulk trends for TN (Figure 5a). Time-series trends indicated a general increase, but the number of nominally significant trends (unadjusted $p \leq 0.05$) was relatively low and restricted to the Sub and Near Arctic regions.

4.4 | Impacted sites

Permafrost thaw is a potential driver of nutrient input to Arctic freshwaters (Abbott et al., 2014, 2015; Bowden et al., 2008; Larouche et al., 2015). Permafrost thaw is hypothesised to increase organic N mineralisation and deepen groundwater flow paths to a level below most plant roots, which will ultimately increase inorganic N in stream water (Frey & McClelland, 2009; Vonk et al., 2015). Thawing permafrost will also expose phosphate-rich minerals, where leaching has been hypothesised to increase concentrations of P in surface waters (Hobbie et al., 1999). Sorption of minerals to newly exposed mineral soils, however, may lead to the opposite effect (Striegl et al., 2005).

Substantial quantities of N, and P released by thawing permafrost are an important source of nutrients for upland vegetation (Gough et al., 2012; McGuire et al., 2012; Shaver & Chapin, 1980). If these nutrients leach to surface waters, they may increase primary productivity. Slump-impacted stream sites had significantly higher levels of TP and TN than were observed in un-impacted sites across all four Arctic regions. Concentrations of DOC were also elevated in slump-impacted sites relative to high-latitude sites in the Low and High Arctic. Measures of TP in particular are likely to over-estimate the biologically available fraction (Chin et al., 2016). However, carbon derived from permafrost thaw has been shown to integrate into stream food webs (O'Donnell et al., 2019). Collectively, the trends in our dataset support the potential for permafrost thaw to substantially alter nutrient dynamics in Arctic freshwaters (Figure 3 and Table S4).

4.5 | Circumpolar monitoring to detect trends in Arctic freshwaters

Drawing conclusions on patterns of regional and circumpolar environmental change based on data compiled from a large number of diverse sources has numerous challenges. Differences in monitoring strategy both within and among data providers resulted in differing frequencies of data collection. While monitoring at some sites was consistent over the study time period, other sites were sampled less frequently, and some were sampled only to provide supporting information for biotic sampling and analysis. Moreover, not all variables were sampled across all sites, limiting the spatial and temporal scope of assessments in some cases.

A key factor in supporting the detection of spatial and temporal trends at such a large scale is establishing a consistent set of variables measured at each site, even if sampling frequency cannot be controlled. The variables reported herein, including nutrients (TN and TP), carbon (DOC), pH/alkalinity, conductivity, and major ions (calcium, magnesium, sodium, chloride, silica, sulfate) provide a clear overview of the chemical habitat of these ecosystems, are relevant for aquatic life, are susceptible to change, (most) are fairly simple to measure even in remote locations, and can reflect landscape-level changes. Including a consistent set of variables in routine monitoring, episodic sampling, or even one-time sample collection would improve our ability to expand on the spatial extent of such assessments, and potentially to detect long-term trends in water chemistry.

Even though our dataset was not designed specifically for broad regional generalisations, the large spread of monitoring sites and analyses performed allowed us to draw broad conclusions concerning spatial and temporal patterns in Arctic freshwater chemistry. The general similarity in results between time-series analysis and analysis of bulk trends supports this. It should be noted that sites with adequate data for time-series analysis for the High and Low Arctic regions were limited ($n = 2$, one in each region), and Near Arctic region sites were mostly located in Sweden and Finland. Thus, the comparisons made between the Near Arctic and more northerly Arctic regions may have been more specific to the factors within these regions or sites. Although the number of sites with extensive time series was limited, it was possible to generalise about data collected across periods (bulk trends) in a way that was meaningful and consistent with previous work.

In our analyses, we report nominal p -values that are, in most cases, not corrected for multiple comparisons. Given the large number of tests we ran, it is most likely that the true p -values are greater than the nominal significance level we use for distinguishing between strong and weak trends. We also adopted the untested (and potentially untestable) hypothesis that the surface waters where chemical measurements were made were drawn randomly from the underlying population of water bodies. While we were not able to perform longitudinal analysis of regional (i.e. bulk) temporal trends for most sites, as sampling frequency was variable, we were able to test the hypothesis that the mean concentrations differed between periods and we assumed that these differences were attributable to temporal change as opposed to being an artifact associated with non-random sampling. Strengthening both the spatial coverage and the number of sites with regular, on-going monitoring in the future will support more robust regional and pan-Arctic assessments of water chemistry, which forms the template for freshwater biodiversity.

5 | SUMMARY

Major changes in water chemistry were detected across the Arctic, both temporally and spatially. For some variables (e.g. TP), changes were divergent across regions, underlining the influence of regionally

relevant drivers of change. On a regional basis, pH, alkalinity, sulfate, chloride, calcium, and magnesium were generally greater in the Sub and High Arctic regions than in the Near Arctic region, and appeared to reflect geological differences between these regions. In contrast, nutrients and carbon displayed latitudinal gradients, with lower concentrations in higher-latitude Arctic regions compared to the southern-most regions. One of the most interesting changes detected in both bulk trend and time-series analysis was the decline in TP in the lower latitude Arctic regions, contrasted with the increase in TP detected in the Low and High Arctic, which probably reflected different mechanisms of P flux between freshwaters and the surrounding landscape. Several spatial patterns and regional trends were consistent with changes to the chemical environment that are predicted to occur with ongoing changes to climate and deposition. For example, comparison of contemporary data with permafrost thaw slump impacted sites indicated strong differences, particularly with respect to nutrients, that spoke to the potential for future change in Arctic systems as changes to climate continue. Across the Arctic, long-term changes in freshwater chemistry have been substantial. Given the expected continued warming across the region, the changes we report in this study are likely to have even greater effects on Arctic water quality, biota, ecosystem function, and human well-being in the future.

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DATA AVAILABILITY STATEMENT

Metadata and (where allowed) data are available on the CAFF Arctic Biodiversity Data Service (ABDS) at abds.is.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

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