A landscape-isotopic approach to the geochemical characterization of lakes in the Kangerlussuaq region, west Greenland

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ABSTRACT
In west Greenland, an approximate chronosequence of landscape evolution and weathering exists between the coast, which has been ice free for long periods, and more recently deglaciated areas along the present day ice margin. Traditional geochemical and isotopic analyses (δ18O, δ13C, δ34S, δ37Cl, δ87Sr/86Sr) along with novel isotopic tools, such as δ13C and δ34S, were used to provide new insights into lake geochemical processes along a transect of lakes from the coast to the ice margin in the Kangerlussuaq region. Evaporation was found to be a key process impacting lake chemistry and isotopic signatures in the ice marginal area, with decreasing importance toward the coast. Evaporative processes were apparent in the δ13C and δ34S isotopic signatures of lake-water chemistry. Consistent with previous work elsewhere (e.g., Blum and Erel, 1995) on increased biotite weathering in glaciated environments, 87Sr/86Sr isotopic ratios were found to be more radiogenic (>0.73) in lakes found in more recently glaciated terrain. Sulfide oxidation was the main source of sulfur (as sulfate) in lakes in the ice marginal area, while the influence of marine aerosols and bacterial sulfate reduction increased further away from the ice sheet around the fjord Kangerlussuaq. Groundwater discharge significant enough to impact lake chemistry was not observed in any of the lakes studied, suggesting that little groundwater–surface water interaction occurs in the study area or that recharge conditions are present in the majority of the lakes studied.

Introduction
The geochemical composition of lake water reflects catchment geology, regional climate, vegetation, and soil development and, importantly, varies on a range of spatial and temporal scales (Kling et al. 2000). The interaction and dominance of these factors will vary over time as climate and vegetation successional states change. Landscape change over time, that is, ontogeny (Fritz and Anderson 2013; Leng et al. 2012), exerts strong controls on biogeochemical processing (Engstrom et al. 2000), but so does position in the landscape, which influences hydrological connectivity, both surface and groundwater (Johansson et al. 2015; Webster et al. 2000). At lower latitudes landscape perspectives of lake biogeochemistry have tended to focus on nutrients, in part because of catchment cultural disturbance (Kendall 1998; Wagner et al. 2011), and DOC, because of the dominance of lakes in boreal landscapes and the key role terrestrially derived dissolved organic carbon (DOC) plays in aquatic ecosystem functioning (Jansson et al. 2007). In many of these studies, the use of multiple isotopic tracers has proven invaluable in understanding landscape evolution, particularly in terms of chemical interactions.

Landscape position has been shown to strongly influence groundwater effect on lake water chemistry, even in areas of low relief (Webster et al. 1996, 2000). In contrast to temperate and boreal zones, Arctic lakes have reduced terrestrial vegetation biomass, limited cultural impacts, and, importantly, permafrost strongly reduces groundwater hydrological connectivity. In areas of continuous permafrost, taliks provide one of the few pathways for groundwater–surface water interaction. Taliks are created in permafrost environments by lakes that do not freeze to the bottom during the winter, resulting in the warming of the ground around the lake bottom to temperatures above 0°C. Taliks have...
been documented beneath lakes in permafrost environments in both the Arctic (Burn 2002; Kokelj, Zijdlik, and Thompson 2009) and Antarctic (Cartwright and Harris 1981; Matsubaya et al. 1979). Subpermafrost waters may only receive recharge from lakes with taliks that fully penetrate the permafrost (through taliks) and from subglacial meltwaters where permafrost is absent. Subpermafrost groundwater discharge will occur in lakes with through taliks, or as discharge into the ocean (e.g., DeFoor et al. 2011).

As well as the overarching effects of low mean annual air temperature, climate can impact Arctic lakes directly through processes such as precipitation and evaporation, as well as indirectly through catchment inputs associated with chemical and physical weathering and impacts on biota (Anderson et al. 2001; Birks, Jones, and Rose 2004). Landscape relief also influences local air temperature and evaporation rates, which can strongly affect vegetation development within a catchment (Anderson 2014). The interplay between vegetation and local climate can influence hydrological retention time and hydrochemistry (Turner et al. 2014). In west Greenland these processes result in immediately adjacent lakes having contrasting major ion chemistry: evaporatively concentrated oligosaline lakes are interspersed among freshwater ones (Anderson et al. 2001; Johnson et al. 2000; Shand et al. 2007).

Isotopic tracers such as δ18O and δ2H, as well as 87Sr/86Sr and δ34S–δ18O (SO4), can reveal whether groundwater discharge is providing a significant component of the water balance within a lake catchment (Gibson, Birks, and Yi 2016; Gibson and Edwards 2002; Gibson et al. 2005; Hagerty and Webb 2008). Additional insight into lake evolution can be gained by using novel isotopic tools, such as δ37Cl and δ81Br (Eggenkamp 2014), in combination with more traditional geochemical and isotopic analyses (δ18O, δ2H, 3H, δ34S–δ18O (SO4), and 87Sr/86Sr). Use of these isotopic parameters has been applied elsewhere to provide increased understanding of evaporation (Gibson, Birks, and Yi 2016; Leng and Anderson 2003), glaciation/deglaciation (Blum and Erel 1997), weathering (Anderson, Drever, and Humphrey 1997; Blum and Erel 1997; Sharp, Creaser, and Skidmore 2002), aeolian transport (Hanlon 2015; Hanlon et al. 2017), marine inputs (McArthur et al. 1989; Negrel and Roy 1998; Wood and Sanford 1995), groundwater–surface water interaction (Bohike and Denver 1995; Bullen and Kendall 1998; Hagerty and Webb 2008; Kendall and McDonnell 1998; Lyons et al. 1995; Negrel et al. 2003), and microbial processes (van Everdingen and Krouse 1985) in glaciated areas.

The Kangerlussuaq area of west Greenland is an excellent natural experimental area in which to address some of these controls on lake geochemistry at the landscape scale using stable isotopes. There are strong gradients in climate (both precipitation and temperature), which are reflected in both soil and vegetation differences as well as limnology, especially conductivity and DOC (Whiteford et al. 2016), with a number of closed-basin oligosaline lakes close to the head of the fjord (Figure 1). Regional geology is relatively homogenous, but there is a gradient in lake age from the coast (formed >10,000 yr BP) to those adjacent to the present ice margin, which are approximately 4,000 years old, and very young lakes formed recently at the edge of the outlet glaciers. A final regionally variable process influencing geochemistry is dust deflation from the outwash plains and its deposition on lakes (Anderson et al. 2017). Here we use novel stable isotope analysis of samples from lakes covering the environmental gradients in the Kangerlussuaq area to provide insight into how lake chemistry evolves at 100–1,000-year timescales following deglaciation. Additionally, two boreholes drilled as part of the Greenland Analogue Project (GAP) provide information on the geochemical and isotopic composition of groundwaters in the study area (Claesson Liljedahl et al. 2016; Harper et al. 2016), allowing the extent to which groundwater–surface water interaction affects lake chemistry in the Kangerlussuaq region to be examined.

**Study area**

The study area centers on the town of Kangerlussuaq (Figure 1), located about 125 km inland from the west coast and 25 km west of the Greenland Ice Sheet and encompassing both a low-Arctic maritime climatic regime in the coastal region and a low-Arctic continental inland regime (Anderson et al. 2001). Inland, continuous permafrost may be greater than 300 m thick near the ice sheet (Harper et al. 2016; Kern Hansen 1990), while closer to the head of the fjord it is reported to be approximately 100–150 m thick (van Tatenhove and Olesen 1994). Toward the coast, permafrost becomes discontinuous around 50 km west of Kangerlussuaq (Weidick 1975). Mean annual temperature (measured 1977–2011) at Kangerlussuaq is −5.1°C and varies from −40°C in winter to 18–20°C in summer, with an annual precipitation of 173 mm (Cappelen 2012). In coastal areas, summer temperatures are cooler and mean annual precipitation is higher: 383 mm per year on the coast at Sisimiut (Figure 1; long-term normal 1961–1990; Cappelen 2012).
The geology of the study area consists of Archean gneisses reworked in the Paleoproterozoic with an east-northeast structural trend. The area is structurally complex, having gone through several episodes of deformation (Engström and Klint 2014), with lakes often occurring along structural lineaments and within closed basins. The dominant rock type is quartzo-feldspathic gneiss that is quartz rich and commonly biotite bearing.

Previous studies have examined lake chemistry, palaeolimnological history, and ecology in the Kangerlussuaq region (Aebly and Fritz 2009; Anderson et al. 2008, 2001; Anderson and Leng 2004; Bocher 1949; Eisner et al. 1995; Fredskild 1977; Fritz and Anderson 2013; Jensen 1889; Leng and Anderson 2003; Leng et al. 2012; McGowan, Ryves, and Anderson 2003; Olsen, Anderson, and Leng 2013; Williams 1991). In the present study, a series of lakes, from the coast to the ice front in the Kangerlussuaq region of southwest Greenland, were sampled (Figure 1).

Lakes in this region are fresh (<1,000 mg/L) to brackish (1,000–10,000 mg/L) based on the
classification of S.N. Davis (Krieger 1964). Most lakes have an electrical conductivity (EC) of less than 800 µS/cm. However, previous limnological studies in the region used a classification of less than 800 µS/cm (approximately 500 mg/L TDS) as dilute and more than 800 µS/cm as “oligosaline”; thus, saline is used in a relative sense when comparing the more brackish lakes to the dilute lakes (Anderson et al. 2001; Leng and Anderson 2003). The convention of referring to lakes with EC greater than 800 µS/cm as “oligosaline” will be used in this article (hereafter, saline). In general, the Kangerlussuaq region of Greenland lacks the very high TDS lakes (TDS > 10,000 mg/L) that may be found in other polar environments, such as the Canadian Arctic (Ouellet, Dickman, and Page 1989) and Antarctica (e.g., Lyons et al. 2002; Matsubaya et al. 1979), which were formed as the result of isostatic uplift and trapping of saltwater. In contrast, saline lakes in the Kangerlussuaq region are primarily closed-basin lakes where evaporation has concentrated salts in the lake water throughout the past 6,000–11,000 years since deglaciation (Anderson and Leng 2004).

**Methods**

Originally, lake samples were taken for the Greenland Analogue Project (GAP; see Claesson Liljedahl et al. 2016; Harper et al. 2016) in the area from the head of the fjord Kangerlussuaq (Søndre Strømfjord) and extending to the ice sheet (Figure 1). Samples for the GAP were acquired between 2008 and 2013. The aim of the GAP was to study the impact of a modern ice sheet on the groundwater system. During the initial phase of the study, the lake sampling program was designed to examine the potential for groundwater discharge to, or recharge from, lakes in the periglacial area. Data collected as part of this study and the GAP include surface-water samples, subpermafrost groundwater samples, spring water samples, and samples of glacial meltwater taken from sub- and supraglacial flow at Isunnguata Sermia, Russell Glacier, and Leverett Glacier (Figure 1).

Water samples from lakes collected for the GAP were generally taken from the shoreline, using nalgene bottles. Care was taken to sample in deeper locations to avoid poorly mixed shallow waters. In some instances, samples were taken at various depths, either from a boat or from holes drilled through ice (depending on the time of year). Sample depths are indicated in the sample name for samples not acquired from the surface. Samples taken at depth were acquired using a kemerer or van Dorn sampler. pH, conductivity, and lake temperature were measured in the field using a portable Oakton pH/conductivity meter. Cation samples were filtered (0.45 µm) and acidified in the field, while anion and isotope samples were untreated. Samples were refrigerated at approximately 4°C, except for brief periods during shipping.

Additional archived water samples were used to extend the study area and complete a transect of lakes from the ice sheet to the coast (Figure 1). These samples consisted of historical samples (2001–2012) from previous studies in the region (see Anderson and Stedman 2007). Archived samples were from tightly sealed bottles, and the possibility of sample deterioration in these older samples was considered unlikely to be an issue, particularly for the isotopic analyses used in this study.

Two research boreholes installed for the GAP provided subpermafrost and talik groundwater samples with which to compare the surface waters (Harper et al. 2016). DH-GAP01 was drilled to intersect a talik beneath lake L26 (Figure 1) and is a total of 221.6 m long and 191 m deep (vertical depth). DH-GAP04 was drilled at the edge of the ice margin at the glacier Isunnguata Sermia. DH-GAP04 is 687 m long and has a vertical depth of 645 m. DH-GAP04 has three sampling sections between the base of the permafrost, at 400 m borehole length, and the bottom of the borehole. The lowest section (604.5–687 m borehole length) of the DH-GAP04 borehole is free of drilling water contamination, while the upper and middle sections still contained significant drilling water contamination (33 and 29 percent, respectively) at the time of sampling in 2014. Both δ18O and δ2H results presented here for the upper and middle sections of DH-GAP04 are calculated using a linear mixing model and drilling-fluid tracer concentrations (Claesson Liljedahl et al. 2016). The drill-fluid tracer allows the proportion of the sample that is drilling fluid, which has a known composition, to be determined and allows the unknown proportion and composition of the groundwater to be calculated (see Harper et al. 2016 for details). Other isotopic results for these sections (δ34S-δ18O of SO4, δ37Cl, δ81Br, and 87Sr/86Sr) can be considered representative of groundwater isotopic values because of much higher concentrations of these ions in the groundwaters compared to the very dilute solute composition of the drilling fluids.

The highly soluble mineral gypsum (CaSO4 • 2H2O) was found as an abundant fracture infilling mineral in the DH-GAP04 core, predominantly below 300 m of borehole length (Harper et al. 2016; Henkemans 2016; Pere 2014). Samples of fracture gypsum were obtained using dental tools under a binocular microscope and were analyzed for δ34S and δ18O of SO4 as well as 87Sr/86Sr. Sulphide
minerals, pyrite (FeS₂), and chalcopyrite ([CuFe]S₂), from gnaiassic bedrock from the DH-GAP01 borehole were extracted and analyzed for δ³⁴S.

Samples of salt crusts that had formed on soils near lake L21 (Hunde Sø; SS3 in the Anderson et al. 2001 numbering system) and lake L32 (Store Saltso; SS17) were preserved in sample bags and analyzed by X-ray diffraction (XRD). XRD was used to determine what salts formed these crusts.

**Isotopic and geochemical analyses**

Isotope analyses were performed at the University of Waterloo Environmental Isotope Laboratory. Tritium preanalysis was determined using the electrolytic enrichment method of Taylor (1977). After enrichment, samples were counted in a LKB Wallac 1220 Quantalus liquid scintillation counter with a detection limit of 0.8 ± 0.8 TU. Deuterium determinations were made following the Cr reduction method outlined by Morrison et al. (2001) and were analyzed on an Isoprime IRMS coupled to a Euroelectron elemental analyzer. Oxygen isotope analysis was performed on an IsoPrime continuous flow isotope ratio mass spectrometer (CF-IRMS), using the preparation procedures of Epstein and Mayeda (1953) with Moser’s modification (Moser 1977). Results for δ¹⁸O and δ²H are reported based on standard corrections to VSMOW (Vienna Standard Mean Ocean Water) and VSLAP (Vienna Standard Light Antarctic Precipitation) from the International Atomic Energy Agency (IAEA). Analytical reproducibility of δ¹⁸O and δ²H are ±0.2‰ and ±0.8‰, respectively.

Sulphate is extracted using BaCl₂, converted to SO₂ by combustion with Nb₂O₅, and analyzed on a Micromass IsoChrom-IRMS. For analysis of SO₄²⁻, the BaSO₄ is combusted to produce CO₂ and is analyzed in a GVI IsoPrime-IRMS (±0.5‰; Morrison 1997). Both ¹⁸O and ³⁴S are corrected to BaSO₄ IAEA-SO5, IAEA-SO-6, and NBS-127.

Chlorine and bromine stable isotope determinations are performed by first precipitating Cl⁻ or Br⁻ as AgCl or AgBr using AgNO₃. Analyses for ³⁷Cl and ⁸¹Br are carried out on CH₃Cl and CH₃Br, respectively, after reacting the silver chloride/bromide with methyl iodide. Both ³⁷Cl and ⁸¹Br were analyzed using continuous flow technology on a Micromass IsoPrime IRMS (±0.2‰), using the methods outlined in Eggenkamp (1994), Kaufmann et al. (1984), and Shouakar-Stash et al. (2005a, 2005b). The reference material used for ³⁷Cl analysis is Standard Mean Ocean Chloride (SMOC), and that used for ⁸¹Br analysis is Standard Mean Ocean Bromide (SMOB).

Analysis of ⁸⁷Sr/⁸⁶Sr isotopic ratios was performed using thermal ionization mass spectrometry (TIMS). Samples are passed through a strontium-specific resin. The isolated strontium is loaded onto a double rhenium filament and analyzed using a Thermo Finnegan Scientific TRITON TIMS and calibrated against the NBS international standard material NIST SRM 987 (Dicken 2000).

Geochemistry samples were analyzed at Labtium Oy (via Geological Survey of Finland) in 2008 and 2010–2013 and at the TVO (Teollisuuden Voima Oy) laboratory in Finland in 2009. Geochemical analyses were performed using the following methods at Labtium Oy: alkalinity was measured using a titrimetric determination, anions were measured by Ion Chromatography (IC), and cation multielement determination was performed by a combination of Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES). At TVO, multielement analyses were performed using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) and ICP-MS analysis. Results with charge balances exceeding 10 percent were discarded, with the exception of the sample from lake L20 (01/07/2008), which had a charge balance error of ~12.5 percent. Both Cl⁻ and SO₄²⁻ concentrations from the 2008 L20 sample (L20-1) are used in order to include L20 δ³⁴S and δ¹⁸O (SO₄) isotopic values in Figure 4 and Figure 7. Sampling and analytical methods for historical samples are documented in Anderson et al. (2001).

**Results**

**Geochemical**

The dilute lakes were generally of Ca or Mg -HCO₃ or of Ca or Mg-HCO₃-SO₄ type, while the more saline lakes tended to be of Na or Mg-HCO₃ Cl-type waters. A small subset of lakes contained a high percentage of anions as sulfate: lake L20 from the GAP and lakes SS70–76 in Anderson et al. (2001). Lake L20 was highly acidic, with a pH of 3.5, whereas the majority of lakes had a pH greater than 7.

Lakes were divided into four categories based on size, salinity, and water source. (1) **Ponds** are lakes that are approximately 2 m deep or less, and may evaporate completely in the summer or freeze to the bottom in the winter. Ponds may show seasonal salinity variations because of evaporation. For example, lake L20 had an electrical conductivity of 354 µS/cm when sampled in early June 2008, and 1,750 µS/cm when sampled in early September 2010. Lake L20 had visibly
shrink by a significant amount when sampled in 2010. (2) **Dilute lakes** are those that were unlikely to freeze to the bottom in winter (>2 m deep) and had conductivities of less than 800 µS/cm, following the classification of Anderson et al. 2001. (3) **Saline lakes** were also of sufficient size to not freeze to the bottom in winter and had conductivities of more than 800 µS/cm. In general, saline lakes are located in closed basinal settings with no visible outflow. (4) **Meltwater/thaw lakes** include glacial meltwater-fed lakes (e.g., lake L6) and thaw lakes that derive water from the melting of frozen till and precipitation. Thaw lakes include lakes L12–14 and L23–24, all of which are located on the till plain in front of Leverett Glacier (Figure 1). While the thaw lakes are likely to be more geochemically evolved than the meltwater-fed lakes, they are similar in terms of low conductivity and depleted δ¹⁸O/δ²H isotopic signatures (Table 1) and access to relatively fresh glacial sediment. Meltwater and thaw lakes tend to plot together in many of the figures.

A summary of average, minimum, and maximum values for concentrations of dissolved species, EC, pH, alkalinity, as well as a subset of isotopic results are presented by group (ponds, dilute lakes, saline lakes, and meltwater/thaw lakes) in Table 1. Table 2 presents results for a representative subset of lakes with chemistry typical of each of the areas described in Figure 1 (ice marginal, upper fjord, and coastal).

### Isotopic results

When considering the isotopes of water, lakes fall along local evaporation lines (LELs) similar to those described by Leng and Anderson (2003) for lakes close to the ice (zone 5) and lakes around the upper fjord (zone 4; Figure 2A). Meltwater lakes tend to show greater isotopic depletion and fall along an evaporation line with lower deuterium-excess (D-excess) relative to the LELs described by Leng and Anderson (2003; Figure 2A). The precipitation average of δ¹⁸O was estimated to be −19‰ by Leng and Anderson (2003), based on the intercepts of the LELs with the GMWL and Kangerlussuaq’s position between the IAEA-monitoring stations at Gronnedal and Thule. A snowfall sample taken July 1, 2009, near the ice sheet had a δ¹⁸O of −20.16‰ and a tritium concentration of 13.8 TU. Lakes had a range of tritium concentrations from 6.6 to 13.2 TU (mean of 9.6 TU for all lakes). Average, minimum, and maximum tritium concentrations are broken down for each lake grouping in Table 1.

The δ⁷³Sr/δ⁸⁶Sr isotopic ratios in lakes have a large range of values from 0.7154 to 0.7580 (subset of results in Table 2). Both δ⁵³Cl and δ⁸¹Br are measured against standard mean oceanic chloride (SMOC) and bromide (SMOB), respectively, and thus, marine chlorine and bromine have isotopic signatures of 0‰. Fifteen lakes were analyzed for δ⁵³Cl, which ranged from −0.41‰ to 0.04‰ (Table 3). Six lakes were analyzed for δ⁸¹Br, which ranged from −0.06‰ to 1.76‰ (Table 3).

Groundwaters tend to be of Ca-Na-SO₄ type and are depleted in δ¹⁸O and δD relative to lake waters. DH-GAP01 intersects the talik beneath lake L26. Pressure measurements indicating a downward gradient in hydraulic head suggest recharge conditions in the talik (Johansson et al. 2015). Both δ¹⁸O and δD isotopic values that are more enriched than the DH-GAP04 groundwaters and plot below the GMWL further support the recharge and mixing of lake water into the talik (Figure 2B). Groundwater from DH-GAP04 indicates that subpermafrost groundwater at the ice margin is brackish to a depth of 450 m and contains concentrations of sulfate up to 1,900 mg/L and Cl concentrations of 176 mg/L. The δ⁸⁷Sr/δ⁸⁶Sr ratios of groundwater (0.7033–0.7075) reflect that of fracture minerals, such

| Table 1. Summary of major geochemical and isotopic results. Alkalinity is expressed as mmol/L as CaCO₃. Meltwater lakes, lakes that receive meltwater directly, and the thaw lakes on the Leverett Glacier till plain are separated here to illustrate the similarity between these two groups. |
|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| **Group**       | **EC**          | **pH**          | **δ¹⁸O**        | **δD**          | **δH**          | **δ⁴⁰Ca**        | **δ²⁴Mg**        | **δ¹⁶K**         | **δ¹⁰Na**        | **δ¹⁷Cl**        | **δ³⁵SO₄**        | **δ⁴⁰Alk**       |
| **Ponds n = 8** |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| Avg.            | 845             | 7.6             | −10.5           | −107.9          | 10.2            | 84.5            | 46.7            | 16.7            | 17.7            | 21.4            | 360             | 1.6             |
| Min.            | 164             | 3.4             | −16.2           | −134.1          | 9.4             | 3.5             | 1.1             | 1.7             | 0.9             | 1.4             | 6.1             | 0.2             |
| Max.            | 1797            | 9.2             | −5.2            | −84.5           | 12.0            | 210             | 135             | 45              | 78              | 102             | 949             | 3.0             |
| Min.            | 50              | 7.2             | −15.1           | −131.6          | 7.4             | 2.7             | 2.2             | 0.7             | 1.3             | 1.7             | 0.0             | 0.1             |
| Max.            | 616             | 9.6             | −6.8            | −91.9           | 12.9            | 42              | 36              | 28              | 53              | 69              | 88              | 4.5             |
| **Dilute Lakes n = 26** |               |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| Avg.            | 2,848           | 8.9             | −8.8            | −99.9           | 10.5            | 25.9            | 161             | 113.9           | 350             | 487             | 174             | 14.0            |
| Min.            | 902             | 8.2             | −11.4           | −113.2          | 8.2             | 4.0             | 57.3            | 35.3            | 74.2            | 121             | 18.2            | 6.6             |
| Max.            | 4,500           | 9.2             | −6.5            | −93.9           | 13.2            | 108             | 254             | 190             | 590             | 782             | 403             | 22.2            |
| **Saline Lakes n = 14** |               |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| Avg.            | 223             | 7.7             | −15.1           | −128.8          | 10.5            | 25.5            | 9.3             | 3.4             | 3.1             | 3.0             | 43.6            | 1.23            |
| **Meltwater Lakes** |               |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| L6              | 62              | 6.8             | −18.1           | −145.5          | 9.4             | 5.9             | 3.3             | 0.4             | 1.8             | 1.8             | 3.3             | 0.43            |
| Avg.            | 238             | 8.4             | −14             | −130            | 8               | 32.7            | 5.0             | 6.3             | 4.2             | 2.6             | 57.2            | 0.9             |
| Min.            | 61              | 7               | −15             | −136            | 7.2             | 0.9             | 1.7             | 1.1             | 0.9             | 12.0            | 0.3             |
| Max.            | 327             | 8.4             | −13.1           | −127.2          | 10.5            | 46              | 7.3             | 8.5             | 5.2             | 3.8             | 77              | 1.6             |
Table 2. Summary of representative chemistry and isotopic results. P = pond, DL = dilute lake, SL = saline lake, and MW = meltwater lake. *Indicates that the results for dissolved chemical species are average results throughout multiple years as measured by John Anderson (ex., Anderson et al. 2001). Alkalinity is expressed as mg/L as CaCO$_3$. NM = not measured.

| Location | Name | Sample Date | Lake Type | EC µS/cm | pH | Ca mg/l | Mg mg/l | K mg/l | Na mg/l | Cl mg/l | SO4 mg/l | Alk µg/l | Sr µg/l | Br µg/l | δ18O ‰ | δ2H VSMOW | δ87/86Sr |
|----------|------|-------------|-----------|----------|-----|---------|---------|-------|---------|---------|----------|----------|---------|--------|--------|--------|-----------|---------|
| Ice Marginal | L8 | 2008-06-28 | P | 342 | 7.9 | 32 | 20 | 2 | 3.5 | 2 | 132 | 76 | 62 | 36 | −16.2 | −134 | 10 | NM |
| | L20 | 2010-09-05 | P | 1750 | 3.4 | 182 | 74 | 29 | 17 | 14 | 949 | <2 | 217 | 123 | −10.6 | −107 | 10 | NM |
| | L18 | 2008-07-01 | P | 1797 | 8.9 | 130 | 135 | 45 | 78 | 102 | 725 | 304 | 540 | 420 | −5.2 | −85 | 10 | 0.7459 |
| | L1 | 2008-06-27 | DL | 381 | 8.4 | 27 | 19 | 11 | 9 | 31 | 2 | 303 | 94 | 156 | −11.0 | −111 | 11 | 0.7553 |
| | L15 | 2008-06-30 | DL | 74 | 8.0 | 5 | 4 | 1 | 1.8 | 2 | 1 | 57 | 15 | 16 | −14.8 | −132 | 8 | 0.7536 |
| | L26 | 26m Talik Lake | 2010-05-11 | DL | 232 | 7.7 | 18 | 11 | 6 | 6.7 | 10 | 3 | 190 | 63 | 40 | −12.9 | −118 | 10 | 0.7385 |
| | L11 | 2011-09-12 | DL | 150 | 7.7 | 12 | 7 | 3 | 6.9 | 9 | 3 | 112 | 40 | 84 | −12.5 | −115 | 8 | 0.7439 |
| | S332 | 2003-08-21 | DL | 51 | 7.3 | 4 | 3 | 1 | 2 | 2 | 1 | <1 | NM | NM | NM | NM | 0.7557 |
| | L17 | 2008-07-01 | DL | 902 | 9.0 | 23 | 85 | 35 | 74 | 121 | 24 | 816 | 72 | 277 | −11.4 | −113 | 10 | 0.7510 |
| | L29 | 2010-09-04 | SL | 1250 | 9.2 | 15 | 85 | 88 | 105 | 152 | <1 | 859 | 70 | 79 | −6.5 | −96 | 10 | NM |
| | L6 | 2008-06-28 | MW | 223 | 7.7 | 26 | 9 | 3 | 3.1 | 3 | 44 | 123 | 52 | 40 | −15.1 | −129 | 11 | 0.7514 |
| | L12 | 2008-06-29 | TH | 297 | 7.8 | 40 | 6 | 8 | 4.9 | 4 | 71 | 130 | 130 | 27 | −13.6 | −128 | 7 | 0.7533 |
| Upper Fjord | L33 | Lake Ferguson | 2010-09-04 | DL | 66.1 | 7.6 | 7 | 3 | 2 | 2.6 | 3 | 2 | 57 | 41 | 21 | −12.7 | −127 | 12 | 0.7187 |
| | S52* | 2003-08-20 | DL | 321 | 7.9 | 26 | 13 | 4 | 18 | 15 | 3 | NM | NM | NM | NM | NM | 0.7533 |
| | S58/L46 | 2012-09-03 | DL | 236 | 8.9 | 23 | 15 | 11 | 21 | 28 | 2 | 237 | 96 | 103 | −10.2 | −95 | 7 | 0.7310 |
| | L45 | 2012-08-28 | SL | 3470 | 8.3 | 108 | 150 | 80 | 340 | 722 | 384 | 656 | 883 | 1960 | −9.0 | −101 | 9 | 0.7267 |
| | L21 | Hunde Sø | 2010-09-04 | SL | 4030 | 8.8 | 16 | 203 | 182 | 545 | 797 | 180 | 2060 | 574 | 980 | −10.1 | −101 | 15 | 0.7298 |
| | L32 | Store Saltsø | 2010-09-04 | SL | 3110 | 8.9 | 21 | 155 | 181 | 343 | 372 | 18 | 1880 | 238 | 667 | −9.9 | −104 | 8 | 0.7540 |
| | L22/S54 | Braya Sø | 2011-09-10 | SL | 3000 | 8.9 | 20 | 149 | 128 | 362 | 493 | 91 | 1560 | 356 | 759 | −8.9 | −95 | 12 | 0.7295 |
| | L39/S55 | Limnea Sø | 2011-09-10 | SL | 4500 | 9.0 | 15 | 233 | 190 | 588 | 782 | 190 | 2230 | 337 | 944 | −7.7 | −93 | 11 | NM |
| | S51122* | SL | 3167 | 8.8 | 58 | 141 | 156 | 418 | 664 | 243 | NM | NM | NM | NM | NM | NM | 0.7275 |
| | S51421 2m* | 2004-06-23 | SL | 2690 | 9.1 | 18 | 155 | 4181 | 337 | 415 | 175 | NM | NM | NM | NM | NM | 0.7314 |
| Coastal | S548* | 12.08.03 | DL | 291 | 6.6 | 3 | 1 | <1 | 2 | 4 | 2 | NM | NM | NM | NM | NM | 0.7154 |
| | S576* | 12.08.03 | DL | 345 | 7.7 | 42 | 10 | 3 | 9 | 13 | 88 | NM | NM | NM | NM | NM | 0.7410 |
as gypsum (0.7023–0.7080), and are less radiogenic than surface waters (0.7154–0.7580; Henkemans 2016). Lake sulfate values have a much larger range of $\delta^{34}$S and $\delta^{18}$O (SO$_4^{2-}$) values than groundwaters or sulfide and sulfate minerals. Full results for $\delta^{34}$S and $\delta^{18}$O (SO$_4^{2-}$) are given in Table 4.

Salt crusts collected from the vicinity of L21 (Hunde Sø) and L32 (Store Saltsø) were analyzed by X-ray diffraction. Salts around lake L21 included antarcticite

Table 3. Results of $\delta^{37}$Cl and $\delta^{81}$Br analyses on lakes and borehole groundwaters. Analytical uncertainty on $\delta^{37}$Cl and $\delta^{81}$Br is ±0.2‰. Locations with * next to the name indicate average lake chemistries. NM = not measured.

| Location | Sample Type | Cl (mg/l) | Br (µg/l) | $\delta^{37}$Cl | $\delta^{81}$Br |
|----------|-------------|-----------|------------|-----------------|-----------------|
| L32      | pond        | 372       | 667        | −0.2            | −0.1            |
| L22/SS4  | pond        | 493       | 759        | 0.0             | 0.8             |
| SS1122 8m* | pond       | 664       | NM         | 0.0             | 0.9             |
| L29      | pond        | 152       | 79         | −0.3            | 1.0             |
| L41      | pond        | 40        | 141        | 0.0             | 1.7             |
| SS1421 18m* | pond      | 415       | NM         | −0.5            | 1.8             |
| SS1421 2m | pond        | NM        | NM         | −0.3            | NM              |
| SS1421   | pond        | NM        | NM         | 0.1             | NM              |
| SS3*     | pond        | 669       | NM         | −0.6            | NM              |
| L18      | pond        | 102       | 420        | −0.4            | NM              |
| L21      | pond        | 797       | 980        | −0.2            | NM              |
| SS6*     | pond        | 567       | NM         | −0.2            | NM              |
| L26 5m   | pond        | 10        | 40         | −0.1            | NM              |
| SS5 0.5m | pond        | 782       | 944        | 0.0             | NM              |
| SS1164 8m* | pond       | 173       | NM         | 0.0             | NM              |
| L12      | pond        | 4         | 27         | 0.0             | NM              |
| DH-GAP01 | groundw. BH | 8         | 90         | −0.1            | 0.3             |
| DH-GAP04U | groundw. BH | 94        | 1,260      | 0.3             | 0.4             |
| DH-GAP04M | groundw. BH | 109       | 1,520      | 0.2             | NM              |
| DH-GAP04L | groundw. BH | 173       | 2,390      | 0.4             | 0.6             |

Table 4. Isotopic composition and concentration of sulfate in surface waters and groundwaters. Sulfate minerals (predominantly gypsum) from fracture infillings and sulfide minerals from the rock matrix are included from the DH-GAP01 and DH-GAP04 cores. Gypsum was found abundantly as a fracture filling below a depth of 300 m in the DH-GAP04 borehole, while only one occurrence of gypsum was noted in the DH-GAP01 borehole. NM is not measured because of insufficient sample material. NA = not applicable.

| Location | Sample Type | $\delta^{34}$S | $\delta^{18}$O |
|----------|-------------|----------------|----------------|
|         |             | % CDT          | % VSMOW        |
| L20 2   | pond        | −0.7           | −3.0           |
| L20 1   | pond        | 0.4            | −3.9           |
| L18     | pond        | 1.1            | 3.4            |
| SS75    | dilute lake | 1.2            | −2.2           |
| L43     | dilute lake | 12.5           | −1.0           |
| L45     | saline lake | 14.1           | NM             |
| SS5     | saline lake | 19.3           | 8.1            |
| SS5     | saline lake | 20.2           | 8.0            |
| SS4     | saline lake | 22.9           | 5.5            |
| SS4     | saline lake | 23.3           | 4.6            |
| L21     | saline lake | 23.4           | 5.3            |
| L12 1   | thaw lake   | 3.8            | −11.0          |
| DH-GAP01 | groundw. BH | 4.9            | 5.9            |
| DH-GAP04-Up | groundw. BH | 5.0            | 3.9            |
| DH-GAP04-Mid | groundw. BH | 4.5            | 3.1            |
| DH-GAP04-Low | groundw. BH | 5.0            | 2.9            |
| Leverett Spring | groundw. Spr | 10.9         | 5.9            |
| DH-GAP01 | sulfide minerals | 2.3–3.7 | NA             |
| DH-GAP04 | fracture sulfate | 2.6–10.7 | 4.5–9.1 |
Evaporation in closed-basin lakes is an important source of salinity elsewhere in Greenland (Williams 1991) and in the McMurdo Dry Valleys, Antarctica (Gooseff et al. 2006; Matsubaya et al. 1979). Evaporation is a major control of the isotopic signature (δ18O/δ2H) of lakes in the region. Leng and Anderson (2003) analyzed δ18O and δ2H in lakes from the coast to the ice sheet and found that lakes fall along LELs, which vary in slope by region (Figure 2A). Coastal lakes have an LEL with a slope (s = 5.4–5.5) closest to the slope of the GMWL (s = 8) because of higher humidity (Gibson, Birks, and Yi 2016), while in the dry, low precipitation regions close to the ice sheet, the slope was much lower (s = 3.9). Continental climates at high latitudes can produce highly evaporative signatures because of pronounced evaporation under arid conditions during the seasonal thaw period (Gibson, Birks, and Yi 2016). Depending on the volume of precipitation, surface area, and lake depth, the δ18O isotopic signature can vary annually by more than 2‰ (Leng and Anderson 2003). The lakes sampled for the GAP show similar trends to those sampled by Anderson et al. (2001; Figure 2A). Evaporative effects are greatest in the ice-marginal lakes (area 1) and decrease toward the coast (area 3). Lakes for the GAP were sampled in the ice-marginal (area 1) and upper fjord (area 2) regions, which coincide with areas 4 and 5 in Leng and Anderson (2003; Figure 2A).

The majority of lakes whose main water source is glacial meltwater have a lower D-excess and plot below the LEL for the ice-marginal lakes (area 1; shown as line 5 in Figure 2A), because the initial source of water for the meltwater lakes would have the more depleted isotopic signature observed for meltwaters (Figure 2B; Gibson, Birks, and Yi 2016).

Evaporation in shallow areas around lakes or from soils can lead to salt-crust formation. An XRD analysis of salt crusts found around lakes L21 and L32 showed the presence of gypsum as well as antarcticite and hydrohalite: chloride salts that occur in cold-climate conditions. Salt crusts may be redissolved during precipitation events or snowmelt, contributing to lake salinity. Salts may also be removed by wind and transported out of the lake catchment basin.

The stable isotopic ratio of chlorine (chloride), 35Cl, can be used to identify sources of chloride salts and processes. Chlorine-37 is preferentially incorporated into the solid phase during halite (or other chloride salt) precipitation, causing the residual solution to become progressively lighter as halite precipitates (Magenheim et al. 1995; Wood, Sanford, and Frape 2005). In a closed system, where halite is precipitated and then redissolved, the δ37Cl signature should not change significantly from the initial value. In the case of

\[(CaCl_2 \cdot 6H_2O), \text{calcite (CaCO}_3)\), and hydrohalite (NaCl \cdot 2H_2O), while around L32 (Store Saltsø) the salt crust was composed primarily of gypsum (CaSO_4 \cdot 2H_2O).

**Discussion**

The lakes sampled represent a transect (Figure 1) from the coast north of the fjord Kangerlussuaq to the ice sheet east of Kangerlussuaq. The transect represents both a climatic gradient from maritime to continental interior (Anderson et al. 2012; Leng et al. 2012) as well as a chronosequence of lake and lake catchment development since glacial retreat (Fritz and Anderson 2013). The lakes along this transect can be roughly divided into the three areas shown on Figure 1B. The first (area 1) is from the ice margin to the head of the fjord (ice marginal). The second (area 2) is the area around the head of the fjord that Leng and Anderson (2003) termed “upper fjord,” and the third (area 3) is coastal and covers from the head of the fjord to the west coast (Figure 1).

From the coastal to ice marginal areas, the processes that affect the chemical evolution of lakes vary in importance. Evaporation decreases toward the coast as humidity increases (Anderson et al. 2012; Leng and Anderson 2003). Weathering rates are assumed to decrease in the less vegetated, cooler, and dryer areas close to the ice sheet (Anderson and Brodersen 2001; Anderson, Drever, and Humphrey 1997). Microbial activity, such as that of sulfate-reducing bacteria, increases in the warmer temperatures around the upper fjord. Toward the coast there is increased influence of marine aerosols (Anderson et al. 2001). These processes and others are discussed in detail further on.

**Evaporation and salts**

The influence of distance from the coast, which controls marine inputs and the relative influence of evaporation versus precipitation, is apparent in several isotopic systems related to lake-water composition (Leng and Anderson 2003). Anderson et al. (2001) indicate that the change to negative effective precipitation occurs approximately halfway between the coast and head of the fjord (Figure 1). Together with the balance between evaporation and precipitation, the presence of a lake outlet appears to be the main control on lake salinity. Lakes can be isolated during periods of low precipitation as lake levels fall and eventually become cut off from drainage routes, forming closed basin lakes where salinity increases over time because of evaporation (Aebly and Fritz 2009; Anderson et al. 2001; Leng and Anderson 2003; Williams 1991).
the lakes in the Kangerlussuaq region, Cl is likely to be of marine origin from sea-salt aerosols and have a value of approximately 0‰ (Eggenkamp 1994; Eggenkamp, Kreulen, and Koster van Groos 1995; Zhang et al. 2007). In an open system, for example, if halite is removed by aeolian activity, δ37Cl should be depleted relative to the marine value, yielding negative isotopic signatures. Alternatively, lakes with positive δ37Cl values may be receiving δ37Cl-enriched chloride, deflated from halite precipitated from waters whose initial chloride input was marine.

δ81Br follows the opposite trend to δ37Cl, becoming more enriched in the residual solution (Hanlon 2015; Hanlon et al. 2017). In the Sand Hills of Nebraska, δ81Br was found to be more enriched than δ37Cl in the waters, while δ37Cl in salt crusts was found to be more enriched than δ81Br. δ37Cl in salts crusts was slightly enriched relative to δ37Cl in waters, while δ81Br in salt crusts was depleted relative to the waters (Frape, Stotler, and Harvey 2013; Hanlon 2015; Hanlon et al. 2017).

Lakes analyzed for δ37Cl and δ81Br are within areas 1 and 2, because the dilute coastal lakes (area 3) had insufficient concentrations of halides to perform the analyses. Both the ice-marginal and upper fjord areas were predicted to show isotopic depletion of δ37Cl values and enrichment in δ81Br because of evaporation (Frape, Stotler, and Harvey 2013; Magenheim et al. 1995; Wood, Sanford, and Frake 2005). Two trends in the δ37Cl-δ18O isotopic results are indicated on Figure 3A. The first trend (1) describes the evaporative enrichment of δ18O, which is correlated with the depletion of δ37Cl. The second group (2) of samples includes two saline and one dilute lake (L41, SS4, and 1122), maintaining a marine δ37Cl isotopic signature (0‰) despite evaporative enrichment of δ18O. However, lakes L41, SS4, and 1122 do have enriched δ81Br values, indicative of salt deflation (Figure 3B).

The majority of lake samples analyzed for δ81Br are positive, as predicted (see Figure 3B). However, a comparison plot of the δ37Cl and δ81Br results shows a poor correlation. That is, the most depleted δ37Cl values do not necessarily correspond to the most enriched δ81Br values (Figure 3C). It is clear that the impact of evaporation on δ37Cl and δ81Br signatures is complex and the limited data provided here are only a first attempt at understanding δ37Cl and δ81Br processes in Arctic lake environments.

Saline lakes tend to have excess Na versus Ca when compared to dilute lakes (Figure 4). As lake salinity increases through evaporation or by salt exclusion during winter ice formation, CaCO3 saturation may be reached and CaCO3 will precipitate, removing Ca2+ and increasing the Na/Ca ratio. CaCO3-rich layers can be observed in lake cores around the fjord Kangerlussuaq (Anderson et al. 2002, 2001; Bennike 2000; McGowan, Ryves, and Anderson 2003), supporting the precipitation of CaCO3 as a mechanism for Ca2+ depletion in saline lakes. Anderson et al. (2002) also suggest that CaCO3 precipitation may occur during the summer months when pH is increased because of photosynthesis.

**Weathering and water rock interaction (strontium isotopes)**

The 87Sr/86Sr isotopic ratios can be a useful tool for tracing the sources of salinity, because the isotopes do not fractionate during mineral dissolution and precipitation (Frape et al. 2014; McNutt et al. 1990). Thus, the 87Sr/86Sr isotopic ratio in water will directly reflect the 87Sr/86Sr isotopic ratios of the sources of strontium, such as weathering, sea-salt aerosols, or mixing with another water source (i.e., groundwater discharge). In Antarctica, 87Sr/86Sr isotopic ratios in lake waters were often observed to be similar to the 87Sr/86Sr isotopic ratios found in soil salts, derived from the weathering.

![Figure 3.](image-url) (A) δ37Cl compared to δ18O isotopic composition. δ81Br values compared to (B) δ18O isotopic values and (C) δ37Cl isotopic values. Analytical error corresponds to symbol size except where otherwise indicated by error bars.
of silicates in the nearby bedrock and regolith (Jones and Faure 1978). Green and Canfield (1984) demonstrated that the Onyx River derived a significant fraction of its salts from interaction with soils in the Wright Valley, Antarctica.

Research concerning strontium in recently exposed glacial and proglacial environments suggests that biotite weathering may have a strong influence on $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (Anderson, Drever, and Humphrey 1997; Blum and Erel 1997; Sharpe, Creaser, and Skidmore 2002). Generally, during chemical weathering, feldspars, specifically plagioclase and K-feldspar, are initially weathered (Grant 1963; Nesbitt and Young 1996). However, in glacial environments, where biotite is enriched in fine-grained sediment, biotite weathering occurs rapidly and then decreases during a relatively short time scale (10 ky) because of the loss of reactive mineral surfaces (Anderson, Drever, and Humphrey 1997; Blum and Erel 1995, 1997; Eggleston, Hochella, and Parks 1989; Nesbitt and Young 1996; Taylor and Blum 1995). The large range of $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios covered by ice-marginal lakes represents the relative influence of various mineral controls, such as biotite weathering versus feldspar (Blum and Erel 1997; McNutt et al. 1990).

A plot comparing the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio in lake waters with increasing distance from the coast (shown as Eastings, which represents the distance east from the coast toward the ice margin) reveals that lakes become increasingly radiogenic with distance from the coast (Figure 5). The regional geology in the Kangerlussuaq area is dominated by felsic and intermediate gneisses and amphibolite facies mafic intrusions. These rock types are rich in biotite and feldspars and provide a potential source for a radiogenic strontium signature. The $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic signature of the ice-marginal lakes exceeded 0.73 and is dominated by weathering of soil and rock material recently released from the melting ice. Many of these lakes, surrounded by relatively fresh glacial sediment, can have a highly radiogenic strontium isotope signature approaching 0.76 (Figure 5). For example, lakes L12, L13, and L14 are located in nonvegetated till in front of Leverett Glacier (Figure 1) and have $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of 0.7533 and 0.7576, respectively. The input of recently thawed, fine sediments transported as aeolian dust into many lakes near the ice margin would also rapidly increase the radiogenic signature.

Aeolian dust can provide a significant chemical flux, especially in glacial environments where a large amount of sediment with small particle size is available (Lamoureux, Gilbert, and Lewis 2002; Lawrence and Neff 2009). The rate of dust deposition has been shown to be greatest during glacial maxima (Lambert et al. 2008), although the Kangerlussuaq region would have been covered in ice at this time. The mineralogical composition of dust can be variable, depending on local geology, but generally contains silicate minerals such as quartz, feldspars, and phyllosilicates (Schütz and Sebert 1987). Winds in the Kangerlussuaq area are dominantly katabatic winds from the east that come off the ice sheet; however, coastal storms from the west do occur (Aebly and Fritz 2009). Easterly winds coming from the ice sheet pick up dust from the exposed glacial sediments, the ice surface, and large sand flats present in front of the ice sheet (personal observation). Aolian deposits were found to overlay glacial till over much of the L26 lake catchment (Johansson et al. 2015). It is likely that there will be increased inputs of aeolian dust...
to lakes in the ice-marginal area as glaciers retreat, leaving more extensive areas of unvegetated glacial deposits, and a corresponding increase in the input of radiogenic strontium to lakes.

In the upper fjord and coastal areas, highly radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are not observed. In these older, more weathered or geologically mature landscapes, the influence of feldspar weathering, marine aerosols, and/or deflated marine sediments, with much less radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic signatures, becomes more significant (Figure 5). Overall, a trend of decreasing $^{87}\text{Sr}/^{86}\text{Sr}$ can be observed with distance from the ice sheet (Figure 5), reflecting the reduced impact of radiogenic mineral input and weathering with time since deglaciation (Blum and Erel 1997), and an increased marine input closer to the coast. The influence of biotite weathering on $^{87}\text{Sr}/^{86}\text{Sr}$ ratios can also be observed on a plot of $(\text{Mg}^{2+} + \text{K}^+)/\text{Na}^+$ (Figure 6). Biotite weathering will contribute $\text{Mg}^{2+}$ and $\text{K}^+$ to the lake chemistry, while feldspar weathering and marine inputs will contribute $\text{Na}^+$. As a result, a pattern of increasing $\text{Mg}^{2+}$ and $\text{K}^+$ relative to $\text{Na}^+$ with increasingly radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ values is observed. Ponds and meltwater lakes show a large range of high $(\text{Mg}^{2+} + \text{K}^+)/\text{Na}^+$ ratios and radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios, suggesting that biotite weathering is a key process for these water bodies. Two outliers exist in Figure 6: L33 (Lake Ferguson) is impacted by marine $^{87}\text{Sr}/^{86}\text{Sr}$, as discussed previously. Lake L11 had high $(\text{Mg}^{2+} + \text{K}^+)/\text{Na}^+$, possibly a result of removal of $\text{Na}^+$ through halite precipitation or because of local geology (mafic material in the surrounding rock).

It has been proposed that increased weathering of glacially comminuted sediments during continental scale glaciations may have affected the marine $^{87}\text{Sr}/^{86}\text{Sr}$ record during short and longer time periods (Anderson, Drever, and Humphrey 1997; Hodell, Mueller, and Garrido 1991; Zachos et al. 1999). With an increase in glacial melting because of climate warming and the potential for enhanced geochemical loading to the oceans, further understanding of processes that affect $^{87}\text{Sr}/^{86}\text{Sr}$ in this proglacial environment will aid in the study of the interaction between the marine $^{87}\text{Sr}/^{86}\text{Sr}$ signature and glaciation.

**Sulfur oxidation and reduction**

The variability in $^{34}\text{S}$ and $\delta^{18}\text{O}$ of sulfate (Figure 7) in the lakes of the Kangerlussuaq region could be indicative of several processes: sulfide oxidation, sulfate reduction, and mixing with marine sulfate. Sulfide oxidation may cause a slight depletion of sulfur-34 (approximately 2–5.5‰) in the resulting sulfate (Toran and Harris 1989), or may result in no discernible fractionation between sulfide and sulfate (Gavelin, Parwel, and Ryhage 1960; Nakai and Jensen 1964; Seal, Alpers, and Rye 2000). Sulfate reduction that is mediated by sulfate-reducing bacteria causes enrichment in the remaining $^{34}\text{S}$ ($\text{SO}_4^{2-}$), because the bacteria preferentially use the lighter sulfur isotopes because of their lower activation energy, which maximizes the energy yield for bacterial metabolic processes (Clark and Fritz 1997; Kaplan and Rittenberg 1963). Finally, marine sulfate may be a significant source of sulfate close to the coastal regions (area 3) and the head of the fjord (area 2).

**Oxidation of sulfides**

The $\delta^{18}\text{O}$ of the sulfate produced during sulfide oxidation is a mixture of atmosphere $\delta^{18}\text{O}$ (+23‰) and the $\delta^{18}\text{O}$ of the water present during oxidation. The relative importance of atmospheric $\delta^{18}\text{O}$ versus water $\delta^{18}\text{O}$ depends on the degree of water saturation. With increasing saturation, the $\delta^{18}\text{O}$ isotopic signature of the sulfate will more closely resemble that of the water, while in relatively dryer conditions the $\delta^{18}\text{O}$ isotopic signature of the sulfate will more closely resemble that of the atmospheric $\text{O}_2$ (Clark and Fritz 1997; van Everdingen and Krouse 1985).

Sulfide minerals, including pyrite and chalcopyrite, from the bedrock core obtained during drilling of the research borehole DH-GAP04 (Figure 1), have a $\delta^{34}\text{S}$ of from 2.3‰ to 3.7‰ (Figure 7A; Table 4). A dilute lake (lake SS75) and a large, acidic (pH 3.4) pond (lake L20)
have $\delta^{34}S-\delta^{18}O$ signatures, indicative of sulfate produced during the oxidation of sulfide in the presence of modern meteoric water and atmosphere (Figure 7A). That is, the $\delta^{34}S$ (SO$_4^-$) values of lake 20 and lake SS75 ($-3.0\%o$ and $-3.9\%o$, respectively) are slightly depleted relative to that of the sulfides ($2.4\%o$ and $3.7\%o$). The $\delta^{18}O$ (SO$_4^-$) of lake 20 and lake SS75 ($-0.7\%o$ and $1.2\%o$, respectively) reflects the oxidation of sulfide in the presence of meteoric water ($\sim -19\%o$) and atmospheric oxygen ($+23.5\%o$; van Everdingen and Krouse 1985). Lakes L20, SS75, and SS76 have low Cl/SO$_4^-$ ratios (Figure 4), high SO$_4^{2-}$ as percentage of anions, and low proximity to the ice sheet (area 1). Further from and 1.2 and 3.7 $\delta^{34}S$ (SS75 and SS76, respectively) are slightly depleted $\delta^{18}O$ of lake SS75 and SS76, which are presumed to be similar to the weathered sulfur-rich rocks that were observed around lake L20. Sulfide oxidation generates H$^+$, resulting in low alkalinity and low pH (lake L20 had a pH of 3.5). Weathering of these sulfide-rich rocks would contribute increased sulfate concentrations (lower Cl/SO$_4^-$ ratios; see Figure 4). The mineral jarosite (KFe$_3$(OH)$_6$ (SO$_4$_3$_2$) was also observed in gossans in the Kangerlussuaq region (Pratt and Peng 2014), which may also impact the isotopic signature of sulfate in adjacent lakes.

A thaw lake located within the till at the front of Leverett Glacier (L12) has a similar $\delta^{34}S$ (SO$_4^-$) and depleted $\delta^{18}O$ (SO$_4^-$) to the lakes indicated earlier, where sulfide oxidation occurred in the presence of modern meteoric water (Figure 7A). In the case of L12, the $\delta^{18}O$ (SO$_4^-$) is a mixture between glacial meltwater or melt from frozen till, which has highly depleted $\delta^{18}O$ and atmospheric oxygen, resulting in sulfate with depleted $\delta^{18}O$ relative to sulfate produced from modern precipitation (Clark and Fritz 1997; van Everdingen and Krouse 1985). In other cases, such as L18 ($\delta^{18}O$ (SO$_4^-$) = 3.4%) where high evaporation rates enrich the $\delta^{18}O$ of the water ($\delta^{18}O = -5.2\%o$) and sulfide oxidation may occur under relatively dry conditions, an enriched $\delta^{18}O$ (SO$_4^-$) signature was observed (Figure 7A).

A similar effect on the $\delta^{34}S$ and $\delta^{18}O$ of SO$_4^{2-}$ as that produced by the oxidation of inorganic sulfide minerals may be caused by oxidation of organic sulfur compounds (Canfield 2001; Clark and Fritz 1997). However, the extent of SO$_4^{2-}$ derived from organic sulfur may be limited in the ice-marginal area where vegetation is fairly sparse, and cold, dry conditions limit productivity.
reduction removes sulfate from lakes by reducing \( \text{SO}_4^{2-} \) to \( \text{H}_2\text{S} \), increasing the \( \text{Cl}/\text{SO}_4 \) ratio and enriching the remaining sulfate in \( \delta^{34}\text{S} (\text{SO}_4) \). The \( \text{H}_2\text{S} \) may then react with metal ions to precipitate metal sulfides such as FeS. \( \text{H}_2\text{S} \) may also be utilized by purple sulfur bacteria, observed in the Kelliville Basin lakes (Figure 1), which oxidize the sulfide to elemental sulfur (Fry et al. 1988). The addition of marine sulfate creates a similar effect because of the higher \( \text{Cl}/\text{SO}_4 \) ratio of seawater (Figure 7B). The enrichment of \( \delta^{34}\text{S} (\text{SO}_4) \) beyond that of seawater indicates that sulfate reduction is occurring along with mixing with marine sulfate. The saline lakes around the head of the fjord are the most impacted by SRB enrichment of \( \delta^{34}\text{S} (\text{SO}_4) \). Surface-water environments become less hostile to microbial communities with distance from the ice sheet because of warmer temperatures and increased time for the evolution and development of communities. For example, cooler temperatures at about 8,200 years BP caused a definitive decrease in productivity in a lake close to the head of the fjord near Kangerlussuaq, based on isotopic and fossil records from a lake core (Anderson et al. 2008). The cooler, dryer conditions at approximately 8,200 years BP and the reduction in productivity may be related both to the close proximity of the ice sheet to the lake at that time (Anderson et al. 2008) and to thermohaline slow down documented in the North Atlantic (e.g., Alley and Ágústsdóttir 2005).

**Marine influence**

Marine aerosols have been found to impact lake chemistry in both the Arctic and Antarctic (Green and Canfield 1984; Harris et al. 2007; Jones and Faure 1978; Keys and Williams 1981; Kokelj, Zijdlik, and Thompson 2009; Lyons et al. 2005). Anderson et al. (2001) found that the percentage of Cl\(^-\) in lakes increased toward the west coast of Greenland. In southeast Greenland, an increased influence of marine-derived Na\(^+\) and Cl\(^-\) was found in shallow groundwater toward the coast (Kristiansen et al. 2013). Kristiansen et al. (2013) found that both present-day salt spray and Holocene marine sediment deposits contributed to the marine Na\(^+\) and Cl\(^-\) in the shallow groundwater. Several lines of evidence indicate that marine aerosols affect lake chemistry in the Kangerlussuaq region:

- Chloride from marine aerosols has a Na:Cl equivalent ratio of 1 and an initial \( \delta^{37}\text{Cl} \) signature of 0\%. Many of the sampled lakes have Na:Cl ratios that fall along or close to the 1:1 line (Figure 4). Some excess Na\(^+\) is observed, likely because of weathering or aeolian inputs (Anderson et al. 2001). Many of the dilute lakes as well as several saline lakes analyzed for stable chlorine isotopes have a \( \delta^{37}\text{Cl} \) isotopic value that is within analytical error of 0\%, providing further evidence of marine Cl\(^-\) input (Figure 3) throughout the study area.
- The enriched \( \delta^{34}\text{S} - \delta^{18}\text{O} \) of \( \text{SO}_4^{2-} \) found in many of the upper fjord lakes, which trend toward a marine signature, indicate mixing with marine sulfate (Figure 7A); however, it is difficult to separate this effect from enrichment because of microbial sulfate reduction.
- \( ^{87}\text{Sr}/^{86}\text{Sr} \) ratios show a pattern of decreasing radiogenic Sr toward the coast (Figure 5). While some of the decrease in \( ^{87}\text{Sr}/^{86}\text{Sr} \) ratios may be explained by a shift from biotite-dominated weathering with distance from the ice sheet, it is likely that marine aerosols (with a \( ^{87}\text{Sr}/^{86}\text{Sr} \) signature of ~0.7092) are also a factor.

The majority of lakes in the study area are above the marine limit (approximately 100 m above modern sea level; Anderson et al. 2001; Funder and Hansen 1996). The marine limit is the maximum elevation at which relict marine shorelines are observed, and thus lakes above this elevation are unlikely to contain relict seawater. A prominent exception is Lake Ferguson (L33; Figure 1), which is located below the marine limit. The \( ^{87}\text{Sr}/^{86}\text{Sr} \) ratio of Lake Ferguson (0.7187) is atypical for its distance from the coast (Easting; Figure 5) and is closer to that of seawater (0.7092). Lake L45 (Figure 1) is also below the marine limit at 96 m elevation. The \( ^{87}\text{Sr}/^{86}\text{Sr} \) isotopic ratio of lake L45 (0.7267) is typical of lakes at similar distance from the coast (Figure 5), suggesting that the lake salinity is derived from local weathering processes and aeolian activity rather than relic seawater. Lake L45 is unique in its high concentration of Cl\(^-\) relative to Na\(^+\) (Figure 4) and has a Na/Cl ratio of 0.73. The Na/Cl ratio of lake L45 is less than that of seawater (0.86), suggesting either an additional removal mechanism for Na\(^+\) or an additional source of Cl\(^-\).

The majority of ponds and dilute lakes fall along a trend of increasing Cl\(^-\) compared to \( \text{SO}_4^{2-} \) (Figure 4). Ponds, meltwater lakes, and runoff tend to have higher \( \text{SO}_4^{2-} \) relative to Cl\(^-\). The ponds and meltwater lakes sampled were located in the ice-marginal area and, as such, most likely have decreased input of marine Cl\(^-\) and limited or no removal of \( \text{SO}_4^{2-} \) by sulfate reduction. Winds in the ice-marginal area are dominantly katabatic winds from the ice sheet (Aebly and Fritz 2009), which may limit marine Cl\(^-\) inputs.
Evidence for deep groundwater discharge

It has been previously assumed that groundwater discharge into lakes in the Kangerlussuaq region is insignificant (e.g., Anderson et al. 2001). However, a number of conceptual and numerical models for groundwater flow under and adjacent to continental-scale ice sheets predict significant groundwater discharge at the front of the ice sheet (Boulton et al. 1996; Lemieux et al. 2008), with the potential for highly saline groundwater discharge (Starinsky and Katz 2003). Determining the extent of groundwater discharge into lakes in the study area was an important objective of surface-water studies. van Tatenhove and Olesen (1994) estimate that in the Kangerlussuaq area, lakes with diameters greater than 30 m do not freeze completely during the winter and, thus, are capable of supporting a closed talik. Thermal modeling may be used to predict the size of lake necessary to create a through or closed talik (e.g., Burn 2002). Thermal modeling performed as part of the GAP indicated that lakes with a width greater than 200 m have the potential to form a through talik, while lakes that are at least 100 m wide may support closed taliks (Harper et al. 2011). Thermal modeling also indicated that taliks may form during only a few 100 years (Harper et al. 2011). The relatively short time scale on which taliks may form implies that the majority of lakes capable of supporting a through talik have had sufficient time to form the talik.

Deep (subpermafrost) groundwaters measured by the GAP boreholes tend to have δ18O/δ2H isotopic signatures that fall along the GMWL and are depleted relative to surface waters. The subpermafrost groundwaters are similar in composition to glacial meltwaters (Figure 2B). These groundwaters have high sulfate concentrations with a characteristic δ34S/δ18O of SO4 (Figure 7). Groundwaters do not contain tritium and have low strontium isotopic signatures (<0.71). Significant discharge of groundwater into lakes via a through talik would cause the lake chemistry and isotopic signature to shift toward the groundwater end member; that is, anomalously low δ18O, δ2H, δ3H, and 87Sr/86Sr isotopic values and higher concentrations of SO4. None of the lakes sampled in the ice-marginal area (area 1) show evidence of groundwater discharge (e.g., Figures 2 and 7) and have 87Sr/86Sr isotopic ratios greater than 0.73.

In area 2 (head of the fjord), the geochemical and isotopic composition of the Kellyville Basin lakes bear some similarities to groundwater, such as high chloride concentrations, less radiogenic 87Sr/86Sr ratios, and enriched δ18O of sulfate; however, alternative explanations have been presented for these characteristics earlier in the article. The most compelling argument against significant groundwater discharge into lakes in the region is the lack of a mixing trend with a more depleted (δ18O/δ2H) groundwater source (Figure 2B). If groundwater discharge is occurring, volumes are too small to affect the lake isotopic composition or other geochemical parameters of the lake waters discussed in this article.

Conclusions

The use of a variety of established and novel environmental isotopes provided new insight into a number of key processes in the Kangerlussuaq region. The impact of evaporation, weathering, transport of marine aerosols, and microbial activity on lakes was found to relate to distance from the coast and ice margin, providing insight into the role that climatic gradients and glacial processes play in lake geochemical evolution.

Evaporation is one of the main processes affecting the geochemical evolution of lakes in this study, especially in the inland areas close to the ice sheet. Evaporation in closed-basin lakes is interpreted to be responsible for the higher, though still brackish, salinities (>800 µS/cm) observed in a number of the Kangerlussuaq-region lakes. Both δ37Cl and δ81Br provide new insight into the importance of salt precipitation and recycling in soils and shallow lake areas. Chlorine and bromine isotopes provide further evidence that evaporation and salt precipitation are important processes to lake geochemistry.

Highly radiogenic 87Sr/86Sr ratios indicate that biotite weathering is enhanced in more recently deglaciated catchments. Marine aerosols and feldspar weathering become increasingly important with distance from the ice sheet and with proximity to the coast. These findings support previous work (Anderson, Drever, and Humphrey 1997; Blum and Erel 1995, 1997; Sharp, Creaser, and Skidmore 2002) on enhanced biotite weathering in proglacial environments and indicate that these theories are applicable to deglaciated areas of Greenland. The findings from the Kangerlussuaq study area may have implications for the impact of glaciation and climate warming on the potential changes to the marine 87Sr/86Sr signature (Anderson, Drever, and Humphrey 1997; Hodell, Mueller, and Garrido 1991; Zachos et al. 1999). With the increased observed (e.g., van As et al. 2012) and predicted melting of the ice cap, increased runoff rates, and the potential increase in solute loadings to the
ocean (Anderson, Drever, and Humphrey 1997; Hasholt et al. 2012), increased radiogenic strontium from glaciated and recently deglaciated terrain will enter the ocean, potentially shifting the marine $^{87}\text{Sr}/^{86}\text{Sr}$ signature.

In more recently deglaciated areas, sulfide oxidation is the main source of sulfate in lakes, while the influence of marine aerosols increases around the fjord. The impact of sulfide oxidation under varying conditions (saturation, water source) is apparent in the $\delta^{18}\text{O}$ signature of the resulting sulfate. Bacterial sulfate reduction does not appear to be an important process in most of the lakes in close proximity to the ice sheet, but, in the warmer, more productive lakes around the head of the fjord, significant $\delta^{34}\text{S}$ enrichment because of microbial sulfate reduction is observed. However, it is difficult to separate the impact of sulfate reduction from mixing with marine aerosols. This study provides new information on the extent of bacterial sulfate reduction in lakes in the Kangerlussuaq area and the relationship between SRB and landscape evolution after deglaciation.

An important objective of the GAP surface-water studies was to determine the role of taliks in the groundwater system and the extent of groundwater–surface water interaction. Characterizing groundwater–surface water interaction is important for understanding lake hydrology, water balance, and chemistry. Previously, studies of lakes in the area had assumed little interaction between groundwater and surface water (e.g., Leng and Anderson 2003). Providing direct evidence for this assumption, a comparison of surface and groundwater isotopic ($\delta^{18}\text{O}, \delta^2\text{H}, \delta^{34}\text{S}, \delta^{18}\text{O}$) and geochemical data collected as part of this study found no evidence for significant groundwater discharge within the sampled lakes. The lack of observable groundwater discharge may indicate low discharge volumes and little groundwater–surface water interaction or recharge conditions, such as those observed between L26 and the DH-GAP01 borehole.

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