Lake water chemistry and the changing arctic environment

Topographic or climatic control?

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Abstract
The arctic is expected to be one of the regions most affected by ongoing climate change, with relative changes in air temperatures significantly higher than the global mean. Lakes are recognized for their potential role in the global climate system and as ecosystems of importance for local societies. As such, there is a scientific interest regarding how arctic lakes and their geochemistry will respond to climatic changes. Lakes around Kangerlussuaq (66.99 N, 51.07 W), south-west Greenland, are known for their unique geochemical composition, including oligosaline lakes, of which some are enriched in colourless dissolved organic carbon (DOC). The origin of this DOC and the importance of local catchment properties for the general water chemistry is currently being debated. This thesis aimed at: i) exploring the extent and effect of catchment morphology on lake-water chemistry in the Kangerlussuaq area; ii) determine the predominant origin of DOC, aquatic or terrestrial. I used a remote-sensing approach based on satellite imagery and digital elevation model (DEM) in deciding landscape influence on water chemistry (pH, alkalinity, conductivity, base cations, sulphate, nitrogen and absorbance). To trace the origin of the organic sources behind DOC lake water and sediments, I used a hydrogen isotope tracing method. The remote sensing approach revealed that morphological characteristics serving as proxies for lake water residence time and hydrologic connectivity (e.g. lake altitude difference and absence of outlets) explained up to 77% of the variations in lake water chemistry. The hydrogen isotopic signature of the DOC indicated a predominantly autochthonous origin, i.e. 59 to 78% was estimated to originate from algae. I conclude that lake water chemistry of the lakes in the study area is primarily controlled by the precipitation : evaporation balance, enhanced by static catchment characteristics regulating water age. Thus, the examined lake water chemical properties are likely to remain across future climatic scenarios, providing the current precipitation : evaporation balance prevails.

Key words: Søndre Strømfjord, water chemistry, morphology, remote sensing, stable isotopes.
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1 Introduction

1.1 Arctic environmental change driven by permafrost thaw
In a global warming perspective, the arctic is a key actor. Due to its high responsiveness to
global temperature rise, the arctic region has been referred to as “the canary in the coalmine”
(ACIA 2004), being a herald of the global impacts a changing climate might bring. Whilst
global mean annual temperatures (MAT) has seen increases of ~0.8°C between the period of
1888 to 2012, the arctic has displayed an accelerated warming of up to twice the global rate
during the last 50 years (IPCC 2014). By some projections, MAT in the region is set to further
increase with up to 12°C until the end of the century (AMAP 2017).

One alarming consequence of the warming arctic climate, is that permafrost temperatures
across the region have seen increases of 0.4°C to 1°C per decade since the 1980’s (AMAP 2017).
Permafrost, defined as perennially frozen ground, is playing a crucial role in the arctic
geobiosphere. Where present, it is setting the scene for terrestrial biogeochemical processes
and is a major factor in dictating arctic terrestrial ecosystems (CAFF 2013). Through inhibiting
weathering and degradation of organic compounds, permafrost is capturing and preserving
soil nutrients, major ions and carbon (C) (Frey & McClelland 2009; Schuur et al. 2015; Vonk
et al. 2015). In fact, some estimates claim that northern permafrost soils roughly contains twice
as much C as the current atmospheric C, or approximately 50% of the total global soil C pool
(Zimov et al. 2006; Tarnocai et al. 2009; Hugelius et al. 2014; Schuur et al. 2015). The high
soil C content has led to northern permafrost soils being called the “Pandoras freezer” (Brown
2013), as an input of this old C in to the atmosphere would further accelerate global warming.
Furthermore, the importance of mobilised C from thawing permafrost has been highlighted

Pan-arctic increases in permafrost temperatures have already initiated a general thickening of
the active layer (the upper soil stratum exposed to seasonal freezing and thawing), leading to
an increasing thermokarst occurrence and, in some areas, even a complete loss of permafrost
(AMAP 2017). Permafrost further exercises a primary control on landscape hydrology due to
its low hydraulic conductivity (Walvoord & Kurylyk 2016; AMAP 2017), indicating that future
impacts on freshwater might not only come from elements released from thawed permafrost,
but also via hydrological changes. Where permafrost is present, subsurface waterflows are
generally restricted to the active layer or non-frozen aquifers and taliks, effectively leading to
complex hydrological landscape dynamics on local, regional as well as temporal scales (Grosse
conditions, however, the soil hydraulic regime is set to shift, e.g., leading to a higher hydraulic
connectivity in permafrost landscapes (Walvoord & Kurylyk 2016), and deeper soil-water
pathways (Frampton & Destouni 2015; Pokrovsky et al. 2015). The thickening active layer is
thus leading to, e.g., an increased mobilization, transport and bio-availability of nutrients,
major ions, contaminants and C, previously contained within the permafrost (Hobbie et al.
1999; Klaminder et al. 2010; Pokrovsky et al. 2015; Schuur et al. 2015; Vonk et al. 2015). Upon
release, these solutes has the potential of altering fundamental geochemical and ecological
processes in terrestrial and aquatic systems throughout the landscape, with consequences to
biota reaching all trophic levels (CAFF 2013).

1.2 SW Greenland lakes, an important piece in the arctic environment
Worldwide, lakes are effective in both storing, altering and enhancing landscape
biogeochemical components. As such, they have been called “Sentinels, integrators and
regulators of climate change” (Williamson et al. 2009). Although varying between estimates
and models, northern high latitudes have been shown to contain ~200 000+ larger lakes (10
to 5000 ha; Smith et al. 2007), or one quarter of all lakes on earth (Lehner & Döll, 2004; Grosse
et al. 2013). If also taking smaller waterbodies into account, the number increases even further. In fact, according to Verpoorter et al. (2014), the highest concentration, area and perimeter of water bodies, globally, exist at latitudes between 45°N to 75°N. It has further been shown that this high-latitude abundance of water bodies is highly correlated to the presence of permafrost and glaciation history, compared to other regions of the world (Smith et al. 2007).

The Greenland landmass, even though being primarily covered by its ice-sheet, is by no means exempt from changing climate and permafrost conditions affecting hydrology and freshwater systems. The largest ice-free land mass in Greenland is located in the southwest (Figure 1; A and B), stretching from the coastal town of Sisimiut to the ice margin ~170 km to the east and is hereafter referred to as the ‘Søndre Strømfjord region’. This part of Greenland contains a remarkable concentration of lakes, with most being smaller than 10 ha (Anderson et al. 2009). Whilst most of these lakes are oligotrophic and dilute (Anderson et al. 2001), ~10% of them are oligosaline (Anderson et al. 2001; Anderson & Stedmon 2007). Several research initiatives have been carried out in the region, resulting in an extensive documentation on, e.g., lake

![Figure 1. Location maps over: A. Søndre Strømfjord region, B. Greenland as well as the C. Kellyville and D. Ice margin areas. All locales contributing with samples and analysed data are displayed with their pre-existing identification number (where existing).](image)
ontogeny, palaeoclimate and lake water chemistry. For instance, Anderson et al. (2009) showed that the standing C-stock in sediments of smaller lakes (<100 ha) make up ~50% of the soil C pool in the Sondre Stromfjord region, in only ~5% of the total land area. A large part of this sediment C is further expected to be autochthonous (Anderson et al. 2009). As described by, e.g., Anderson and Stedmon (2007), concentrations of dissolved organic carbon (DOC) in the water of many of these lakes are unusually high for these latitudes (and for lakes in general). Where DOC-concentrations are sometimes exceeding 100 mg L⁻¹, often in correlation with high conductivity. Furthermore, the DOC in these high-DOC lakes has an uncharacteristically low light absorbance (Abs.), essentially appearing as colourless (Anderson & Stedmon 2007).

The origin of the colourless DOC in the Greenland lakes is debated. Based on lake-water spectral analyses, general DOC properties and catchment residence time inference, Anderson and Stedmon (2007) made the claim that this colourless DOC was mainly a result of autochthonous DOC being strongly photodegraded. However, this has since been questioned by Holmgren (2016) who used a hydrogen (H) isotope tracing approach to estimate DOC origin. By analysing DOC in a lake known to contain colourless DOC, Holmgren found the δ²H signature of DOC (one sample) and sediments suggesting a dominance of allochthonous, rather than autochthonous, C sources. These findings, if representative of arctic lakes, indicate that even in arid arctic regions with minute runoff, terrestrial catchment processes are fundamental for lake-water chemistry. Interestingly, work by Saros et al. (2015) showed that lake-DOC in the eastern Sondre Stromfjord region decreased with 14 – 55% between 2003 and 2013. As increases in lake-water sulphate was simultaneously measured, it was hypothesised that sulphate deposition might be increasing soil ionic strength, leading to lower DOC input from the catchments. However, it was also noted that this would be somewhat contradictory to the overall limited surface runoff and groundwater input to these lakes. Hence, DOC loss within the water column through e.g. flocculation and/or microbial degradation was also suggested as plausible factors behind the decreasing DOC levels of the lakes (Saros et al. 2015; Fowler et al. 2018). Indeed, determining the original source(s) would improve our understanding regarding the drivers of the observed trend in lake water DOC.

The current paradigm on the reason for the oligosalinity and the unusually high DOC concentrations as hypothesized by, e.g., Anderson and Stedmon (2007), is that elements and organic compounds are ‘up-concentrated’ via a strong net-evaporation from the lakes (henceforth referred to as ‘evaporation effect’). This evaporation effect is a result of local climatic conditions (with evaporation superseding precipitation during summer). However, this effect appears likely to be partially coupled to catchment properties not directly linked to climate, such as topography, as catchment morphology is expected to regulate hydrological connectivity and lake water residence time (Anderson et al. 2001; Anderson & Leng 2004; Law et al. 2015). Thus, morphology regulates water age and subsequently strength of the evaporation effect. In accordance with this theory, some of the oligosaline lakes on Greenland, and elsewhere, are lacking active outflows, resulting in very long residence times (Anderson et al. 2001; Anderson & Stedmon 2007; Anderson et al. 2009).

### 1.3 Climatic and non-climatic drivers of lake water chemistry

Various efforts in reconstructing palaeoclimate and palaeohydrology have also been carried out in the Sondre Stromfjord region. By using palaeolimnological sediment records (e.g. McGowan et al., 2003; Anderson & Leng, 2004; Anderson et al. 2009; Perren et al., 2012; Law, et al. 2015), lake water sampling (Anderson et al. 2001) and through geomorphic and stratigraphic analyses (Aebly & Fritz 2009), a good understanding of lake history and ontogeny, as well as local climate fluctuations, of the region has been achieved. Especially the three large saline lakes (ss03, ss04 and ss05) and their surroundings (Figure 1; A) have been
thoroughly scrutinized. Even though climatic variations are generally being held as primarily accountable for recorded Holocene variations in e.g. lake surface levels, water chemistry and in-lake biotic composition around Kangerlussuaq (Anderson & Leng, 2004; Anderson & Stedmon, 2007; Anderson et al., 2008; Osburn et al., 2018), catchment morphology and vegetation change cannot be disregarded as possible drivers of the observed variation between lakes.

Sobek et al. (2007) showed that, generally, regional climate and topography is hierarchically acting together with lake and catchment properties to govern e.g. lake water DOC. Furthermore, biotic and abiotic catchment scale properties are well known to have an important influence on surface water chemistry at higher latitudes (Laudon & Sponseller 2018). For example, catchment scale vegetation type variations has been shown to influence DOC variability (Ågren et al. 2007) and pH (Buffam et al. 2007; Petrin et al. 2007), and catchment topography is known to influence catchment runoff and surface water residence time (Quinton & Marsh 1999; McGuire et al. 2005; Paquette et al. 2018) as well as water chemistry (D’Arcy & Carignan 1997; Laudon & Sponseller 2018; Neilson et al. 2018).

Even though less debated, the importance of catchment properties other than climate have been discussed for Greenland lakes as well. For instance, despite presenting a thorough Holocene ontogenetic record for lakes in the area around Kangerlussuaq, Aebly and Fritz (2009) states that further modelling on the hydrology and chemistry in lake basins would be beneficial to better understand the complexity of hydrologic, limnologic and biologic interplays here. This notion is further emphasized by, e.g., Law et al. (2015), who noted that the encompassing effects of climatic variations can conspire with catchment characteristics (e.g. morphology) and give rise to more individualised lake responses over time. A dynamic which has since also been observed in this region (Law et al. 2018). As initially mentioned, a rapidly changing climate is expected to lead to potential increases in hydrologic connectivity in permafrost catchments. Given the well documented history of Holocene fluctuations in climate and lakes in the Søndre Strømfjord region, further investigations in catchment specific impacts on lake water chemistry in the area around Kangerlussuaq is of importance. A better understanding of the drivers behind lake ontogeny on a catchment scale would not only complement existing work in the region, but also give insights in to potential future changes. Furthermore, thoroughly understanding hydrologic drivers in this arid paraglacial landscape could likely promote the understanding of future hydrology and water chemistry in similar regions affected by permafrost.

1.4 Aim and hypotheses

The encompassing goal for this thesis was to increase our knowledge regarding the importance of catchment landscape characteristics for lake water-chemistry in south west Greenland lakes. As mentioned, most research in this area emphasizes the importance of climatic control over these lake systems (Anderson & Leng, 2004; Anderson & Stedmon, 2007; Anderson et al., 2008; Osburn et al., 2018), only attributing catchment-scale terrestrial processes minor influence. Hence, I hypothesized:

i) That catchment properties, at least partly, explains the observed between-lake variability in water chemistry.

ii) That lake water, and hence sediment, DOC has a predominantly autochthonous origin.
2 Method
2.1 Study area
The study areas are situated at the head of the 170 km long Søndre Strømfjord, near the airport and village of Kangerlussuaq, SW Greenland (Figure 1; C). The Søndre Strømfjord region is part of the largest ice-free margin in west Greenland with some 20 000 lakes (~1 ha to >1000 ha) covering ~14% of the landscape, of which ~80% are <10 ha (Anderson et al. 2009). The bedrock mainly consist of granodioritic gneiss (Frost et al. 2001; van Gool et al. 2002; Nielsen 2010) which also forms the base material for soils in the region (Nielsen 2010). These soils are constituted of quaternary till deposits and are strongly influenced by Holocene eolian transport (Willemse et al. 2003).

The entire region is located in the continuous permafrost zone, with a varying active layer depth of ~30 cm to ~1.8 m in this area (Anderson et al. 2017). Climate in the region follows a gradient along the ~170 km long sea to ice-margin stretch. The maritime climate at the coastal town of Sisimiut (Figure 1; A), characterised by variable weather during the relatively mild winters (average January temp. -12.8°C) and cool summers (average July temp. 6.3°C), changes eastwards to a more weather-stable low-arctic inland climate, with colder winters (average January temp. -19.8°C) and warmer summers (average July temp. 10.7°C), at the town of Kangerlussuaq (Nielsen 2010). Along the same gradient, the precipitation also changes considerably from >500 mm year⁻¹ at Sisimiut to ~150 mm year⁻¹ at Kangerlussuaq (Anderson & Stedmon 2007). From about 52°W and eastwards, there is a negative precipitation : evaporation (P:E) ratio, leading to considerable water stress (Hasholt & Søgaard 1978; Anderson & Stedmon 2007; Mernild et al. 2015). Hence, hydrology is mainly driven by winter snow cover, with surface runoff only taking place during spring melt, despite maximum precipitation occurring in August (Anderson & Stedmon 2007; Nielsen 2010; Mernild et al. 2015). The water stress, in turn, leads to clear differences in vegetation cover and species composition depending on slope aspect (Nielsen 2010), with visibly scarcer cover on the more water stressed south-facing slopes. The Søndre Strømfjord region has experienced a fluctuating climate throughout Holocene with profound changes to an overall warmer and drier period during mid-Holocene (8000 – 5000 BP) that resulted in summer temperatures ~2.5°C above present (McGowan et al. 2003; Law et al. 2015). Depending on the studied lake, a period between ~5800 to 4500 BP is given as the onset of catchment changes in, e.g., vegetation following precipitation increases that preceded a generally wetter and colder period from ~4000 BP to present (Law et al. 2015). The period following ~4000 BP has seen more rapid shifts in climate (decadal to centennial) than preceding periods (McGowan et al. 2003; Aebly & Fritz 2009; Law et al. 2015). However, as suggested by Aebly and Fritz (2009), the last 700 years appear to be drier than as far back as ~6000 BP. As a result, lake water depths in the region have varied, with present day conditions resulting in several lakes east of the 52°W longitude being permanently endorheic (Aebly & Fritz 2009), or lacking outlet and/or inlet for large parts of the year. As mentioned, the majority of lakes are oligotrophic, but ~10% are oligosaline and display high conductivity (2000 – 4000 μS⁻¹) (Anderson et al. 2001; Anderson & Stedmon 2007). DOC concentrations in lakes in the east of the Søndre Strømfjord region are generally higher than would commonly be expected of high latitude lakes, with highest DOC concentrations being recorded in the oligosaline lakes west of Kangerlussuaq (Anderson & Stedmon 2007). It is noteworthy that, as the marine limit in the studied area is at ~60 m a.s.l. (due to isostatic uplift) and lakes with increased conductivity are found well above 100 m a.s.l., the salinity is not of relict marine origin (Anderson et al. 2001).

Field work was conducted between the 3rd and 14th August 2017, in the area around the town of Kangerlussuaq. Two areas of interest (AOI) were selected; the Ice Margin area (IM), located by the Russel glacier, and the Kellyville area (KV), situated south west of Kangerlussuaq.
(Figure 1; C and D). A total of 17 sample sites, in 16 lakes, were used and denoted Kangerlussuaq Sampling (KS). Five lakes (KS1 – KS5) were sampled for reference at IM as lake DOC concentrations are expected to be much lower at IM than at KV (Anderson & Stedmon 2007). IM is also affected by strong katabatic winds, descending from the nearby Greenland ice-sheet. As a consequence, lakes in this area are under higher influence of eolian material input than those at KV, with a potentially higher effect on lake water chemistry (Rydberg et al. 2016). At KV, eleven lakes (KS6 – KS17). Note that the majority of lakes sampled in 2017 are occurring in published data (e.g. Anderson & Stedmon, 2007; Saros et al., 2015; Osburn et al., 2018) and that a comparison between pre-existing and my labelling is provided in Table 1. To facilitate cross-study referencing, the established denominations will hereafter be favoured. The most notable feature of the KV area is the three large oligosaline lakes, ss03, ss04 and ss05, locally known as Hundeso, Brayaso and Limnaeso, to the south in this area (Figure 1; C). These three lakes (together with several other of the smaller nearby lakes) have previously been joined together (Aebly & Fritz 2009). Thus, forming one large waterbody that drained over a sill at 186 m a.s.l. to the south of ss05. Evidence of this can, e.g., be seen in sets of palaeoshorelines adjacent to ss03, ss04 and ss05. Furthermore, Aebly and Fritz (2009) recorded additional, submerged, palaeoshorelines in Brayaso. Together with sediment sample analyses, a reconstruction of lake-level fluctuations and regional climate was made. This showed that lake surface in the basin of the three lakes has varied ~29 m during Holocene, following the previously described climatic variations. Furthermore, the lake level of ss05 and ss06 has increased during the last 20 years (Leng & Anderson 2003; Law et al. 2018). Consequently, ss05 and ss06 are no longer separated, but connected via an estimated 2 – 3m shallow submerged ridge which gives that sample points KS6 and KS7 are located in the same lake.

2.2 Obtaining, treating and analysing data
The 2017 sampling effort was supplemented with sediment-, biota- and soil samples from several additional lakes, sampled during previous research initiatives in the region. Similarly, geochemical data from water and sediments was synthesised from existing data sets of such initiatives. These covered the 2017 sampled lakes, as well as numerous additional lakes from the coast to the ice sheet, ranging back to the year 1998. All supplemental data and samples were kindly provided by Prof. John N. Anderson of Loughborough University.

2.2.1. Solid Phase Extraction
Solid phase extraction (SPE) was used in field extraction of lake-water DOC for H-isotope analysis, similar to the method employed by Holmgren (2016). The SPE method, described by Dittmar et al. (2008), provides a simple, weight-efficient and effective method of extracting and storing water DOC until analysis. As such, the method is highly suitable for, e.g., sampling procedures under logistical constraints. Briefly, a set volume of sampled water is passed through a prepared cartridge containing a sorbent of styrene divinyl benzene polymer (PPL-cartridge). DOC is filtered out in the sorbent where it is efficiently stored until extraction procedures can be applied. Through this method Dittmar et al. (2008) reported DOC

<table>
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<th>2017 sampling</th>
<th>Pre-existing denomination</th>
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<tr>
<td>KS1</td>
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<td>KS2</td>
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extraction efficiencies of ~62% in ocean water. Prior to fieldwork around Kangerlussuaq, PPL-cartridges were primed in laboratory at the department of Ecology and Environmental Science, Umeå University, by rinsing with one cartridge volume of methanol, then letting the cartridges soak overnight in methanol before rinsing with two cartridge volumes of Milli-Q water. An additional cartridge volume of methanol was then passed through. Cartridges was subsequently kept methanol saturated until usage.

2.2.2 Field sampling
Logistical constraints limited SPE water-sampling to sets of maximum 3 lakes per day. From each lake, 2 L water (3 L from lakes ss901, ss902, ss903 and KS3) was obtained from approximately 0.5 m below the surface. This was done from the shoreline, using a 4 m long telescopic rod with a 1 L bottle attached. Samples were subsequently brought back to base camp (BC) for further processing. Each sample container was rinsed in lake water 3 times prior to sampling. SPE in BC was performed using an adaptation of extraction procedures described by Dittmar et al. (2008). To increase the PPL-cartridge adsorption efficiency, two cartridge volumes of 0.02 M HCL were passed through prior to filtering. DOC/H extraction was then made by gradually filtering 1 L (2 L for lake ss901, ss902, ss903 and KS3) of water through pre-combusted (4 h, 500°C) 0.45 µm Whatman GF/F glass fibre filters, prior to passing it through a Varian Bond Elut 6 mL PPL-cartridge. Roughly 9 h was needed per 1 L DOC/H extraction, except for ice-margin lakes where approximately half the time was needed per litre. Excess sample-water was used for backup and equipment rinsing. After completed extraction, sample cartridges were emptied of porewater using a vacuum pump and stored cool (sealed in zip-lock bags in the soil at permafrost depth) until they could be kept frozen. All sample treatment procedures were kept to one of the two tent vestibules in BC, to minimize risks of contamination and sample disturbances. The vestibule in tent shadow was especially chosen for this, to keep the temperature as level as possible and avoid sunlight during extraction. Extraction setup is shown in Figure 2.

Figure 2. Field DOC/H extraction set up. a) Loaded 60 mL syringes with attached filters, b) Styrofoam cooler box for sample transportation, c) Sample bottles used for extraction, d) Reserve samples for extraction and equipment cleanings, e) PPL extraction setup: 60 mL syringe tubes as containers connected to PPL cartridges with adapters, which are in turn connected to custom made light-weight 300 mL vacuum containers.
Samples for absorbance as well as DOC concentration and total dissolved nitrogen (DN) was also taken in conjunction to DOC water-sampling, by filtering sampled water through Whatman 0.2 µm single use filters in to a 50 mL falcon tube. In addition to this, individual samples of two common macrophyte groups were collected from lakes ss1400 and ss07 for reference analysis of H-isotopic signal (4 samples in total). These were collected in the pelagic zone and put in water-filled 50 mL falcon tubes. All samples were subsequently field-stored in the ground at permafrost depth together with PPL-cartridges until they could be frozen. Throughout the entirety of the fieldwork period, lakes, catchments and features of particular interest were photographed and documented for spatial analysis and reference.

2.2.3 Extraction of DOC for hydrogen isotope analysis
Prior to laboratory extraction of DOC for H analysis, PPL cartriges were further dried through N2 flushing. DOC extraction was then performed by passing one cartridge volume (6 mL) of methanol through PPL cartridges (Dittmar et al. 2008). The methanol-DOC solution was collected in glass vials and sealed with air-tight Teflon septum. Using a 0.2 mm needles inserted in the septum, a constant flow of N2 at 2 Bar was passed through the vials until the methanol was completely removed. Extracted DOC was then dissolved in Milli-Q water for transfer to polyethylene scintillation vials, and subsequently freeze-dried prior to steam equilibration analysis.

2.2.4 H-isotope analyses and steam equilibration
H isotope tracing is extensively used in determining the origin of organic carbon matrixes, e.g., in sediment or soil samples for palaeoclimatological investigations (Sauer et al. 2009; Holmgren 2016; Gudasz et al. 2017). The method is based on the two stable isotopes of H, protium (1H) and deuterium (2H) having a mass difference by a factor of ~2 (Schimmelmann et al. 2006), which is a higher relative difference than any other two isotopes of the same element (Chriiss 1999). Biological and chemical processes might favour reactions involving the lighter isotope (H1) and thus, continuously change the relative abundance of the two H isotopes. Consequently, material with different biological or chemical origin may differ in their 2H/H ratio (Schimmelmann et al. 2006). This ratio is often expressed as a δ2H value, with a null-reference value established as 0‰ in the reference material Vienna Standard Mean Ocean Water (VSMOW) (Schimmelmann et al. 2006). Studies have shown that there is a difference in the δ2H ratio of organic material coming from algae and that of terrestrial plants (Smith & Ziegler 1990; Karlsson et al. 2012; Rose et al. 2015). As described by, e.g., Smith and Ziegler (1990), some of the reasons behind this difference are (as listed by Rose et al. 2015): i) Enrichment of δ2H in terrestrial plants due to evapotranspiration, ii) concentration differences of lipids between algae and terrestrial plants (lipids tends to be δ2H depleted) and iii), discrimination against δ2H within the algae during photosynthesis.

The rationale for the use of hydrogen isotopes in my study is, in short, that terrestrial and aquatic/pelagic DOC sources will have vastly differing isotopic signatures. Hence, by establishing δ2H reference values for terrestrial and aquatic endmembers (such as humus-soil and plankton, respectively), it is possible to estimate the relative origin of, e.g., the DOC within the water column. A major issue with H-isotope tracing however, is the effect of exchangeable H (Hex) that are adsorbed on the surface of the material of interest that may distort the signal from the solid organic matter (Schimmelmann 1991; Wassenaar & Hobson 2000). Hex can bind to both the mineral matrix, as well as to the organic material of samples and does not reflect the isotopic signature of the organic material itself, as it equilibrizes in ambient temperature and moisture (Ruppenthal et al. 2013). Hence, to obtain accurate δ2H values of the solid matter in question (in this case, DOC), it is imperative to remove all the Hex fraction in samples. Removal of the mineral matrix is often done through the use of hydrofluoric acid (Ruppenthal
et al. 2013). Another method, e.g. applied by Holmgren (2016), instead applies calibration through the use of composite samples with known C and H concentrations. Removal of \( H_{ex} \) from the organic matrix can be done through, e.g., dual isotope steam equilibration. Here, two sets of sample replicates are exposed to two different water samples of known H isotopic compositions in an equilibration process. A correction for the \( H_{ex} \) found in the organic matrix can then be made to obtain a correct \( \delta^2H \) value for the organic matrix (Ruppenthal et al. 2013). As shown by Holmgren (2016) and Gudasz et al. (2017), it is then further possible to utilize the obtained \( \delta^2H \) value from, e.g., lake-sediments together with \( \delta^2H \) values from terrestrial (plants) and aquatic (pelagic algae) endmembers to ascertain the level of allochthony of the examined matrix.

In my study, I adopted the steam equilibration method previously used by Holmgren (2016) when analysing the H-isotopic composition in lake water DOC, macrophytes, lake sediments and reference endmembers (algae samples for water and terrestrial plants and humus soil samples for terrestrial). Briefly, sample preparation consisted of thoroughly grinding samples in a steel ball mill and passing them through a sieve (212 µm pore size) to remove larger fractions. To establish the target weights for adequate H mass (18 µg to 25 µg) in further analyses, the samples were analysed at the Evolutionary Biology Centre at Uppsala University for Nitrogen (N), C and H concentrations. A total of 118 individual sub-samples, together with 15 samples of the reference material KHS (Kudu Horn Standard; Coplen 2017), was then extracted in to 0,04 mL (Ø 3,5 mm / 15,5 mm) silver containers (Analysentechnik GmbH & Co. KG) prior to being dried. As described by e.g. Qi and Coplen (2011), it is imperative to thoroughly dry samples prior to steam equilibration for optimizing high-quality measurements. As such, each batch of 133 samples was placed in holding brass plates in four sets of 28 and one of 19 with every plate containing 3 reference samples spread out over the plate (Figure 3; A). The brass plates containing the samples, together with a sixth (empty) to maintain even temperature, were subsequently placed in a vacuum oven (Figure 3; B) and passed through an 8-hour equilibration protocol at 105°C. This included two initiating cycles of air evacuation with subsequent helium flushing. The initiation protocol was followed by 6

Figure 3. A) The five brass-plates used for holding silver capsules during the drying process. Red circles on plate A are displaying the reference sample positioning used on all plates. B) Vacuum oven loaded for drying protocol. Note that six brass plates (one empty) were used to ensure even temperatures during the drying processes.
days of drying at 60°C and 0.1 millibar air pressure. Dried samples were then removed from the oven and immediately crimped tight to avoid moisture uptake. These were subsequently stored in helium flushed cylinders until δ²H analysis with a thermo official Isotope ratio Mass Spectrometry (IRMS) was performed at the Swedish University of Agricultural Sciences, Umeå. This largely followed the method described by Ruppenthal et al. (2013) and further adopted by e.g. Holmgren (2016) and Gudasz et al. (2017). However, here I used two waters with known H isotopic compositions of ~400‰ and 385‰.

2.2.5 Light absorbance, DOC and DN concentration
Light absorbance analysis on lake water was performed, using a 50mm cuvette, in a JASCO Corp, V-560, spectrophotometer with the Rev. 1.00 software. Samples KS6 – KS17 were also analysed using a 10mm cuvette to account for low absorbance. Calibrations with milli-Q as blanks was made prior to each cuvette batch as well as approximately every fifth sample. DOC (mg L⁻¹) and DN (µg L⁻¹) concentrations were analysed with a Formacs combustion TOC/TN analyser (Skalar Analytical B.V.) at the department of Ecology and Environmental Science, Umeå University.

2.3 Remote sensing and spatial analysis
Prior to fieldwork, location data of previously sampled lakes in the region was obtained from published material (Anderson & Stedmon 2007) to ensure a level of comparability through the use of existing data. Together with satellite imagery from the European Space Agency (ESA) Sentinel 2 satellite (https://scihub.copernicus.eu/dhus/#/home), location data was used in ESRI™ ArcMAP 10.5® to produce maps for in situ evaluation and decision of sample locations.

2.3.1 Digital Elevation Model processing
To extract landscape morphology parameters, catchments were digitally delineated for 190 water bodies (~0.07 ha to ~510 ha) in and around the two AOIs, using the ArcHydro tool-package in ESRI™ ArcMAP 10.5®. After extensive search for an adequate elevation model, the Arctic DEM (Digital Elevation Model) mosaic data set with a resolution of 5 by 5 meters (0.0025 ha pixel⁻¹), provided by Polar Geospatial Centre at the University of Minnesota (https://www.pgc.umn.edu/data/arcticdem/), was chosen for spatial and morphological analyses due to its high resolution and availability. The lakes in the AOIs were manually delineated as polygons, using satellite imagery from Sentinel 2 together with the DEM elevation data as a base reference. Due to the endorheic nature of the lakes in the AOIs, adaptation of established watershed delineation routines (e.g. ESRI 2011) was necessary. This was done by creating holes in the input DEM raster, effectively making each individual lake a “bottomless” sink. In the workflow, this process is added between pre-processing the DEM by levelling the lake surfaces (’Level DEM’ tool) and smoothing the DEM surfaces (’Fill depressions’ tool), prior to the flow pointer raster creation. From the extensive catchment data output, the relevant catchments were then chosen for use in further data extraction.

2.3.2 NDVI
Normalized difference vegetation index (NDVI) was calculated and used for vegetation cover analysis, using spectral bands 4 (visible red, 664.5 nm ±49 nm) and 8 (near infra red (NIR), 835.1 nm ±72.5 nm) from Sentinel 2 (SUHET 2013). This imagery was obtained by satellite passes on 20170710 for the Kellyville area, and 20170706 for the Ice Margin area and has a resolution of 0.01 ha pixel⁻¹. The NDVI calculations were performed using raster calculator in QGIS 2.18.16 (QGIS Development Team 2018), applying equation (Eq. 1)

\[ NDVI = \frac{NIR-Red}{NIR+Red} \]  

Eq.1
Where *Red* is the visible red spectrum. Resulting raster output was imported to ESRI™ ArcMAP 10.5®, where further classification and referencing of NDVI-values was done through visual cross-referencing with photographs taken in the AOIs during sampling. A method example is given in Figure 4 with reference photographs as well as NDVI raster and satellite imagery. Lake water areas in the NDVI raster was removed prior to catchment specific calculations in areal coverage of eight defined NDVI classes, as well as total mean NDVI value, that were used in further analyses.

![Figure 4](image)

*Figure 4. Example of cross-referencing process using remote sensing and photography, around lakes ss06 and ss07. Images at the top are showing; a) Intermittent inlet flow-path with richer vegetation, b) Dry outlet zone from ss07, predominantly dried grass and gravel with some small shrubs, and c) View from West over North and South facing slope aspects, note that the south facing slope is primarily soil, rocks and some dry grass. Locations of photographed areas are marked in: 1) NDVI raster classified in 8 classes, and 2) Satellite image. Note that light-green areas in 2.) are shallower lake bottom between ss06 and ss05, visible thanks to low light absorbance in the water.*

### 2.3.3 Morphological analyses

Morphological landscape features were analysed using the Spatial Analyst toolbox in ESRI™ ArcMAP 10.5®. Data were subsequently extracted for each relevant lake-catchment using the tool ‘Zonal Statistics as Table’, after which further selection ensued. Additionally, lake depths were compiled from Anderson and Stedmon (2007), Whiteford *et al.* (2016), Osburn *et al.* (2018) and from data provided by Anderson, N. J. of Loughborough University. The selected morphological features used in this study are summarized in Table 2.

To assess the potential for lakes to have in- and outlets, a combination of visual assessment, spatial and photogrammetric analyses, as well as review of previously published material on the area was employed. The water stress on the area is driving most lakes to be primarily endorheic and is further limiting the conditions for surficial runoff through streams. Hence,
classifying potential in- and outlets and assessing landscape flow paths was done with two main assumptions; 1) Each lake has an altitude threshold that, when reached by rising water level, causes the lake to overflow and subsequently accelerate the turnover rate of the lake water. 2) Paths of flowing water in the area can generally not be regarded as conventional streams. Instead, they should be viewed as intermittent flow paths with a catchment area threshold (i.e. the drainage area needed for a flow-path to occur). Most often, the vegetation will respond to these flow paths as they, even in non-surface flow conditions, have a higher soil moisture content. As such, this should be traceable through NDVI analysis of vegetation health and type (Figure 4; a). The flow-path can however exist with a smaller catchment area if it is “draining” an overflowing lake (Figure 4; b). Using these assumptions, the working order for deciding lakes with potential in- and outlets of significance for the water chemistry went as follows: Using the levelled DEM, all lakes were virtually filled to their overflow level. Multiple, potential, flow path layers were then created, each having a different catchment area threshold. These flow path layers were then cross-referenced to the NDVI classes, field observations and to different spectral band-combinations of the Sentinel 2 imagery (SUHET 2013) highlighting wetter ground. A satisfying match between stream occurrence and vegetation occurrence was achieved at 17.5 ha drainage area to each stream pixel. Furthermore, by using the obtained filled lake DEM and the initially levelled DEM, the lake altitude difference was obtained by using the Raster-Calculator tool. ‘Mean slope’ was chosen as a rough representation of catchment morphometric variations, as topography has been known to have a high order on influence on water runoff rates (Quinton & Marsh 1999; McGuire, K. J. et al. 2005; Paquette et al. 2018) and surface water chemistry (D’Arcy & Carignan 1997; Laudon & Sponseller 2018). ‘Mean aspect’ was included with regards to the findings of e.g. Nielsen (2010) and Rydberg et al. (2016). Their results indicating that, as south facing slopes are having poor vegetation cover (seen in Figure 4; c), the prevalence of a south facing slope aspect in a catchment would potentially influence terrestrial sediment input through increased eolian erosional transport or through surficial runoff.

<table>
<thead>
<tr>
<th>Morphologic factors</th>
<th>Explanation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean slope</td>
<td>Extracted mean catchment slope as indication of catchment steepness.</td>
</tr>
<tr>
<td>Mean aspect</td>
<td>Extracted mean catchment slope aspect (of 360 degrees) as proxy for vegetation coverage/quality and sun irradiance.</td>
</tr>
<tr>
<td>Lake area</td>
<td>The lake area as extracted by manually created polygons.</td>
</tr>
<tr>
<td>Lake depth</td>
<td>Maximum depths are compiled from Anderson and Stedmon (2007), Whiteford et al. (2016), Osburn et al. (2018) as well as from data provided by Prof. Anderson, N. J. of Loughborough University.</td>
</tr>
<tr>
<td>Lake altitude</td>
<td>Present day altitude, extracted from the leveled DEM.</td>
</tr>
<tr>
<td>Lake altitude difference</td>
<td>The difference between present day lake altitude and the potential maximum lake-level altitude.</td>
</tr>
<tr>
<td>Outlet within 3m</td>
<td>Possibility for lake to have an outlet within 3m of lake level increase (analysed as two factors, where; 1 = yes, 2 = no).</td>
</tr>
<tr>
<td>Lake volume / Catchment area</td>
<td>Calculated area quota as proxy for catchment area influence.</td>
</tr>
<tr>
<td>Catchment area / Lake area</td>
<td>Proxy for runoff ratio.</td>
</tr>
<tr>
<td>Lake area / Lake depth</td>
<td>Calculated quota as proxy for volume and residence time.</td>
</tr>
</tbody>
</table>
2.4 Handling analyses and calculations

2.4.1 $\delta^2$H analysis

Output data of the steam equilibration analysis was treated as described by Holmgren (2016), with the final step being a binary mixing model yielding the percentage of allochthony within samples. The steps involved in calculating the $H_{ex}$ percentage, the non-exchangeable H isotopic composition and the allochthonous C percentage are shown in Eq. 2 to 4;

$$H_{%ex} = \frac{\delta^2H_{L} - \delta^2H_{ex}}{\delta^2H_{LRW} - \delta^2H_{HRW}}$$ Eqa. 2

With $H_{%ex}$ being the $H_{ex}$ percentage, $\delta^2H_{L}$ and $\delta^2H_{ex}$ being the isotopic composition of samples equilibrated with light and heavy (respectively) reference water and $\delta^2H_{LRW}, \delta^2H_{HRW}$ being the isotopic compositions of light, respectively heavy, reference water.

$$\delta^2H_{Non-ex} = \frac{\delta^2H_{L} - H_{Non-ex} \times \delta^2H_{LRW}}{1 - H_{%ex}}$$ Eq. 3

Where $\delta^2H_{Non-ex}$ is the isotopic composition of non-$H_{ex}$ in the sample.

$$Allochthonous\ C(\%) = 100 \times \frac{(\delta^2H_{Non-ex} \times \delta^2H_{Aq})}{(\delta^2H_{Terr} - \delta^2H_{Aq})}$$ Eq. 4

With $\delta^2H_{Aq}$ being the isotopic composition of the aquatic endmember and $\delta^2H_{Terr}$ being the terrigenous endmember isotopic composition.

2.4.2 Absorbance measures

For ease of comparing lake water absorbance to previous studies (e.g. Holmgren 2016, Anderson & Stedmon 2007), Specific Ultra Violet Absorbance at 254 nm (SUVA254) (Abs. 254 DOC$^{-1}$) and the absorbance at 375 nm ($a_{375}$) ratio to DOC concentration ($a^*_{375}$) (Abs. 375 DOC$^{-1}$) was calculated for each sample. SUVA254 is a commonly used proxy for measuring DOC aromaticity in water (Traina et al. 1990; Weishaar et al. 2003), whereas $a^*_{375}$ has been employed by e.g. Anderson and Stedmon (2007) and Osburn et al. (2018) as a qualitative proxy for indicating the rate of DOM absorbance in lakes of this region.

2.5 Statistics and testing of hypothesis 1

Statistical analysis and exploration was done in the software R (R Core Team 2017) and SPSS (IBM Corp. 2016). To convert measured between-lake variability in the water chemistry into simpler dimensions, I performed a Principal Components Analysis (PCA). Briefly, a PCA enables the breakdown of data sets of multiple variables (dimensions), such as the lake water chemical factors in this case, in to fewer dimensions by grouping variables that are showing similar variance. These dimensions are called principal component (PC) scores, with scores (n $> = 1$) in numerical order corresponding to the amount of variance (highest to lowest) explained in the data set. Furthermore, when grouped in to PC scores, additional correlation tests can be done on factors suspected to influence the PCA variable data set. The PCA was conducted on data sampled in July 2001, synthesised from the datasets provided by Anderson N.J. and consisted of the twelve variables: pH, Alkalinity (Alk.), conductivity (Cond.), natrium (Na), ammonium (NH$_4$), potassium (K), magnesium (Mg), calcium (Ca), chloride (Cl), nitrate (NO$_3$), sulphate (SO$_4$) and Abs. 250nm. These parameters were acquired for the 29 lakes of the KV area displayed in Figure 1; C. I performed a correlation analysis using the PCA scores as proxy for water chemistry to further test hypothesis 1; that catchment properties are regulating lake water residence time. And hence, that the influence of ‘Evaporation effect’ is larger than that of landscape features such as slope, vegetation type (and coverage) and altitudes, as opposed
to regions with stronger hydrological connectivity. Given the non-normal distribution of the landscape properties, I used Spearman’s rho rank analysis.

3 Results

3.1 Water chemistry

Concentrations of DOC and DN for the 2017 sampling revealed higher concentration in the KV area than at the IM (Figure 5). Most notably, the oligosaline lake ss1421 was displaying highly elevated concentrations (126.4 mg DOC L\(^{-1}\) and 3126 µg DN L\(^{-1}\)) in comparison to the other lakes of the KV area (including the other oligosaline lakes ss06, ss05, ss04 and ss03), whilst the adjacent lake ss1381 is showing some of the lowest concentrations of the KV samples (49.2 mg DOC L\(^{-1}\) and 1271 µg DN L\(^{-1}\)). Lake ss02 showed comparably low concentrations of both DOC and DN (40.3 mg DOC L\(^{-1}\) and 1049 µg DN L\(^{-1}\)), being between lake ss1341 (36.6 mg DOC L\(^{-1}\) and 851 µg DN L\(^{-1}\)) and ss1381, approaching the concentrations of IM samples.

![Combined diagram over the two absorbance measures (top) and DOC and DN concentrations (bottom) for the 17 sampled lakes in 2017. Crosses show SUVA254 (Abs. 254 DOC\(^{-1}\)) and filled triangles a\(^{375}\) (Abs. 375 DOC\(^{-1}\)). Empty diamonds show DN (µg L\(^{-1}\)) and filled circles DOC (mg L\(^{-1}\)). Dashed line separates IM and KV areas.](image-url)

Absorbance, as SUVA\(_{254}\) and a\(^{375}\), also displayed marked difference between the IM and KV areas, with IM lakes generally showing higher levels of absorbance and ss096 having the highest measured absorbance (a\(^{375}\) = 0.0035 DOC\(^{-1}\), SUVA\(_{254}\) = 0.0247 DOC\(^{-1}\)) of all the samples. The two exceptions to the low absorbance in the KV area, ss1400 (a\(^{375}\) = 0.0013 DOC\(^{-1}\), SUVA\(_{254}\) = 0.0136 DOC\(^{-1}\)) and ss07 (a\(^{375}\) = 0.0020 DOC\(^{-1}\), SUVA\(_{254}\) = 0.0177 DOC\(^{-1}\)), share the common traits of: i) being relatively small (< 5 ha), ii) hosting thriving macrophyte
communities (Figure 6), iii) having visually coloured water and, iii) not having another lake higher up in the catchment.

Figure 6. Pictures of lake ss1400 and ss07 in the KV-area. Notice the abundant macrophyte communities.

3.2 Catchment properties and lake water chemistry
Visual selection of catchment NDVI into landscape classes resulted in 8 classes, ranging from 1: No vegetation (water and/or rocks), to 8: Healthy vegetation (rarely occurring). A complete list of the classes, their NDVI values as well as a brief description of their criteria is found in Table 3.

<table>
<thead>
<tr>
<th>NDVI values</th>
<th>Class number</th>
<th>Landscape type</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 - 0.1</td>
<td>1</td>
<td>Water or rocks. No vegetation.</td>
</tr>
<tr>
<td>0.1 - 0.3</td>
<td>2</td>
<td>Rocks, bare earth / soil, sparse and / or dry vegetation. E.g. dry wetland grasses.</td>
</tr>
<tr>
<td>0.3 - 0.35</td>
<td>3</td>
<td>Dry grass / high stone content and slightly less dry wetland grasses.</td>
</tr>
<tr>
<td>0.35 - 0.4</td>
<td>4</td>
<td>Intermediately dry vegetation. Increasing dwarfshrub content. Increasing content of healthy vegetation.</td>
</tr>
<tr>
<td>0.4 - 0.5</td>
<td>5</td>
<td>Vegetational shift in the spectra. Beginning with primarily dwarfshrubs, followed by increasing Betula nana (shrubs) and followed by increasing salix spp. occurrence. Also, to some extent, Indicative of increasingly healthy vegetation, e.g. N and NW facing slopes.</td>
</tr>
<tr>
<td>0.5 - 0.54</td>
<td>6</td>
<td>Increasingly healthy dwarf-shrub vegetation. Not necessarily indicative of increasing salix content.</td>
</tr>
<tr>
<td>0.54 - 0.6</td>
<td>7</td>
<td>Mainly salix and / or healthy, &quot;well growing&quot; , vegetation. Mainly occurring in potentially wetter areas such as depressions and potentially intermittent outflow zones.</td>
</tr>
<tr>
<td>0.6&lt;</td>
<td>8</td>
<td>Healthy vegetation (rarely occurring).</td>
</tr>
</tbody>
</table>

From the PCA output, PC scores one to four were significant. These were selected as representative for the water chemistry and subsequently used for evaluation of hypothesis 1. Together they explained ~91.4% of the variance within the data set, with PC1 explaining 60.35% and PC2-4 explaining 11.81%, 10.87% and 8.40% (respectively) of the variance (Table 4). In short, PC1 mainly described salinity coupled variation, while PC 2 explained variation...
relating to nutrients such as nitrogen. PC3 was representing variation in Ca concentrations, whilst PC4 captured variability primarily relating to the lake water colour Table 4).

Table 4. Correlation coefficients between water chemical variables and PC-scores one to four. Percentage of total variance explained in parenthesis and PC score interpretations given. High correlation coefficients highlighted in grey.

<table>
<thead>
<tr>
<th>Variables</th>
<th>PC1 (60.35%)</th>
<th>PC2 (11.82%)</th>
<th>PC3 (10.87%)</th>
<th>PC4 (8.40%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>0.860</td>
<td>-0.218</td>
<td>-0.035</td>
<td>0.249</td>
</tr>
<tr>
<td>Alk.</td>
<td>0.949</td>
<td>0.046</td>
<td>-0.096</td>
<td>0.111</td>
</tr>
<tr>
<td>Cond.</td>
<td>0.994</td>
<td>0.084</td>
<td>-0.038</td>
<td>0.01</td>
</tr>
<tr>
<td>Na</td>
<td>0.988</td>
<td>0.093</td>
<td>-0.08</td>
<td>0.002</td>
</tr>
<tr>
<td>NH₄</td>
<td>-0.297</td>
<td>0.632</td>
<td>-0.536</td>
<td>0.23</td>
</tr>
<tr>
<td>K</td>
<td>0.991</td>
<td>0.089</td>
<td>-0.029</td>
<td>-0.051</td>
</tr>
<tr>
<td>Mg</td>
<td>0.983</td>
<td>0.095</td>
<td>-0.082</td>
<td>0.011</td>
</tr>
<tr>
<td>Ca</td>
<td>0.103</td>
<td>0.078</td>
<td>0.915</td>
<td>0.229</td>
</tr>
<tr>
<td>Cl</td>
<td>0.984</td>
<td>0.101</td>
<td>-0.051</td>
<td>0.006</td>
</tr>
<tr>
<td>NO₃</td>
<td>-0.276</td>
<td>0.682</td>
<td>0.202</td>
<td>0.549</td>
</tr>
<tr>
<td>SO₄</td>
<td>0.729</td>
<td>0.187</td>
<td>0.297</td>
<td>-0.257</td>
</tr>
<tr>
<td>Abs 250 nm</td>
<td>-0.125</td>
<td>0.648</td>
<td>0.149</td>
<td>-0.677</td>
</tr>
</tbody>
</table>

I found significant correlation between landscape features influencing lake water residence time (i.e., supporting the evaporation effect theory) and water characteristics represented by PC1-4 (Table 5). Generally, features coupled to lake morphology and bathymetry (e.g., ‘Lake area’ and ‘Outlet within 3m range’) are showing significant and high correlations with PC1

Table 5. Summary of Spearman’s rho correlations between catchment and lake morphometrics to PC scores one to four with corresponding percent of data set variance displayed in parenthesis. Two levels of significance given as asterisks.

<table>
<thead>
<tr>
<th>Spearman’s rho (2-tailed)</th>
<th>PC1 (60.35%)</th>
<th>PC2 (11.82%)</th>
<th>PC3 (10.87%)</th>
<th>PC4 (8.40%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean slope</td>
<td>0.220</td>
<td>0.049</td>
<td>0.189</td>
<td>-0.199</td>
</tr>
<tr>
<td>Mean aspect</td>
<td>0.105</td>
<td>-0.114</td>
<td>0.045</td>
<td>-0.053</td>
</tr>
<tr>
<td>Lake area</td>
<td>0.631**</td>
<td>-0.410*</td>
<td>0.035</td>
<td>0.386*</td>
</tr>
<tr>
<td>Lake depth</td>
<td>0.499**</td>
<td>-0.196</td>
<td>-0.028</td>
<td>0.298</td>
</tr>
<tr>
<td>Lake altitude</td>
<td>-0.568**</td>
<td>0.408*</td>
<td>-0.201</td>
<td>0.439*</td>
</tr>
<tr>
<td>Lake altitude difference</td>
<td>0.688**</td>
<td>-0.379*</td>
<td>-0.043</td>
<td>-0.243</td>
</tr>
<tr>
<td>Outlet within 3m range</td>
<td>-0.775**</td>
<td>0.498**</td>
<td>-0.046</td>
<td>-0.184</td>
</tr>
<tr>
<td>Lake volume / Catchment area</td>
<td>0.412*</td>
<td>-0.147</td>
<td>-0.058</td>
<td>0.536**</td>
</tr>
<tr>
<td>Catchment area / Lake area</td>
<td>-0.312</td>
<td>0.146</td>
<td>0.100</td>
<td>-0.764**</td>
</tr>
<tr>
<td>Lake area / Lake depth</td>
<td>0.389*</td>
<td>-0.417*</td>
<td>0.042</td>
<td>0.269</td>
</tr>
<tr>
<td>Mean NDVI</td>
<td>-0.389*</td>
<td>0.015</td>
<td>-0.146</td>
<td>-0.054</td>
</tr>
<tr>
<td>NDVI Class 1</td>
<td>0.233</td>
<td>0.135</td>
<td>0.325</td>
<td>0.407*</td>
</tr>
<tr>
<td>NDVI Class 2</td>
<td>0.215</td>
<td>0.170</td>
<td>0.070</td>
<td>-0.110</td>
</tr>
<tr>
<td>NDVI Class 3</td>
<td>-0.212</td>
<td>0.040</td>
<td>-0.123</td>
<td>0.052</td>
</tr>
<tr>
<td>NDVI Class 4</td>
<td>-0.010</td>
<td>-0.140</td>
<td>-0.052</td>
<td>0.103</td>
</tr>
<tr>
<td>NDVI Class 5</td>
<td>0.087</td>
<td>-0.311</td>
<td>-0.034</td>
<td>-0.054</td>
</tr>
<tr>
<td>NDVI Class 6</td>
<td>-0.261</td>
<td>0.280</td>
<td>-0.084</td>
<td>-0.030</td>
</tr>
<tr>
<td>NDVI Class 7</td>
<td>-0.202</td>
<td>0.258</td>
<td>-0.013</td>
<td>0.017</td>
</tr>
<tr>
<td>NDVI Class 8</td>
<td>-0.170</td>
<td>-0.195</td>
<td>0.182</td>
<td>-0.307</td>
</tr>
</tbody>
</table>

* = P < 0.05 ** = P < 0.01
(salinity) and, to some extent, also with PC2 (nitrogen). However, PC1 and PC2 are differing by having inverse correlations. I.e., positive PC1 correlations are being negative to their PC2 counterpart, and vice versa. Of the lake morphometric features only ‘Lake altitude’ and ‘Outlet within 3 m range’ are having negative correlations \((r^2 = -0.568; P<0.01\) and \(r^2 = -0.775; P<0.01\), respectively) to PC1. The reason for negative correlation to ‘Outlet within 3 m range’ being that existing outflow potential was numerically classed as 1 and absence of outflow potential classed as 2 (Table 2). Catchment morphology (‘Mean slope’ and ‘Mean aspect’) and NDVI classes (vegetation) are displaying little to no correlation with any of the PC scores. Exceptions are NDVI class 1 correlating to PC4 \((r^2 = 0.407; P<0.05)\), and PC1 having a weak negative correlation with Mean NDVI \((r^2 = -0.389; P<0.05)\). It is noteworthy that PC1 and PC2 both are showing their highest, respective, correlations to the 3m outlet range factor \((r^2 = -0.755; P<0.01\) for PC1 and \(r^2 = -0.498; P < 0.01\) for PC2). Furthermore, the runoff ratio (‘Catchment area / Lake area’) is showing high negative correlation to PC4 (colour) \((r^2 = -0.764; P<0.01)\), but no significant correlation to PC1 and PC2. PC3 (bedrock) does not display any significant correlations with the analysed parameters, thus indicating other factors affecting Ca concentrations.

### 3.3 The DOC origin

Enough material for \(\delta^2\)H analyses was only obtained in three (out of 17) lake water DOC samples following low recovery rates through the solid-phase extraction method, combined with low DOC levels in some of the lakes. One additional DOC sample (sampled in 2015) was also kindly provided by Holmgren, B. (PhD, Department of Ecology and Environmental science, Umeå university). Reference samples \((n = 5)\) had a mean of -25‰ (SD = ±4.8), and the resulting \(\delta^2\)H values for analysed DOC samples were: ss901 = -171.2‰, ss1400 = -184.0‰, ss04 = -181.7‰, ss02 = -162.0‰ (Figure 7, Table 6). In comparison, terrestrial plant and soil samples ranged between -78.4‰ to -105.4‰ \((n = 8,\) mean: -92.1‰), while algae samples ranged between -194.0‰ to -238.3‰ \((n = 4,\) mean: -210.5‰). Hence, according to the binary mixing model (Eq. 4), about 30% (mean) of the lake water DOC has an allochthonous origin.

![Figure 7](image-url)  
**Figure 7.** Box plot diagram of the non-exchangeable hydrogen calculated from the dual isotope steam equilibration. Whiskers are max./min. values, line in boxes is the median, cross is the mean and dots are values. DOC is shown as triangles and Macrophytes as filled circles. Individual sample site names are given for DOC and macrophytes.
However, macrophyte analysis showed that the binary mixing model might be a too simplistic approach, as these aquatic plants have a primarily terrestrial $\delta^2$H signal (allochthony: total mean = 68%, ss07 mean = 59%, ss1400 mean = 77%) (Table 6). Thus, the DOC could in theory be a mixing solely between macrophytes and algae. Regardless of using macrophytes or terrestrial endmembers, the applied mixing model suggest a dominance of within lake sources for the DOC.

Table 6. Sample type, originating lake, the obtained $\delta^2$H$_n$ value and the calculated rate of allochthony (in %).

<table>
<thead>
<tr>
<th>Lake</th>
<th>Sample type</th>
<th>$\delta^2$H$_n$ / ‰</th>
<th>Allochthony (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AT4</td>
<td>Terrestrial endmember</td>
<td>-78.43</td>
<td>100</td>
</tr>
<tr>
<td>AT5</td>
<td>Terrestrial endmember</td>
<td>-84.85</td>
<td>100</td>
</tr>
<tr>
<td>AT7</td>
<td>Terrestrial endmember</td>
<td>-91.38</td>
<td>100</td>
</tr>
<tr>
<td>AT8</td>
<td>Terrestrial endmember</td>
<td>-89.38</td>
<td>100</td>
</tr>
<tr>
<td>ss02</td>
<td>Terrestrial endmember</td>
<td>-86.93</td>
<td>100</td>
</tr>
<tr>
<td>ss02</td>
<td>Terrestrial endmember</td>
<td>-105.44</td>
<td>100</td>
</tr>
<tr>
<td>ss02</td>
<td>Terrestrial endmember</td>
<td>-95.89</td>
<td>100</td>
</tr>
<tr>
<td>ss02</td>
<td>Terrestrial endmember</td>
<td>-104.17</td>
<td>100</td>
</tr>
<tr>
<td>ss02</td>
<td>Aquatic endmember</td>
<td>-238.31</td>
<td>0</td>
</tr>
<tr>
<td>ss02</td>
<td>Aquatic endmember</td>
<td>-200.08</td>
<td>0</td>
</tr>
<tr>
<td>ss1590</td>
<td>Aquatic endmember</td>
<td>-209.44</td>
<td>0</td>
</tr>
<tr>
<td>ss903</td>
<td>Aquatic endmember</td>
<td>-193.99</td>
<td>0</td>
</tr>
<tr>
<td>ss02</td>
<td>DOC</td>
<td>-162.04</td>
<td>41</td>
</tr>
<tr>
<td>ss04</td>
<td>DOC</td>
<td>-181.73</td>
<td>24</td>
</tr>
<tr>
<td>ss1400</td>
<td>DOC</td>
<td>-184.01</td>
<td>22</td>
</tr>
<tr>
<td>ss901</td>
<td>DOC</td>
<td>-187.99</td>
<td>33</td>
</tr>
<tr>
<td>ss1400</td>
<td>Macrophyte</td>
<td>-106.34</td>
<td>88</td>
</tr>
<tr>
<td>ss1400</td>
<td>Macrophyte</td>
<td>-131.15</td>
<td>67</td>
</tr>
<tr>
<td>ss07</td>
<td>Macrophyte</td>
<td>-128.37</td>
<td>69</td>
</tr>
<tr>
<td>ss07</td>
<td>Macrophyte</td>
<td>-151.95</td>
<td>49</td>
</tr>
</tbody>
</table>

All analysed lake sediment samples ($n = 78$) displayed improbable $\delta^2$H ratios. I.e., the samples had $\delta^2$H ratios lower than the algae endmembers. Therefore, sediment samples, as well as the references samples analysed together with them, were disregarded from all further analysis of relevance for hypothesis 2.
4 Discussion

4.1 Catchment properties and lake water chemistry

4.1.1 The drivers of lake water chemistry

In line with hypothesis 1, my findings concerning water geochemistry and catchment characteristic couplings, support the existing paradigm on lake water chemistry of the Søndre Strømfjord region; that is, the evaporation effect is a major driver of lake water chemistry within this region and catchment features enhancing the evaporation effect are highly influential on the lake water chemistry.

In boreal ecosystems, there is a strong connection between soils and freshwater, where catchment characteristics such as vegetation type (Ågren et al. 2008; Laudon & Sponseller 2018), topography (Blackburn et al. 2017) and landscape hydrology (Klaminder, J. et al. 2011; Laudon & Sponseller 2018), exert strong influence on water chemistry. However, my studied lakes from the Søndre Strømfjord region lack such strong connectivity to surrounding soils. One apparent indicator for the disconnect between lake water chemistry and general catchment characteristics in the KV area, is the lack of significant correlation between factors reflecting topographic proxies (‘Mean slope’ and ‘Mean aspect’) to any of the PC scores. Correlations to such topographic features would be expected if there was a strong control on water chemistry from groundwater, as seen in boreal settings (e.g. Klaminder et al., 2011; Blackburn et al., 2017; Laudon & Sponseller, 2017).

Clearly, overall catchment topography and vegetation in my studied catchments have a weak influence on the lake water chemistry, leaving other factors with a higher order of control over these aquatic systems. Indeed, Spearman’s rho showed that primarily, factors related to morphometry (e.g., ‘Lake area’, ‘Lake depth’) and evaporation rates (‘Lake altitude difference’) of the lake, together with proxies for poor drainage and, thus, of long lake water turnover times (i.e. ‘Outlet within 3m range’) was of significance for lake water chemistry. These findings combined indicate that the main reasons for present day water chemistry are indeed high evaporation rates (stimulated by large lake surfaces) and a slow turnover of water, leading to an endorheic lake regime. Thus, my interpretation agrees with those of previous studies and observation from the area (e.g. Anderson & Stedmon, 2007; Law et al. 2015; Law et al. 2018). Furthermore, by examining lake ontogeny and geochemistry along the precipitation gradient from the coast to the ice sheet, e.g. Law et al. (2015) and Osburn et al. (2018), showed that the KV area displayed a high second order control on lakes through e.g. catchment morphology. Whereas the primary control of climate (through P:E balance) was the main driver along the gradient from Sisimiut to the ice sheet. This condition can be further coupled to the global scale hierarchical lake DOC regulation proposed by Sobek et al. (2007), where climate and topography are acting on different levels in influencing lake DOC.

My main vegetation proxy (‘Mean NDVI’) showed a weak and negative correlation to lake water chemistry captured by PC1 (Table 5). As vegetation is highly controlled by precipitation in the region (Thing 1984; Anderson et al. 2017), this correlation could serve as further evidence of control through the evaporation effect on these lakes. Where a higher ‘Mean NDVI’ expresses higher abundance and quality of vegetation in the catchment and, as such, is being indicative of a weaker evaporation influence. However, there might be an additional way of interpreting this correlation between ‘Mean NDVI’ and PC1. If PC1 was instead regarded as a proxy for alkalinity and pH, this correlation could instead be considered as lower ‘Mean NDVI’ (indicating poor vegetation cover) resulting in higher alkalinity and pH through increased input of terrestrial sediment as hypothesised by e.g. Anderson et al. (2001, 2017) and Rydberg et al. (2016). Conditions with a sparse or completely missing vegetation cover is making exposed soil available for transport in to the lakes, either through direct short range eolian
input, or through surface runoff. However, if this relation between vegetation cover and water chemistry is the main reason for the correlation between ‘Mean NDVI’ and PC1, I would also expect significant correlations between PC1 and the lower NDVI classes, which is not the case. This could be a consequence of that NDVI data processing was not made in a satisfactory way, or that there are additional factors in play which are not accounted for by simple regression alone. Nevertheless, the mechanism behind the negative correlation between ‘Mean NDVI’ and PC1 remains elusive and warrants future research before it can be fully revealed.

As mentioned by e.g. Sobek et al. (2007) and Buffam et al. (2007), surface waters in more humid regions generally experience a dilution of DOC and geochemistry during increased runoff and throughflow events such as intensive rain or snow melt. However, this paradigm is mainly derived from regions with higher input of meteoric water and hydrologic connectivity in and between catchments, than what is found in the eastern Søndre Strømfjord region. The general lack of hydrologic connectivity in, and between, catchments and lakes here could be expected to influence lake water chemistry by leading to continued ‘up-concentration’ of solutes in the lake water, as endorheic lake conditions are effectively acting as a sink for solutes. With no significant correlation found between runoff rate (‘Catchment area / Lake area’) and salinity (PC1), I interpret this as further evidence of the hydrologic disconnect between lakes and their catchments leading to an enhanced evaporation effect. In this regard, it is noteworthy that PC4, proxy for Abs. 250 nm (Table 4), is showing a strong negative correlation to ‘Catchment area / Lake area’. Indeed, this could be due to more pronounced photo-bleaching causing a transformation of DOC into colourless forms as suggested previously by Anderson & Stedmon (2007).

Following a period of high autumn precipitation, Holmgren (2016) noticed a terrestrial seepage input of DOC rich, and coloured, water to lake ss02. Hence, some exchange between the terrestrial carbon pool and the lake water colour can be expected. Indeed, a similar phenomenon has been observed in high arctic lakes of Alaska by Lewis et al. (2011), where DOC and solute enriched soil water entered lakes following late-season rain events capable of flushing out dissolved material from the AL. Similar pulses of groundwater input was also observed in the Søndre Strømfjord region by Osburn et al. (2018). This type of solute enriched groundwater was shown by Lewis et al. (2011) to be highly influenced by small variations in catchment microtopography, even leading to noticeable differences between adjacent catchments. With these indications suggesting locally variable and periodic inputs of solute-enriched and coloured water from surrounding soils, it is tempting to interpret the positive correlation between one of my vegetation classes (‘NDVI class 1’) and colour (PC4) (Table 5) as a result of this mechanisms. However, ‘NDVI class 1’ is indicative of bare rock and (primarily) surface waters, and as such, it contains artefact measures of the lake surfaces where my remote sensing method failed to fully remove lake areas. The correlation between lake colour (PC4) and ‘NDVI class 1’ is therefore not surprising and is only confirming existing knowledge, hence this correlation can further be disregarded.

The PC3 axis, driven mainly by the variability of calcium, was indeed the water chemistry variable least predictable by my proxies. I find that variation captured by PC3 is most likely driven by within lake carbonate formation and subsequent precipitation into the sediment and, thus, unlikely to be correlated to catchment properties. This interpretation is supported by previous studies showing that Ca, derived from bedrock through weathering, is indeed undergoing such processes in the region (Anderson et al. 2001; Pla & Anderson 2005; Scribner et al. 2015).
4.1.2 Temporal water chemistry variations and possible consequences

Lake water chemistry in my analysis was obtained in 2001 while satellite imagery in the GIS analysis was based on data from 2017; hence, this time-gap could generate uncertainties if the water chemistry of the studied lakes, or vegetation cover within their catchments have changed dramatically in between 2001 and 2017. Temperature and precipitation has increased in the region during the last few decades (Mernild et al. 2014, 2015), probably driving observed lake level shifts (Law et al. 2018). Furthermore, there has been recorded increases in e.g. atmospheric nitrogen deposition (Anderson et al. 2017) as well as in lake water sulphate concentrations (Saros et al. 2015). These changes co-occurred with the previously discussed decrease in DOC (Saros et al. 2015).

So, is it likely that my result was affected by the time-gap between satellite images and water sampling? I find that unlikely since trends within the water chemistry sets of 2001 and from the 2017 sampling are in accordance with each other and contain no major deviances. In short, DOC, DN, a*375 and SUVA254 followed the same patterns as previously seen for these lakes, with variance between the IM and KV areas being consistent and in line with previously recorded patterns (Figure 5) (e.g. Anderson et al., 2001; Osburn et al., 2018). Additionally, the main implication this temporal discrepancy might have for this study, concerns correlations between the PC scores and NDVI values, and possibly to lake morphometry. NDVI, even though a widely used measure, is a relatively rough proxy with inherent risks of errors coupled to image resolution as well as inter- and intra-annual variations of, e.g., precipitation events and temporal data availability (Paruelo & Laurenroth 1998; Kerr & Ostrovsky 2003). Thus, utilizing NDVI in the context of this study it is necessary to acknowledge the risk of error and that this could be a partial reason for the confounding NDVI to PC scores regressions. Even if it is used here as a rough measure on catchment vegetation influence on lake water chemistry, where trends of both are expected to act on larger time scales. However, at this spatial scale, and only using imagery acquired on a one-time occasion, the method can be considered precise enough for identifying broader trends and conditions. A similar view should be applied concerning lake morphometry. In the light of recorded lake level changes (Law et al. 2018), the question of analysis error becomes less relevant considering the methodological precision in manual lake delineation based on satellite imagery and DEM data of varying acquiring dates. Again, while accuracy on each lake might not be enough for exact local predictions, the precision within the entire dataset is sufficient for the broader trends evaluated here.

4.2 The DOC origin

My H-isotope analyses indicate that lake water DOC is of a primarily autochthonous origin. As shown by the mean allochthony rate for the sampled lake water, only 30% of the DOC could be traced to terrestrial sources, attributing the major DOC input for these lakes to in-lake sources, such as pelagic algae. Hence, my second hypothesis (that lake water, and hence sediment, DOC have an autochthonous origin) appears valid, given that mixed model calculations suggest that at least >60% of lake water DOC has an autochthonous origin.

Of the four analysed samples lake ss02 displayed the strongest rate of allochthony (41%), with the IM lake ss901 following at 33% (Table 6). As described by Rydberg et al. (2016), the lakes closest to the ice margin are especially susceptible to eolian sediment input due to the strong katabatic winds descending from the ice sheet. As increased DOC concentrations have been found in older seasonal snow, enriched in eolian deposits, it is likely that both lake water chemistry and DOC is under high allochthonous influence in the IM area (Rydberg et al. 2016; Anderson et al. 2017). It is presumably even higher than in the KV area, due to the increased eolian influence closer to the ice sheet. However, upon simple comparison to the KV area lakes, this high terrestrial influence is not clearly reflected in e.g. total concentrations of the IM lakes.
The explanation for this might be dilution as an effect of increased precipitation received at IM, compared to KV, leading to increased throughflow with reoccurring drainage events (Johansson et al. 2015). The high percentage of allochthony displayed by DOC from lake ss02 (Table 6) is largely in line with the findings of Holmgren (2016), where sediment DOC was shown to be of a predominantly allochthonous origin. However, my findings inevitably question the applicability of those findings to the broader scale in the KV area. Furthermore, Ss02 is the only lake of the KV area with a high rate of anthropogenic influence in its catchment. It is the host to remains of a cold-war era radar station (main building still present), an active dirt road, as well as the starting/ending point of a hiking trail between Sisimiut and Kangerlussuaq which is frequented by all-terrain vehicles. Also, the trail is following along on top of one of the zones determined to be in NDVI class 7, i.e. an intermittent flow path in to the lake. With these circumstances in mind, the risk of mobilisation and terrestrial influx through anthropogenic disturbances must be taken in to consideration, especially as the same rate of allochthony is not observed in the other examined KV lakes.

Macrophytes displayed a remarkably strong terrestrial signal (Figure 6), which if included in the binary mixing model (i.e. macrophytes and algae) as the main aquatic source, suggest that these plants could contribute with about 68% of the organic matter in the DOC. The spread between the different species samples, as well as between the different lakes was, furthermore, relatively large, with samples from lake ss1400 having more of a terrestrial signal (77% mean allochthony) than lake ss07 (59% mean allochthony) (Figure 7, Table 6). Consequently, the question of suitability in utilizing δ²H concentrations without accounting for macrophyte influence, when inferring allochthony rates in DOC from lake water or sediments, arises. The high allochthonous isotopic signal in macrophytes indicate that they would potentially be an additional aquatic source of terrestrialiy coded stable H-isotopes. With respect to the abundant macrophyte community of ss1400 it is remarkable that out of the four analysed lake water DOC samples, ss1400 was showing the highest rate of autochthony within aquatic DOC. With the planktonic signal for ss1400 being stronger than even the oligosaline ss04 (second highest rate of allochthony). Ss1400 is one of the two sampled lakes in the KV area with highest absorbances, and it is likely that one of the reasons for the high absorbance is a higher rate of pelagic algae such as the Golden brown algae (Jensen & Christensen 2003; Nielsen 2010). Thus, one possible explanation for the high allochthony rate in the pelagic DOC, is that high pelagic algae production (algae signal) is impeding the influence of the macrophytes (terrestrial signal) in the overall δ²H ratio.

I found the δ²H ratio of the n = 78 sediment samples to be unsuitable for further analysis of DOC origin, given the occurrence of samples with a δ²H ratio outside even that of the two endmembers used in my mixing model. I interpreted this as a consequence of that the analysed sediment H-fraction was having a high rate of residual H_{ex} originating from clay minerals, resulting in an overshadowing of the organic matter signal (Schimmelmann 1991; Wassenaar & Hobson 2000; Ruppenthal et al. 2013). Thus, I could not treat these results as solely representing the organic matter δ²H isotopic composition. Importantly, this condition of residual H_{ex} from clay minerals in the analysed matrix did not influence analysis of remaining DOC samples. While the drying process might not be sufficient enough to remove all H_{ex} in clay-minerals of sediments, it should be for the remaining samples (Ruppenthal et al. 2013); hence, the δ²H analyses of the DOC samples should be trusted.

4.3 Conclusion
My study strengthens the existing paradigm that the P:E balance, governed by local climate is the main driver of lake water chemistry. As such, morphologically speaking, only factors directly affecting the climatic influence on residence time and P:E balance are of importance
to lake water chemistry in the region. Predictions for future climate around Søndre Strømfjord is pointing towards warmer and slightly wetter conditions (Christensen et al. 2016), following the global warming scenarios outlined by IPCC (IPCC 2014). So far, an accelerated increase in mean winter temperature and a deepening of the precipitation gradient from the coast to the Ice sheet, following evaporation still superseding annual precipitation has been recorded during the last few decennia’s in the region (Mernild et al. 2014, 2015). Furthermore, between 2001 and 2012, there has been significant increases in intensive precipitation events (Mernild et al. 2015). Hence, it should be recognized that possible inter- and intra-annual changes of the P:E balance of this region in the future, has the capability to quickly alter the current conditions governing lake water chemistry. Such changes have the potential of highly increasing the importance of catchment morphology and vegetation to lake water chemistry.
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