Carbon translocation from glacial and terrestrial to aqueous systems – characteristics and processing of dissolved organic matter in the endorheic Tibetan Lake Nam Co watershed

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A sense of solemn aspiration comes upon us as we view the mountain. We are uplifted. The entire scale of being is raised. Our outlook on life seems all at once to have been heightened. And not only is there this sense of elevation: we seem purified also. Meanness, pettiness, paltriness seem to shrink away abashed at the sight of that radiant purity.
— *Sir* Francis E. Younghusband

Abstract

The Tibetan Plateau (TP) comprises sensitive alpine environments such as grassland biomes. Climatic changes and intensifying land use threaten these ecosystems. Therefore, it is important to understand the response of ecosystems to changing biotic and abiotic factors. The translocation of dissolved organic matter from glacial and terrestrial to aqueous systems is an important aspect of this response, specifically when characterizing changing conditions of freshwater resources and sensitive limnic ecosystems on the TP. Via changes in its chemical composition, characteristics, transformation and processing of DOM can be tracked. Three catchments of the Nam Co watershed of the TP (Niyaqu, Qugaqie and Zhagu) and the lake were investigated to understand how site specific terrestrial processes and seasonality affect the composition of DOM and alteration of organic compounds in streams and the lake of this endorheic basin.

Four hypotheses were tested: **H1** The natural diversity in the Nam Co watershed controls site specific effects on DOM composition. **H2** Seasonal effects on DOM composition are driven by warm and moist summers influenced from the Indian summer monsoon (ISM) and cold and dry winters. **H3/ H4a** Site specific effects on DOM diminish by means of biological decomposition and photooxidation of DOM during the stream path / in the lake. Alongside **H4b** organic matter of the Nam Co Lake is independent from catchment influences, given by an autochthonous source of DOM.

A multi-parameter approach was applied, consitsing of water chemistry parameters (pH, electric conductivity, cations and anions, dissolved inorganic carbon), concentration of dissolved organic carbon (DOC), DOM characteristics (chromophoric DOM, fluorescence DOM and $\delta^{13}C$ of DOM) and DOM ultra-high resolution Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS). Sampling was conducted for three seasons, freshet in 2018, the phase of the ISM in 2019 and post-ISM baseflow in 2019. Alongside a watershed-wide plant cover estimate was composed, to explore the link between differences in DOM characteristics and degree of green plant cover. Sampling covers stream water, as well as endmember samples such as: glacial effluents, water of springs and water from an alpine wetland. The lake was covered by sampling the brackish zone and the lake pelagial and the lake surface.

The composition of DOM differed between the three endmember groups and between stream samples of catchments. Glaciers showed a dual DOM source, indicating a glacial microbiome and compounds derived from burned fossil fuels. Springs differed based on their geographic location. Upland waters showed limited inputs of alpine pastures: lowland springs displayed influences of yak faeces with microbial reworked DOM, indicated by less negative $\delta^{13}C$ and nitrogen. Wetlands were distinguished by more eutrophic conditions by highest concentrations in DOC and high amounts in N-heteroatoms. Streams were site specific with input sources derived from glaciers, wetlands, groundwater, intense animal husbandry and a plant-derived phenolic signature from alpine pastures aligned to the degree of plant cover. Seasonality affected DOM characteristics in stream water. During freshet, DOM was plant-derived, as was during baseflow conditions. A flush of dissolved organic carbon, accompanied by a compositional shift towards more microbial derived DOM was observed during the ISM season.

Processing of DOM in streams was limited to the biolabile fraction of DOM of the glacial biome. Transformation of DOM was overruled by the constant input of plant derived phenolic DOM compounds from alpine pastures. Consequentially, the brackish intermixing zone showed the inflow of terrestrial DOM into the lake. In contrast, lake water exhibited distinct DOM characteristics, by lowest amounts in aromatic molecular compounds and DOM rich in ^{13}C. This suggested intense processing of phenolic, terrestrial derived DOM by photooxidation, as well as a seasonally stable autochthonous DOM source derived from algae and microorganisms in lake water. In conclusion, DOM characteristics are largely influenced by local endmembers such as glaciers, springs and wetlands. Seasonality shows that shifts in the onset, and changes in the intensity of the ISM can largely modify DOM composition. Processing of DOM took place mainly in the lake. The study revealed that DOM is suited to function as a monitoring agent in this lake watershed. Hence, DOM is a helpful tool to understand changes in ecosystems, and forthcoming, to safeguard sensitive ecosystems of the TP.

Abstract

Keywords: Dissolved organic carbon, Third Pole, High Asia, Water Tower of Asia, Critical zone, Non-metric multidimensional scaling, PARAFAC fluorescence, Fourier transform ion cyclotron resonance mass spectrometry

Zusammenfassung

Das Tibet-Plateau (TP) umfasst empfindliche, alpine Ökosysteme von globaler Bedeutung, wie das größte alpine Weidebiom der Welt. Die Ökosysteme Hochasiens sind durch Auswirkungen des Klimawandels und die Intensivierung der Landnutzung stark bedroht. Um eine Einschätzung des Zustands von Süßwasserressourcen und empfindlichen See-Ökosystemen in Tibet treffen zu können, untersucht die Arbeit die Verlagerung von organischem Kohlenstoff aus glazialen und terrestrischen in aquatische Systeme. Dazu wird die gelöste organische Substanz (DOM) genutzt. Drei Einzugsgebiete des tibetischen Nam Co Sees: Niyaqu, Qugaqie und Zhagu, sowie der See selbst wurden untersucht.

Es wurden vier Hypothesen formuliert: **1**) Standortspezifische Auswirkungen auf die Zusammensetzung des gelösten organischen Kohlenstoffs werden vermutet. **2**) Saisonale Auswirkungen auf die Zusammensetzung von DOM werden angenommen. Weiterhin: **3**) wird der Verlust standortspezifischer Signaturen im gelösten organischen Kohlenstoff durch Verarbeitung während der Fließwege der Flüsse und **4a**) im See durch biologischen Abbau und Photooxidation angenommen. Schließlich **4b**) gelöster organischer Kohlenstoff im Nam Co See wird als eigenständig von Einflüssen der Einzugsgebiete und der Saisonalität angenommen, DOM ist überwiegend mikrobiell geprägt.

Ein DOM Multiparamater-Datensatz wurde verwendet. Hierzu wurden wasserchemische Parameter (elektrische Leitfähigkeit, pH-Wert, Konzentration von Kationen und Anionen, gelöster anorganischer Kohlenstoff), die Konzentration des gelösten organischen Kohlenstoffs, sowie Parameter zur weiteren Charakterisierung erhoben (kolorierte organischer Substanz, fluoreszierendes DOM und Isotopie des ^{13}C Kohlenstoffatoms). Zusätzlich wurde ultrahochauflösende Massenspektrometrie (FT ICR-MS) genutzt, um die molekulare Zusammensetzung zu untersuchen. Die Probenahme erfolgte an drei verschiedenen Zeitpunkten im Jahr, zur Schneeschmelze 2019, in der Regenzeit (2018) sowie während des Basisabfluss (2019). Flusswasser der Einzugsgebiete, sowie Gletscherabflüsse, Quellwasser und Wasser eines alpinen Feuchtgebietes wurden beprobt. Im Seegebiet wurde die Brackwasserzone und das Pelagial in 30 m Tiefe, sowie Oberflächenwasser beprobt.

Die Arbeit zeigt, dass sich die molekulare Zusammensetzung von DOM sowohl zwischen Gletschern, Quellen und Feuchtgebieten als auch zwischen den untersuchten Einzugsgebieten stark unterscheidet. Gletscher wiesen eine duale Zusammensetzung des DOM, aus mikrobiellen und kondensiert-terrestrischen Quellen auf. Quellwasser weist Unterschiede hinsichtlich der Lage in der Landschaft auf. Hochlandquellen zeigten geringe Einträge, die aus dem umliegenden Weidegebiet stammten. Tieflandquellen waren stark durch Tierhaltung in Form von Stickstoffeinträgen und angereichertem ^{13}C beeinflusst. Alpine Feuchtgebiete wiesen hohe Kohlenstoffkonzentrationen und eine hohe Zahl von Stickstoffverbindungen auf. Insgesamt ist das Wasser der Bäche durch hohe Standortspezifität geprägt. Die Saisonalität hatte einen großen Einfluss auf die Eigenschaften des DOM im Flusswasser. Zur Zeit der Schneeschmelze und der Phase des Basisabflusses überwogen pflanzliche Quellen. Wohingegen während des Monsuns ein erhöhter Austrag von organischem Kohlenstoff beobachtet wurde, welcher mit einer Verschiebung der Zusammensetzung hin zu mehr mikrobiell beeinflusstem DOM einherging.

Die Verarbeitung von DOM im Fluss beschränkte sich auf labile Verbindungen aus dem Gletscherbiom. Die Umwandlungsprozesse in Flüssen wurden durch den ständigen Eintrag phenolischer Verbindungen überdeckt. Folglich zeigte sich der Eintrag von terrestrischen Signaturen in der Brackwasserzone des Sees. Das Seewasser im Pelagial hingegen war stark abgereichert an Aromaten und zusätzlich stark angereichert in ^{13}C. Dies lässt auf eine intensive Verarbeitung der eingetragenen terrestrischen Signaturen schließen. Zusätzlich weist der See eine DOM Zusammensetzung auf, die aus mikrobieller Produktion im See selbst herrührt und von saisonalen Veränderungen und Signaturen der Einzugsgebiete unabhängig ist.

Die Studie zeigt, dass DOM Eigenschaften in hohem Maße von lokalen Einflüssen wie Gletschern, Quellen und Feuchtgebieten sowie von weiteren standortspezifischen Auswirkungen abhängig sind. Die Degradation von Feuchtgebieten kann eine Gefahr für empfindliche, oligotrophe Ökosysteme darstellen. Labile Verbindungen aus Gletschern hingegen, werden bereits während des Flusslaufs verwertet. Die Verarbeitung der gelösten organischen Substanz fand hauptsächlich im See statt. Abschließend lässt sich festhalten, dass sich gelöste organische Substanz eignet, um raum-zeitliche Effekte zu untersuchen. Somit ermöglicht sie ein Monitoring, zum Schutz der empfindlichen Ökosysteme Tibets.

Table of content

List of tables

Study I

Study II

Study III

List of figures

General introduction

Study I

Study II

Study III

Synthesis

Abbreviations

A254	UV/VIS absorbance at 254nm
Ai_{mod}	Modified aromaticity index
α	Significance level
AROP	Aromatic O-poor
AROR	Aromatic O-rich
AWM	Asian Winter Monsoon
Cal ka BP	Calibrated kiloyears before present
CDOM	Chromophoric DOM
DBE	Double bond equivalents
DDC	DOM degradation cascade
DIC	Dissolved inorganic carbon
DOC	Dissolved organic carbon
DOM	Dissolved organic matter
EASM	East Asian Summer Monsoon
EC	Electric conductivity
EEM’s	Excitation emission matrices
ESI	Electrospray ionization
FC	Fluorescence component
FDOM	Fluorescence DOM; fluorophore DOM
FT-ICR MS	Fourier transform ion cyclotron resonance mass spectrometry
HCl	Hydrochloric acid
HDPE	High-density polyethylene
HOT	Hawaii DOM ocean time-series
HUSOP	Highly unsaturated O-poor
HUSOR	Highly unsaturated O-rich
K. pygmaea	*Kobresia pygmaea* (alpine sedge)
m. asl.	Meters above sea level
ISM	Indian summer monsoon
LIA	Little ice age
MWP	Medieval warm period
n	Number of observations; sample size
NAMORS	Nam Co Monitoring and Research Station for

	Multisphere Interactions
NMDS	Non-metric multidimensional scaling
N_2O	Nitrous oxide
PCA	Principal component analysis
PCE	Plant cover estimate
PES	polyethersulfone
PPL	Styrene divinyl benzene polymer
R^2	Coefficient of determination
RCC	River continuum concept
RMSE	Root mean square error
RI	Relative intensity
SAT	Saturated
SD	Standard deviation
SMTZ	Summer Monsoon Transition Zone
SOC	Soil organic carbon
SOP	Saturated O-poor
SOR	Saturated O-rich
SPE	Solid phase extraction
$SUVA_{254}$	DOC normalised UV/VIS absorbance at 254nm
SVE	Support vector machine regression
TDC	Total dissolved carbon
TP	Tibetan Plateau
TransTiP	International Research Training Group “Geoecosystems in transition on the Tibetan Plateau”
USN	Unsaturated with nitrogen
USOP	Unsaturated O-poor
USOR	Unsaturated O-rich
UV/VIS	Ultraviolet /visual light source
WRB	World Reference Base for Soil Resources
ypb	Calibrated years before present

General introduction

Organic carbon, the dissolved organic carbon fraction and the critical zone

Carbon is the crucial atom for all processes of life. There would be no living cells, no fungi, no plants and no animals without carbon and especially forms of organic carbon. Although relatively rare in the total elemental share of planet earth, all organic compounds are built up with carbon (Neubauer, 2014). This element is truly essential when investigating any processes in nature. Organic carbon comprises up to several tens of millions of different organic compounds, and plays a central role such as in lignin and cellulose, both in the supporting tissue of vascular plants, as energy source in saccharides and as component in nucleic acids constituting DNA. Summing up, organic carbon is relevant in all processes of life. The carbon cycle elucidates the linkage and continuous exchanges between the biosphere, geosphere, pedosphere, hydrosphere and atmosphere, ultimately sustaining life on earth (Figure 1).

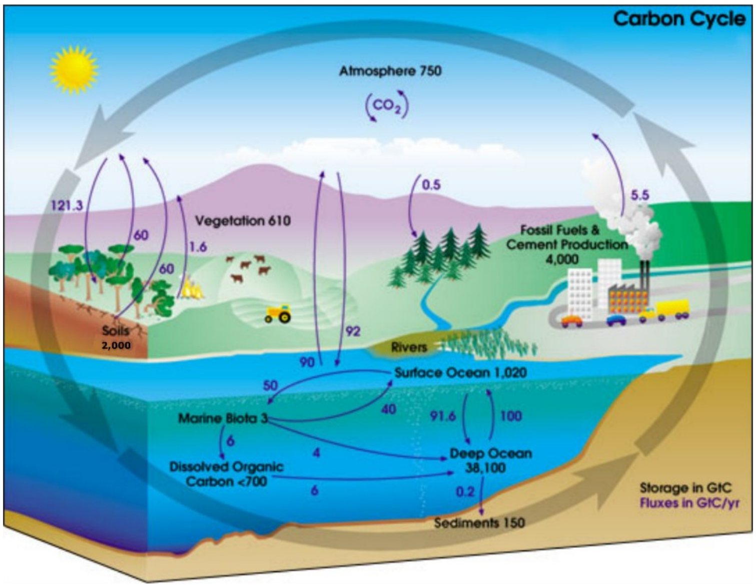

Figure 1: The carbon cycle of the earth including storages (black) and fluxes (magenta). Copyright: US-Government/NASA

Due to its significance, organic carbon is immensely investigated across a wide variety of ecosystems, e.g. for soils (Jackson et al., 2017), glacial ecosystems (Anesio et al., 2009), inland waters (Spencer et al., 2012; Worrall et al., 2004) and oceans (Dittmar and Kattner, 2003). The concept of the critical zone elucidates the exchange of compounds and the interrelatedness of spheres and ecosystems in the near surface environment (Giardino and Houser, 2015). The critical zone puts a particular focus on the hotspot of interaction, located from the top of the tree canopy down to the aquifer bottom. Resulting from this interrelatedness, investigations of natural processes need to expand across spheres and scales, involving many disciplines (Brantley et al., 2007). Therefore, it requires intersectional research, to understand, how processes transform and alter organic carbon in the critical zone (Lin, 2010) in order to understand, how threatened ecosystem services can be safeguarded worldwide (Green et al., 2019; Naidoo et al., 2008).

In order to grasp the multitude of organic compounds that carbon constitutes, size thresholds for organic matter have been introduced, leading to the establishment of organic matter fractions as operational units (Figure 2) (Wander, 2004). Different fractions of natural organic matter hereby are scale variant, meaning natural organic matter fractions with different sizes have diverging properties and also react differently (Aufdenkampe et al., 2001; Chen et al., 2002; Chen et al., 2003). In general, dissolved organic carbon (DOC) can be differentiated from particulate organic carbon (POC) by the operational threshold of 0.45µm in diameter (Raymond and Bauer, 2001). The fraction of particulate organic matter of POC (POM) is further subdivided by density fractionation (Cerli et al., 2012) and thus allows a refined understanding of its reactivity and fate in soils (Schrumpf et al., 2013) and aquatic environments (Chifflard et al., 2019). Besides from particulate fractions, dissolved organic compounds play a key role for nutrient budgets of ecosystems. Dissolved organic matter (DOM) constitutes the largest mobile pool of carbon and nutrients globally and thereby is the most important energy source for many ecosystems worldwide (Kaplan and Cory, 2016). Much of ecosystem natural organic matter losses or gains occur via the DOM fraction, especially if connections are via the lotic environment, such as rivers. DOM is understood as the agent “browning the waters” (Roulet and Moore, 2006), highlighting its importance, but also pointing out its potential to threaten the status quo of connected ecosystems, for example by inducing oxygen depletion. DOM is understood to inherit the chemical signatures of its source area, often soil- and plant-borne (Shogren et al., 2019) or from glacial biomes (Anesio et al., 2009) further, DOM was investigated to have significant effects on receiving ecosystems, as trigger for acidification (Han et al., 2022) or eutrophication (Görs et al., 2007) and by this, effects ecosystem services (Williamson et al., 2016) including water quality (Baker and Inverarity, 2004). DOM characteristics allow to trace changes in the thermal regime of soils, such as warming permafrost (Kawahigashi et al., 2004) or degrading alpine wetlands (Zhang et al., 2020) and influences of agricultural practise (Roebuck et al., 2020), making it an important precursor of effects of climate and land-use change. DOM can connect changing environments in the source area with effects in receiving ecosystems, over distances of several hundreds of kilometres (Seidel et al., 2015). DOM facilitates to trace the connection between ecosystems over temporal (Goodman et al., 2011) and spatial scales (Roebuck et al., 2018) and, given its enormously wide range of chemical composition and its complexity, is an unrivalled promising object of research. DOM hence offers unique opportunities to monitor ecosystems (Jaffé et al., 2012), due to its small size, ubiquitous occurrence and recurrent chemical composition (Wünsch et al., 2019), it can easily be employed as an early warning signal and monitor for processes in connected catchments. The technical (Tian et al., 2020) and computational (Moradi et al., 2020) opportunities to expand this to real-time investigations with big data are currently developed and promise an even better understanding of DOM characteristics as influenced by a changing environment. The current development of standardized DOM fingerprints (Aukes and Schiff, 2021) will also help to make DOM monitoring approaches more applied.

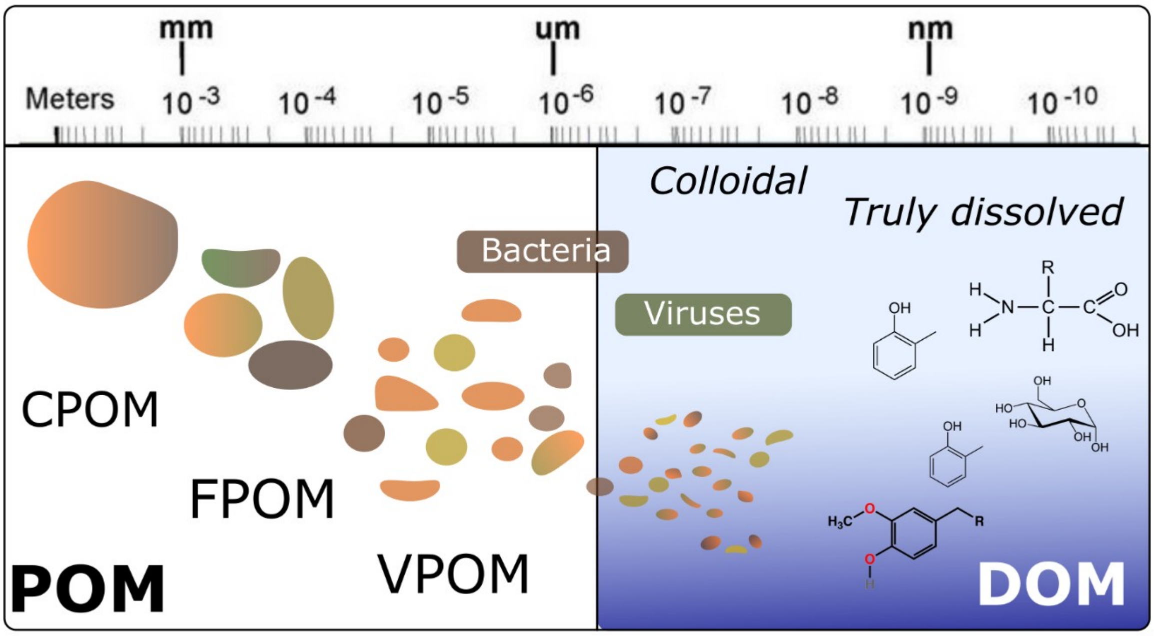

Figure 2: Size thresholds of different organic matter fractions: Particulate organic matter (POM) can be subdivided in coarse particulate organic matter (CPOM), fine particulate organic matter (FPOM) and very fine particulate organic matter (VPOM), colloids and compounds in dissolution below the 0.45µm threshold are defined as dissolved organic matter (DOM).

DOM processing and transformation in the aquatic pathway

When assessing DOM characteristics and the implementation of DOM for long-term monitoring, it has to be understood that DOM is not just translocated in aquatic environments such as rivers or fluvial lakes from the source to terminal environments. The fluvial pathway functions as a DOM reactor not just as a transport module (Riedel et al., 2016). Processing and transformation of DOM can take place by sorptive fractionation between DOM and POM in streams, removing biolabile compounds from DOM (Aufdenkampe et al., 2001) or by photooxidation, especially of DOM rich in aromatic compounds (Hansen et al., 2016; Helms et al., 2014; Spencer et al., 2009) leading to a fundamental transformation of DOM by rendering it less refractory (Miranda et al., 2020). Biological utilisation or bio-degradation of DOM on the contrary, removes lower molecular weight compounds, such as carbohydrates from DOM and leaves aromatic structures, such as lignin behind (Kalbitz et al., 2003; Ni and Li, 2019). Biological and photolytical degradation can change DOM characteristics in opposite directions (Hansen et al., 2016), allowing for a selective interpretation of DOM processing.

The DOM degradation cascade (Zark and Dittmar, 2018) and the river continuum concept (Mosher et al., 2015; Vannote et al., 1980) are two heuristic conceptual frameworks that allow for an understanding of DOM characteristics (this especially by the river continuum concept) and subsequently DOM processing in aquatic environments. The frameworks follow the concept of changing river morphology during the path of flow (Rinaldi et al., 2016) by underlining that high site-specificity exist at low Strahler numbers (Mosher et al., 2015). Strahler number, is the numerical expression of branching complexity, allowing to decipher the maturity of streams. Lowest numbers represent the 'finger-tip' channels (Strahler, 1952). In the headwaters of catchments, DOM signatures are closely connected to catchment specific attributes. With increasing Strahler number, site-specificity decreases (Mosher et al., 2015) as transformation and unification processes, such as bio- and photodegradation gain in relevance. Notably, human-induced influences can hamper this processual development (Roebuck et al., 2020), especially in smaller catchments. Nevertheless, transformation and unification of DOM are globally in force, as was shown for large rivers (Riedel et al., 2016), estuaries and the open ocean (Zark and Dittmar, 2018). Identifying recurring patterns of DOM processing could considerably help in estimating the consequences of global challenges, such as land-use intensification, climate change and eutrophication (Raús Maúre et al., 2021).

Multi-methodological approaches towards a characterisation of DOM

DOM is a highly complex manifold of chemical substances, making a complete understanding of DOM characteristics a potentially challenging and laborious, if not an impossible task (Stubbins et al., 2014). In order to enable drawing conclusions and policy decision-making based on DOM analytical essays, clear scientific coherence has to be established. In the light of this, studies which focus on the critical zone are dealing with the complex interplay of different ecosystems, biomes or landscape units. Both, the properties of DOM and the conceptual dimension of the critical zone call for scientific approaches that utilise and combine proxies and techniques (Kayler et al., 2019), in multi-methodological or multi-parameter approaches (Oliveira et al., 2018).

A major challenge remains for the coupling of different methodological approaches to unify scientific concepts for studies in the field of the critical zone or spanning across different ecosystems (Kellerman et al., 2018; Raymond and Bauer, 2001). This challenge can only be overcome, when contradictions are removed and findings are matched, such as the influence of site specific biomolecules in DOM on global persistent terminal DOM signatures. The analytical toolbox for the analysis of natural organic matters has widely expanded in latest years. This is the case for fluorescence spectroscopy (Fellman et al., 2010) with the establishment of the PARAFAC algorithm (Bro, 1997) and for cyclotron mass spectrometry (Leyva et al., 2020) with enhanced routines for analysis (Merder et al., 2020) and assessment (Bramer et al., 2020). Alongside with technical novelties, the development of coupled approaches allows an even better understanding of the molecular composition and reactivity of DOM. Studies in fluorescence of DOM, for example, greatly benefit from additional high resolution mass spectrometry as described by Stubbins et al. (2014) and fluorescence quantum yields, a coupled approach of chromophoric and fluorescent DOM further help to pinpoint DOM fluorophores with chemical compounds (Wünsch et al., 2015).

It has to be acknowledged that DOM chemistry assays can hardly ever be complete, given the several thousand chemical components existent, for example in the well-investigated Suwannee River DOM standard of the International Humic Substances Society (IHSS) (Green et al., 2015), let alone chiral molecules or isotopologues. All assays differ in their analytical depth, i.e. how exact can be an analysis in identifying a chemical compound and in their analytical width, this referring to the fraction of the total DOM domain that is explored. Bridging drawbacks of analytical depth and width is a necessity when an unskewed DOM assay is aspired (Minor et al., 2014). This can be achieved by use of several methods and proxies (Figure 3).

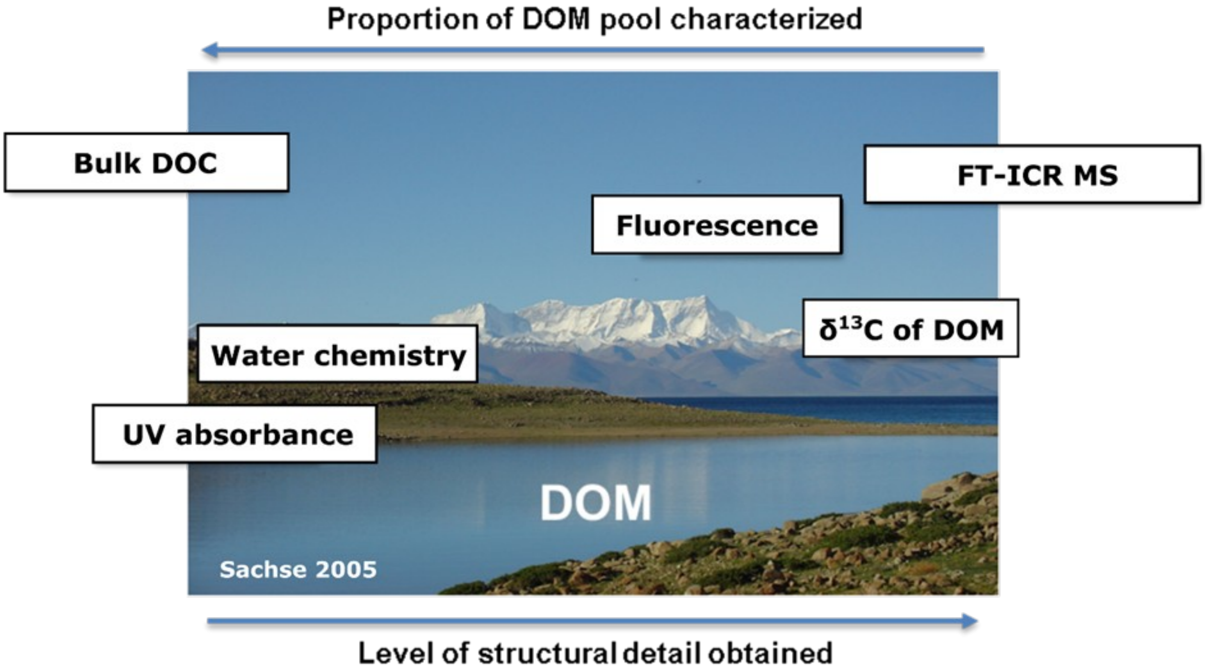

Figure 3: Methodological poles of selected proxies. Following Minor et al. (2014)

An important step in assessing properties of DOM from large multi-parameter approaches is by chemometrical data analysis (Kjeldahl and Bro, 2010). The complexity of datasets increases with the amount of dependent variables. Natural processes often are non-linear and not normal distributed (Schilli et al., 2010). This renders the straightforward analysis of data with classical methods, such as multifactorial analysis of variance (MANOVA) complicated, if not impossible. Notwithstanding, statistical analysis by ordination or dimension reduction techniques is a valuable tool in this respect, because the variety of tools allows a thorough, reproducible and flawless interpretation of data. This calls for tools and methods to decrease complexity by dimension reduction. For this, a wide variety exists between straightforward linear decomposition methods, such as principal component analysis (PCA) up to kernel and machine learning optimized non-linear applications, such as t-distributed stochastic neighbour embedding (t-SNE). In the field of ordination methods, where distance matrices are used to plot observations (samples) in low-dimensional planes, multidimensional scaling approaches (MDS) are available in metric and non-metric (NMDS) formats, up to complex manifold decompositions as can be conducted with isometric mapping (Isomap) and have been applied for long-term monitoring approaches of soil solution (Schilli et al., 2010). The combination of conceptually sound interlinked mechanistic models, together with suited multi-parameter approaches and decision-based (Kjeldahl and Bro, 2010) chemometric evaluation delivers tremendous opportunities to further employ DOM fingerprints as a monitor for the large emerging challenges of the Anthropocene.

High altitude ecosystems – admired, yet threatened

The zeal with which mountains all over the world have been and are still explored, gives evidence that mountains have always been at the core of human enthusiasm and scientific interest (Neuhaus, 2012). Mountains can comprise both, connotation as fascinating extremes or as hostile environments (Macfarlane, 2004). Not surprisingly, natural sciences have a special interest in mountains and other extreme environments, such as the Polar Regions. In the era of the Anthropocene, these areas at least partly bear the chance to be near-pristine and not as much superimposed by modern human action as many other regions worldwide (Meybeck and Helmer, 1989). Concomitant, polar (Turner, 2011) and mountain ecosystems appear to be especially threatened by global climatic change (Birrell et al., 2020; Rangwala and Miller, 2012). Their ecosystem service supply, e.g. for world wind oscillations or the provisioning of freshwater, consequently are especially under pressure, with potential global implications (Kohler et al., 2010). Research activities in remote regions of the world hence are a key mission for the scientific community.

The Tibetan Plateau

The Tibetan Plateau (TP) and southward Himalayas formed by the collision of the Indian subcontinent with the Eurasian plate around 40 Ma ago (Renner, 2016). The Himalayas comprise the highest mountains in the world, with ten summits of elevation > 8000 m. asl. The TP is located in central Asia and spans an area of more than 2.16 Mkm² (Study 1, Figure 1). It has an average elevation of around 5000 m. asl. with an altitudinal gradient decreasing from the south towards the north, where mid-latitudes and hilly regions indicate the flattening toward the north. The TP adjoins the Karakorum to the west and the Altun-Qilian-Kunlun to the north, the Hengduan Shan forms the eastern border of the TP. The Himalayas in the south of the TP form a huge orographic barrier for the dominating Indians Summer monsoon and East Asian monsoon (Wünnemann et al., 2018), leading to increasing aridity northwards (Zhao et al., 2020). The TP is comparably flat, indicating limited folding during orogeny, but uplift (Wang et al., 2012), except for fracture zones, where mountain ranges such as the Nyainqentanglha were formed (Dong et al., 2011). Temperatures on the TP are dominated by the elevation mass effect (Haffner, 1997), leading to mild summers with monsoonal-driven greater humidity and cold and mostly dry winters. The TP comprises several biomes, following the height- and aridity-gradient in north-south direction (Miehe et al., 2019). The Himalayas as the southern fringe of the TP comprise an extended forest belt, followed by alpine scree, glaciers and montane steppe around the highest elevations of the Himalayas. Further northward, alpine pastures, composed in majority of the sedge *Kobresia pygmaea*, form the world's largest connected alpine pasture area (Miehe et al., 2008) with active pastoralism (Gongbuzeren et al., 2018) and rich cultural heritage (Bellezza, 1996), with the yak livestock in the centre of local agronomy (Miehe et al., 2019). Due to long-term reported pasture degradation, alpine pastures have been in the core of scientific concern (Damm, 1998; Harris, 2010; Schleuss et al., 2015), putting the livelihoods of many communities at risk (Hopping et al., 2018). Further northward, alpine steppe vegetation dominates (Miehe et al., 2011), interchanging with alpine wetlands, sedge swamps and salt marshes (Wang et al., 2020). Alpine steppe has been reported to benefit from reported warming and wetting on the TP (Li et al., 2010; Nieberding et al., 2021)- At the same time, alpine pastures and alpine wetlands are threatened by degradation (Gao, 2016), leading to eutrophication and browning of freshwater resources (Zhang et al., 2020).

The TP is recognised as the Asian water tower (Bandyopadhyay, 2013), as it forms the sourcing area of several large streams that drain east and south-east Asia and deliver huge freshwater resources to billions of people downstream of the TP (Chen et al., 2016b). The TP is also known as the 'third pole environment' due the large extent of glaciation (Qiu, 2008; Yao et al., 2012), comprising the only cold pole outside of the circumference polar regions. This large cold pole also bears implications for global wind oscillations, as was shown by numerical modelling for the dynamics of the Indian and East Asian summer monsoon (Dell'Osso and Chen, 1986; Lau and Li, 1984; Liu and Yin, 2002). The reported warming of the TP could hence lead to important ramifications for supra-regional climatic correlations. The large glacial inventory of the TP and Himalayas is well investigated (Bolch et al., 2010; Spencer et al., 2014; Xie Shu-cheng et al., 1999; Zhou et al., 2019) and has been reported to be threatened by accelerated climatic change (Song et al., 2020; Yao et al., 2019). The implications of pronounced warming and related increase of glacier wastage translate to local effects, such as the hazard of larger sediment resuspension (Chen et al., 2016a) and loss of soil resources (Menges et al.,

2019) but also threaten the water supply of whole Asia (Gao et al., 2019) and even have global implication e.g., since the larger output of cold water from the melting glacial environment is shown to hamper winter time water mixing in the Arabian Sea and presents a driver for algae bloom, leading to a threating of fish biodiversity and local populations welfare (Goes et al., 2020).

The TP is a fascinating environment, of unparalleled natural beauty and scientific value, it harbours an hotspot of biodiversity (Salick et al., 2007) and is an important repository for the history of human evolution (Chen et al., 2019). Nonetheless, TP's nature is affected by multiple and multiplied threats (Qiu, 2014), not only posing a risk at the livelihood of local communities, but further impacting ecosystem services and welfare for billions of people in Asia and even beyond.

The Nam Co watershed

The large, world important streams have their sourcing catchments in the fringe areas of the TP, while the largest part of the plateau is arheic (Meybeck, 1995). It comprises a large number of endorheic lakes, especially larger lakes with an area >100 km². In total 2 % of the plateaus surface area is covered with lakes (Meybeck, 1995). Endorheic basins respond sensitively to changes of biotic and abiotic factors by physical, chemical and ecological parameters (Frondini et al., 2019; Mason et al., 1994. Endorheic lakes therefore allow excellent opportunities for research activities related to climatic change and other human-induced challenges, such as land-use intensification. Lakes and their endorheic watersheds hence can be used as sentinels for larger-scale responses (Adrian et al., 2009). Consequentially, plateau lakes and their catchments have been in the centre of scientific interest for several disciplines, including glaciology (Li et al., 2014), paleoecology (Gasse et al., 1991; Kang et al., 2019), limnology (Wang et al., 2011) and biogeoecology (Gao et al., 2020) in the light of current climatic change.

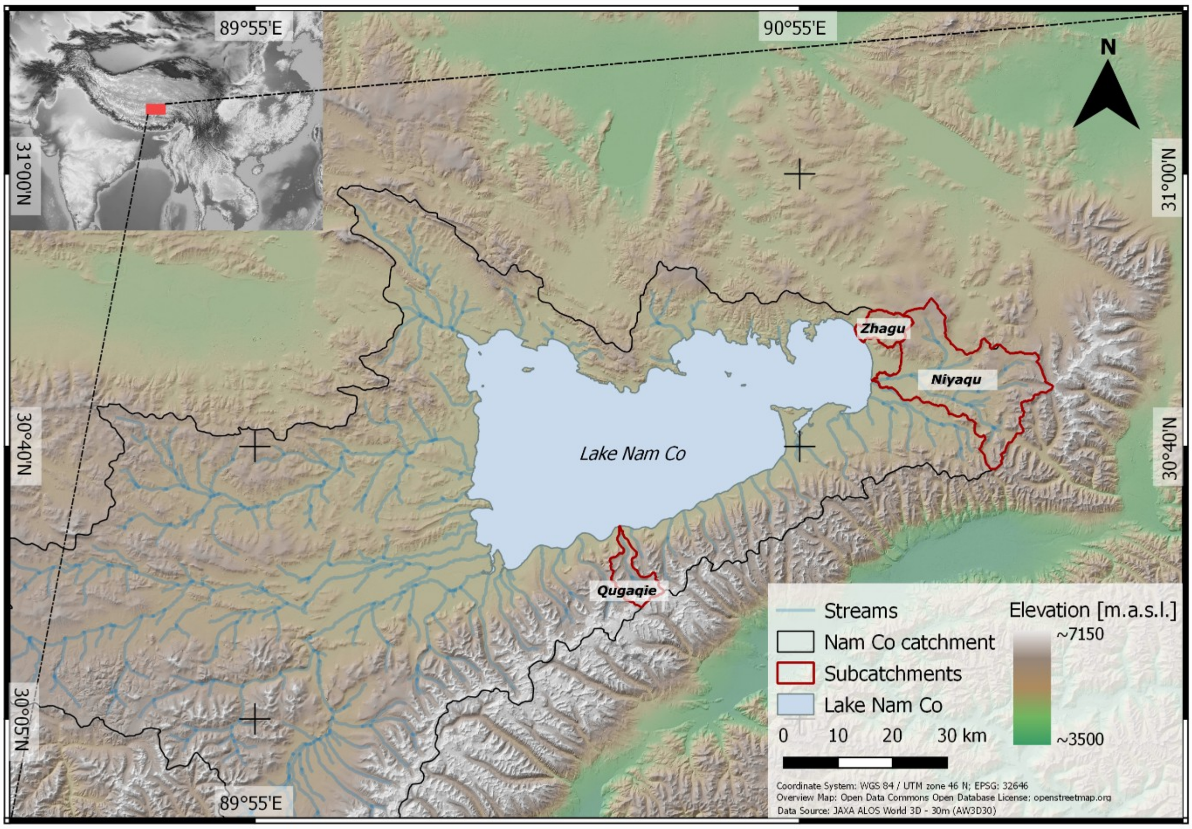

Figure 4: Location of the Nam Co watershed and lake on the southern Tibetan Plateau (TP)

While there are numerous examples of lakes, this thesis zooms in to Lake Nam Co and its watershed. This lake is located in the south of the TP (Figure 4). The Nam Co watershed is especially suited for investigation, by its positioning in several transition zones. A) The progression between the relatively moist, mountainous, highly to extremely dissected (Meybeck et al., 2001) Himalaya-affected southern zone and the arid, more flat, poorly dissected northern parts of the plateau. The Nam Co watershed is delimited by the outcrops of the glaciated Nyainqentanglha mountains to the south and south-east, while the northern and north-western margins stretch in hilly uplands. B) The watershed further lies in a transition of two major biome types (Miehe et al., 2019). The alpine *K. pygmaea* pasture expanding from the south intersects here with the alpine steppe, dominating the more arid inward plateau. The Nam Co watershed was found to be especially affected by warming (Frauenfeld, 2005) and wetting, as tracked by lake level rises (Zhang et al., 2011). The moisture surplus was attributed to increased glacial runoff (Lei et al., 2013) and precipitation gains (Zhu et al., 2010) resulting in greening. Gains in green plant cover were reported in the alpine steppe biome (Nieberding et al., 2021), while so far no reports are available for alpine pastures. Given the special positioning of the Nam Co watershed in two transitions, the catchments of Nam Co represent different landscape units in close proximity (Figure 5). The catchments have different tectonic origin, resulting in different geology, orientation and steepness. Abiotic and biotic factors superimpose catchments to a differing degree by geomorphological, hydrological and climatic forcing (Keil et al., 2010).

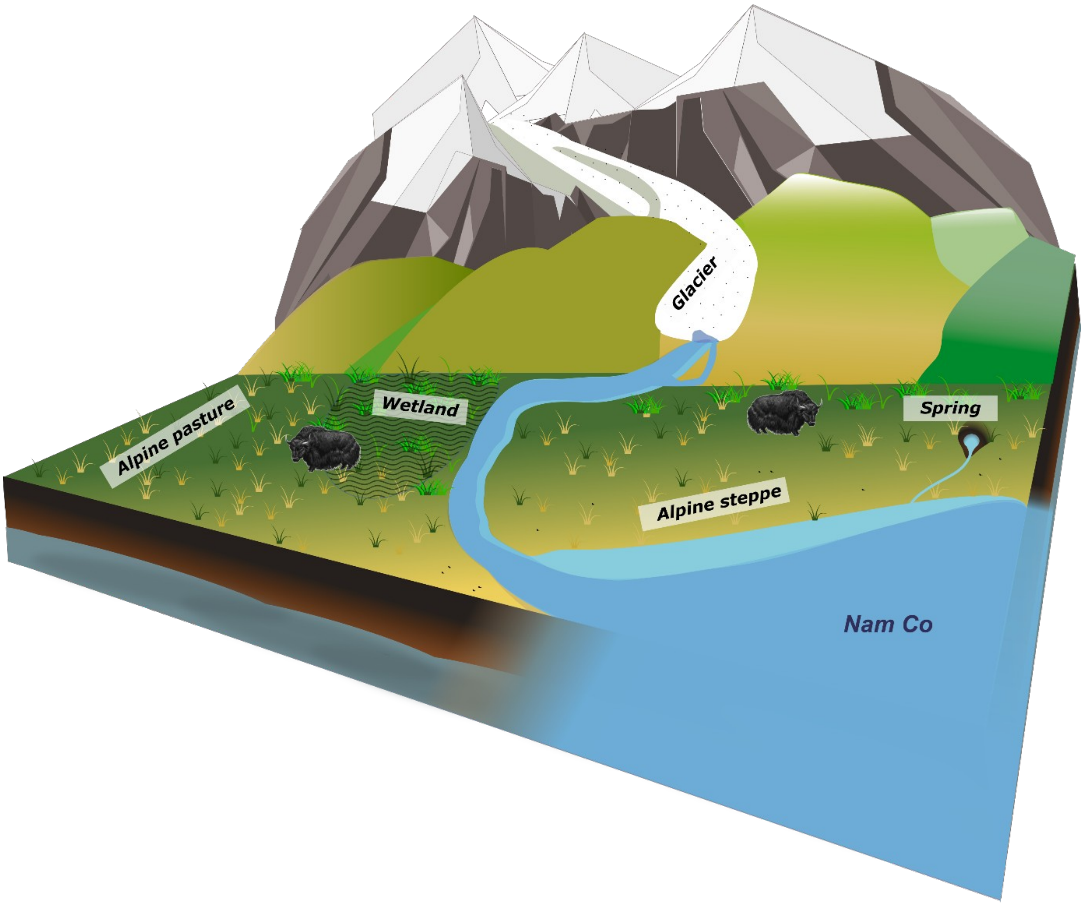

Figure 5: Block image of the investigated watershed and lake

Rationale and objectives

Finding early warning signals for critical transitions in nature is a global challenge (Scheffer et al., 2009). Comprehending the fluxes and dynamics of dissolved organic matter between different landscape units of the critical zone, is a cornerstone to predict ecosystem change (Chorover et al., 2011; Perdrial et al., 2010; Simon, 2021) in an ever more challenged world (Meybeck, 2003). DOM can be employed using its monitor function, tracking early warning signals of critical transition processes of ecosystems ecological state (Ritson et al., 2014). Globally, especially lake ecosystems have fallen short in investigating DOM dynamics (Minor and Oyler, 2021). Even more so, a gap on characteristics of organic matter discharge, composition and processing persists for entire High Asian ecosystems on catchment scale (Li et al., 2018) and in particular for glaciers (Zhang and Kang, 2017) and wetlands (Gao and Li, 2016). This is grave, given the significance of the TP for world climate and transregional water security and with respect to its high natural sensitivity. Studies of DOM characteristics and processing are scarce on the TP and either employ overview studies from large rivers (Qu et al., 2017) or draw their conclusions from small sample sizes and single investigations (Li et al., 2021; Spencer et al., 2014), by this potentially over generalising research findings. Endorheic watersheds and terminal lakes are known for their sentinel function (Adrian et al., 2009; Williamson et al., 2016). These systems are promising study objects, for different reasons: the short-circuited ecosystem cycles allow 1) easy sampling due to relatively short distances, 2) comparably short response times of terminal aquatic systems to changes in the source area and 3) the utility that comes from the response integration effect of closed basins. Investigations of catchments of endorheic watersheds on the TP hence give the opportunity to downscale excessively generalized research findings, by focusing on site-specific and small–scale environmental properties and seasonality, often found to govern characteristics of soil organic matter (Spielvogel et al., 2009) or watershed DOM signatures (Kai et al., 2019; Mosher et al., 2015).

Our objective therefore is to employ the monitor function of DOM, to track influences of sites and seasons, employed by a multi-parameter dataset. This allows to utilise the sentinel function of the endorheic Nam Co Lake basin, in order to investigate ecosystem responses, for example on degradation of rangeland or glacier melt. Addressed to the outlined research gap, of DOM as the missing link between studies of pasture degradation and research dealing with lake responses to climatic change, we aim to draw conclusions about DOM characteristics, dynamics, processing and transformation, representative for wide parts of the southern TP.

Hypotheses

H 1 Catchment biogeodiversity in the endorheic Nam Co watershed governs DOM characteristics and water chemistry parameters. Investigated endmembers (glaciers, springs and alpine wetlands) possess unique DOM signatures compared to the DOM of streams, influenced by biomes, the brackish intermixing zone and the terminal lake.

H 2 Seasonality influences DOM signatures, given the climatic controls on the TP with cold and dry winters and warm and moist summers. Freshet has minor effects on DOC concentrations and DOM composition due to the absence of a closed snow cover, hampering DOM. In contrast, the Indian summer monsoon delivers warm and moist air masses, leading to a temperature increase and sufficient humidity, promoting a compositional change in DOM.

H 3 Processing of DOM in form of biotic utilisation and photooxidative degradation is expected during the fluvial pathway from the source area towards the terminal lake. DOM undergoes a degradation cascade.

H 4 As a consequence of DOM biotic and photooxidative degradation, DOM composition of the terminal aquatic basin in Lake Nam Co, **H4a** is chemically distinct from sources in the catchments. Due to altitude-dependent strong solar radiation and a clear and deep water column of the Lake with two yearly phases of full circulation (dimictic), aromaticity-markers, such as chromophoric DOM (CDOM) originating from terrestrial DOM sources are expected to be preferentially depleted. Lake DOM is further expected to be relatively uninfluenced from seasonality by its large water volume and removal of seasonality influences by DOM processing. **H4b** DOM of lake Nam Co is supposed to be of an autochthonous microbial derived origin.

In order to test the hypotheses, the following studies were carried out:

I. **Reviews and syntheses: How do abiotic and biotic processes respond to climatic variations in the Nam Co catchment (Tibetan Plateau)?**
Study I included an extensive, multidisciplinary review of the Nam Co watershed as a case study area for closed basins in the southern TP. The paper deciphered the state of the art of scientific activities in the fields of organic carbon, sediments and water. By this insights into conducted investigations at the Nam Co watershed were gained, their outcome

identified and persisting knowledge gaps were outlined, leading to the formulation of hypothesis **H1** to **H4**.

With focus on climatic changes and other present challenges in the Nam Co watershed, the paper outlines responses of biotic and abiotic factors. Socio-cultural aspects such as intensification of land use by pastoral practise, together with fencing activities received special attention. It was defined, that the Nam Co watershed acts as a natural laboratory, to investigate ecosystem responses on climatic change and land use intensification, constituting a so-called: sentinel function. Building upon this, the paper delineates persisting research gaps in the field of biogeochemistry and natural organic matter research. The dynamics and fate of dissolved organic matter were identified as major missing link between two strong focal areas: 1) Studies on terrestrial processes, such as increased glacial wastage, and degradation of alpine pastures that remain plot based or are limited to effects in their particular biome / area of study and 2) processes in the lake, such as changes of the water balance, the foodweb of the makrozoobenthos and paleoecological studies, which seldom take present-day catchment processes into account. Based on the reviewed literature, the importance of DOM investigations as a link of processes in the terrestrial sphere with highly potential reactions in the aquatic regime were emphasised. Perspectives to further comprehend changes of geo- and biodiversity as affected by land use and climatic change in the Nam Co watershed are presented.

II. The glacial – terrestrial – fluvial pathway: A multiparametrical analysis of spatiotemporal dissolved organic matter variation in three catchments of Lake Nam Co, Tibet, PR China

Study II first aimed at closing the knowledge gap of plant cover information in the Nam Co watershed. Information about significant changes in green plant cover, i.e. greening in terms of a significant increase of plant cover, or browning, in case of a significant loss, was compiled by application of a 30 year watershed-wide satellite dataset. This dataset serves as the basis to draw conclusions which vegetation types influence DOM in the streams. This information contributed to **H1**.

The second aim was the compositional characterisation of DOM, with respect to site specific influences from three catchments within the Nam Co watershed by employing a large multi-parametrical dataset. The catchments were chosen to cover the transition between biome types and between the mountainous southern branch of the Lake Nam Co watershed and the more arid plateau stretching northwards. Different sampling locations were selected to cover the influence of landscape units, such as glaciers, springs, alpine wetlands, streams and the lake. To gain maximum insights into DOM diversity and water chemistry, several parameters were combined: water chemistry parameters (dissolved inorganic carbon, electric conductivity, pH, concentration of cations and anions), dissolved organic carbon concentrations and DOM characteristics such as chromophoric DOM, PARAFAC-resolved fluorophore DOM and $\delta^{13}C$ of DOM. In total 138 samples were taken from the respective source area (including endmembers: glaciers, springs and wetlands) until the river mouth, enabling the testing of **H1** and **H4**. Also, the change of DOM characteristics along the flow path of streams was covered, allowing to test for processing and transformation of DOM, as implemented by hypothesis **H3**. Water samples were taken in three sampling campaigns, in order to cover the effect of freshet, the onset of the Indian summer monsoon and the late year baseflow, hypothesis **H2** was tested by this.

III. Dissolved organic matter sources and processing in the endorheic Lake Nam Co catchment (Tibet) as assessed by ultra-high resolution Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS)

In study III an ultra-high resolution assessment of a subset of 48 samples by undirected electrospray ionisation Fourier transform ion cyclotron resonance mass spectrometry (ESI FT-ICR MS) of solid phase extracted (SPE) DOM was conducted. The samples were taken during the late year baseflow sampling campaign at Nam Co in 2019. The methodology employed in this paper allowed to decipher the DOM characteristics of the three catchments under investigation in the Nam Co watershed and the effect of landscape units, including the three endmember groups: glaciers, springs and alpine wetlands as well as streams, the brackish intermixing zone and the lake. By this, **H1** was further corroborated with a better in-depth understanding of the molecular composition of DOM. This application also led to a deeper investigation of DOM processing and transformation of stream samples, taken along the stream flow route. By this, a critical discussion of **H3** was possible, while the ultra-high analytical resolution of chemical composition in samples allowed confirmatory conclusions concerning hypothesis **H4**.

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Study I

Reviews and syntheses: How do abiotic and biotic processes respond to climatic variations in the Nam Co catchment (Tibetan Plateau)?

Contribution: I contributed equally to the content of the Manuscript and structured the main text body. I authored the following chapters: 'Vegetation, soils and pasture degradation in the catchment', 'Effects on carbon cycling in alpine ecosystems' as well as 'Holocene vegetation cover and climate reconstruction based on pollen records' and composed Figure 4.

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Reviews and syntheses: How do abiotic and biotic processes respond to climatic variations in the Nam Co catchment (Tibetan Plateau)?

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Abstract. The Tibetan Plateau (TP) is the largest alpine plateau on Earth and plays an important role in global climate dynamics. On the TP, climate change is happening particularly fast, with an increase in air temperature twice the global average. The particular sensitivity of this high mountain environment allows observation and tracking of abiotic and biotic feedback mechanisms. Closed lake systems, such as Nam Co on the central TP, represent important natural laboratories for tracking past and recent climatic changes, as well as geobiological processes and interactions within their respective catchments. This review gives an interdisciplinary overview of past and modern environmental changes, using Nam Co as a case study. In the catchment area, ongoing rise in air temperature forces glaciers to melt, contributing to a rise in lake level and changes in water chemistry. Some studies base their conclusions on inconsistent glacier inventories but an ever-increasing deglaciation and thus higher water availability have persisted over the last decades. Increasing water availability causes translocation of sediments, nutrients and dissolved organic matter to the lake, as well as higher carbon emissions to the atmosphere. The intensity of grazing has an additional and significant effect on CO_2 fluxes, with moderate grazing enhancing belowground allocation of carbon while adversely affecting the C-sink potential through reduction of above- and subsurface biomass at higher grazing intensities. Furthermore, increasing pressure from human activities and livestock grazing are enhancing grassland degradation processes, thus shaping biodiversity patterns in the lake and catchment. The environmental signal provided by taxon-specific analysis (e.g. diatoms and ostracods) in Nam Co revealed profound climatic fluctuations between warmer/cooler and wetter/drier periods since the late Pleistocene and an increasing input of freshwater and nutrients from the catchment in recent years. Based on the reviewed literature, we outline perspectives to further understand the effects of global warming on geo- and biodiversity and their interplay at Lake Nam Co, which acts as a case study for potentially TP- or even worldwide processes that are currently shaping high mountain areas.

Key words: bioindicators, carbon cycle, climate change, climate reconstruction, deglaciation, geobiodiversity, lake level change, Lake Nam Co, paleo-environmental proxies, pasture degradation

1 Introduction

The Tibetan Plateau (TP), often referred to as “The Third Pole” and “The Water Tower of East Asia”, is the highest and largest alpine plateau on earth (Qiu, 2008). With an area of about 2.5 million km² at an average altitude of > 4000 m above sea level (a.s.l.), it includes the entire southwestern Chinese provinces of Tibet and Qinghai, parts of Gansu, Yunnan, Sichuan and neighboring countries (Fig. 1). The southern and eastern plateau and the adjacent Himalayas regions are forming the headwaters of several major rivers (i.e., Brahmaputra, Ganges, Hexi, Indus, Mekong, Salween, Yangtze, and Yellow River), providing fresh water for ~1.65 billion people and to many ecosystems in greater Asia (Cuo and Zhang, 2017). Large proportions of the inner TP are endorheic and therefore do not drain into the large river systems. On the TP, the effects of climate change are expressed stronger than the global average, showing a steep rise in air temperature of about 0.3°C per decade since 1960 (Yao et al., 2007) and a moderate rise in precipitation during the last decades (Dong et al., 2018). The warming rate increases with altitude (Pepin et al., 2015) which is why the air temperature on the TP is soaring roughly twice the global average, thus substantially affecting the geo- and biodiversity. Glaciers and lakes are the dominant components for the Tibetan water sources, and their actual status and future development are strongly impacted by global warming. Since the 1990’s, nearly all glaciers on the TP have exhibited retreat, causing a 5.5 % increase in river runoff from the Plateau (Yao et al., 2007). The consequences of deglaciation and permafrost degradation (Wu et al., 2010) are observable in higher water and sediment fluxes, relief changes and arising natural hazards (floods, rock falls, landslides, desertification, ecosystem degradation). Consequently, landscapes are continuously being rearranged which alters the spatial distribution and composition of the inhabiting species, many of which are endemic to the TP (Walther et al., 2002). Even conservative estimates predict substantial species extinction and considerable changes to the ecosystems (Chen et al., 2011; Bellard et al., 2012). The future trajectory of such complex processes is difficult to map accurately, thus it is important to monitor the current state as well as the evolution of this highly sensitive region. The large number of water bodies on the TP, its geological diversity, climatic setting as well as sensitivity to climate change make it a unique natural laboratory, which could be used as an early warning system for other alpine environments. Many lakes on the TP are superficially closed systems, which is why they are particularly suitable as "thermometers" and “rain gauges” to measure the climatic, hydrological, geomorphological, pedological and ecological changes in their respective catchments. With an area of 2018 km², Nam Co is the second largest lake on the central TP. Currently, Lake Nam Co represents an endorheic system, acting as a sink for water, sediment and carbon fluxes. The existence of a former drainage (“Old Qiangtang Lake”) towards the northwestern Siling Co and further east, down from the TP is still under discussion (Li et al., 1981; Kong et al., 2011) (see Sec. 3.1). With good accessibility and infrastructure such as the Nam Co Monitoring and Research Station for Multisphere Interactions (NAMORS), the Nam Co catchment has become a frequent study location for monitoring and tracking of environmental changes over various timescales.

Here we present an interdisciplinary overview of how earth-surface fluxes have developed with changing environmental conditions and which consequences are to be expected for biodiversity, as well as for water, sediment and carbon fluxes within the study area of the Nam Co catchment on the central TP. In particular, this review considers past and modern geobiodiversity changes with focus on glacier retreat in relation to hydrological patterns and changes in lake water chemistry. The corresponding changes in terrestrial ecosystems concerning carbon cycle, greenhouse gas releases, as well as pasture degradation are discussed. We provide an overview of how the paleoenvironment on the Tibetan Plateau with respect to landscape evolution around Nam Co was shaped by geodiversity, lake level changes and Holocene vegetation cover. Lastly, based on the available studies, this review identifies the major research gaps that are awaiting further exploration and comparison with other high-altitude environments.

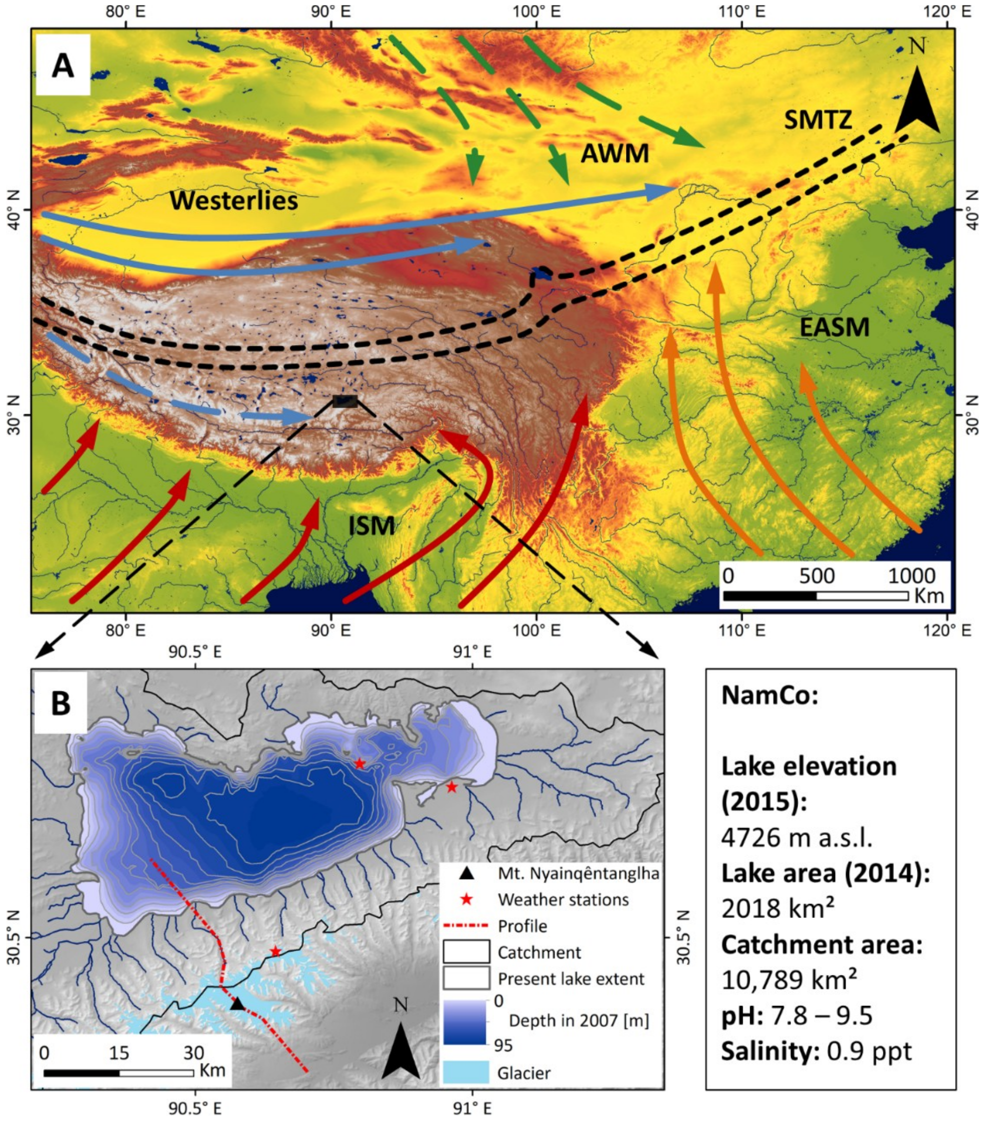

Figure 1. Major atmospheric systems governing the climate in China (A); Nam Co study site (B); and characteristics of Nam Co's catchment (C). A: Continuous arrows indicate systems active in summer. These are the Indian Summer Monsoon (ISM) in red, the East Asian Summer Monsoon (EASM) in orange and the Westerlies in blue. Dashed arrows represent systems active in winter. These are the Asian Winter Monsoon (AWM) in green and southern parts of the Westerlies in blue. The black dotted lines denote the Summer Monsoon Transition Zone (SMTZ) (after Wünnemann et al., 2018). Background elevation data according to SRTM DEM v4 (Jarvis et al., 2008). B: Nam Co catchment, including the current lake extent (based on Copernicus Sentinel data 2018, processed by ESA), its bathymetric depth in 2007 (Wang et al., 2009a), the outline of the catchment (after Keil et al., 2010), glaciers of the Nyainqêntanglha Range (GLIMS and NSDIC 2005, updated 2018) and rivers discharging into Nam Co (SRTM DEM v4; Jarvis et al., 2008). The red dotted line indicates the profile position of Figure 2. C: Characteristics of Nam Co: Lake elevation (Jiang et al., 2017), lake surface area (Zhang et al., 2017), catchment area, lake pH and salinity (Keil et al., 2010).

2 Environmental changes in lake Nam Co and its catchment

2.1 Climatic characteristics of the Nam Co basin

The prevailing climate at Nam Co is characterized by strong seasonality, with long, cold winters and short but moist summers. During winter, the Westerlies control the general circulation and lead to cold and dry weather, with daily temperature minima below -20 °C. In springtime, the TP heats up and allows the melt water to percolate to deeper soil layers. The drought situation increases gradually until the monsoon rains arrive, typically between May and June. During autumn, weather shifts again to clear, cold and dry conditions (Yao et al., 2013). The mean annual temperature measured at the NAMORS research station (Fig. 1) between 2006 and 2017 was -0.6 °C and the annual precipitation was between 291–568 mm (mean = 406 mm), with the majority occurring during the monsoon season from May to October (Tab. 1). The onset and strength of monsoonal precipitation varies substantially between individual years and can be delayed by up to six weeks, depending on the altitude and latitude on the TP (Miehe et al., 2019). Precipitation rates are subject to spatial variations due to the > 7000 m high Nyainqêntanglha range which represents the southern border of the lake catchment. This leads to considerably larger glacial areas in the southwestern part (~700 km^2) than in the northeastern part of the mountain range (~100 km²) (Bolch et al., 2010).

Table 1: Average daily air temperature (maximum, mean, minimum in °C) and average daily precipitation (sum in mm) from NAMORS from 2006 to 2017. Calculations were performed using the tidyverse package family in R on RStudio environment (Wickham, 2017; RStudio Team, 2018; R Core Team, 2019). Data provided by ITP Beijing, for details about sensor eqiupment see (Ma et al., 2009).

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Ø / Σ
T max	-0.7	-1.5	1.5	4.7	11.6	13.3	12.6	12.2	11.3	8.7	2.5	1.2	6.4
T mean	-10.8	-9.7	-5.7	-1.4	3.1	7.9	9.1	8.3	6.5	0.3	-6.5	-8.4	-0.6
T min	-21.5	-20.5	-14.3	-7.3	-4.5	1.1	5.2	3.2	-1.3	-14.7	-15.3	-19.1	-9.1
Precip.	4	1	3	13	23	41	85	117	81	34	5	1	406

2.2 Glacier retreat and hydrological patterns of Nam Co

The rise of satellites such as Envisat, CryoSat and ICESat and the increasingly wide-spread availability of their data, have enabled the accurate study of lake and glacier parameters as far back as the early 1970's (Wu and Zhu, 2008; Zhu et al., 2010b; Liao et al., 2013). The size of Nam Co as well as the extent and distribution of glaciers in the Nyainqêntanglha range have been the subject of many publications over the recent years (Yao et al., 2007; Frauenfelder and Kääb, 2009; Bolch et al., 2010; Wang et al., 2013; Fig. 2; Table 1). Due to different data sources with varying resolutions as well as different mapping procedures, the estimated glacier area varies between different studies (Fig. 2; Table 1), as the delineation of debris- and snow-covered glaciers is rather subjective (Wu et al., 2016). This is especially true for the first glacier inventory (Li et al. 2003), which has been discussed in various studies due to inaccuracies and the quality of its base data (Frauenfelder and Kääb, 2009; Bolch et al., 2010). Nevertheless, recent studies show glacier shrinkage in the Nyainqêntanglha range at a rate of 0.3–0.5 % yr^{-1} as measured since 1970 when the first satellite images were acquired (Fig. 2; Table 1). As a result of this glacier melting, the lake surface area has expanded from ca. 1930 km² to ca. 2018 km² at a rate of 2.1 km² yr^{-1} (Fig. 3A), and the lake level has risen at a rate of 0.3 m yr^{-1} until approximately 2009, and at lower rates since then (Fig. 3B). The initial rising trends of both lake level and surface area are mirrored by most lakes in the southern part of the TP, but the slowdown of this trend observed at lake Nam Co around 2009 seems unique (Jiang et al., 2017). This suggests that the lakes on the TP react to changing environmental parameters in a variety of different ways, and that geographical proximity among lakes does not necessarily produce similar reactions to change. The effects on freshwater input to the lake are discussed in the following section (2.3). Although changes in monsoonal precipitation and wind direction may influence glacial retreat rates (Wang et al., 2013), rising temperatures remain their primary cause (Ji et al., 2018). The total contribution of glacial melt water as surface runoff to this lake level increase has been estimated ranging from 10 % to 53 % (Zhu et al., 2010b; Lei et al., 2013; Wu et al., 2014; Li and Lin, 2017), with recent studies at the lower end of this spectrum. Increased precipitation is estimated to be responsible for 50–70 % of lake growth (Zhu et al., 2010b; Lei et al., 2013). Whether there is a change in evaporation remains unclear as studies for approximately the same time period have suggested both a slightly increasing and a slightly decreasing evaporation rate since the late 1970´s (Lazhu et al., 2016; Ma et al., 2016).

Table 2. Overview of glacier area changes (%) in the western Nyainqêntanglha range (changed after Wu et al., 2016).

Period	Region of the Nyainqêntanglha range	Glacier shrinkage (%)	Reference
1970–2000	Southeastern slope	-5.2	Shangguan et al. (2008)
1970–2000	Northwestern slope	-6.9	Shangguan et al. (2008)
1970–2000	Western	-5.7	Shangguan et al. (2008)
1977–2010	Western	-22.4 ± 2.9	Wang et al. (2013)
1970–2009	Western	-21.7 ± 3.4	Wu et al. (2016)
1970/80–2000	Southwestern	-19.8	Frauenfelder and Kääb (2009)
1970–2000	Nam Co Basin	-15.4	Wu & Zhu (2008)
1976–2001	Nam Co Basin	-6.8 ± 3.1	Bolch et al. (2010)
1976–2001	Southeastern slope	-5.8 ± 2.6	Bolch et al. (2010)
1976–2009	Detailed glaciers (Zhadang, Tangse No.2, Lalong, Xibu, Panu)	-9.9 ± 3.1	Bolch et al. (2010)

The rises in temperature and precipitation are also affecting permafrost soils that are extending over an area of ca. 1.4 million km^2 (Yang et al., 2004) on the TP. The permafrost layers can be described as relatively warm and thin, with temperatures mostly > -1.5 °C and < 100 m thickness (Wu et al., 2010). The mean annual soil temperature of permafrost in particular areas of the TP has increased by 0.1–0.3 ˚C between 1970–1990 (Cheng and Wu, 2007). Simulation studies have shown that along with climate warming, the permafrost extent may decrease by 9–19 % by 2049 and by 13–58 % by 2099 (Li and Cheng, 1999; Nan, 2005). Although there is no clear estimate of permafrost extent in the Nam Co basin, Tian et al. (2009) reports a lower limit of permafrost at an elevation around 5300 m a.s.l. along the northern slopes of Mt. Nyainqêntanglha (7162 m). A frost lens was also encountered 9 m below surface (4738 m a.s.l.) while sampling an outcrop along the right bank of the Gangyasang Qu close to the northwestern lake shore in 2005 (Schütt et al., 2010). Thus, due to increasing temperatures, permafrost degradation may serve as an additional recharge factor to groundwater, resulting in increased subsurface inflow into the lakes.

Focusing on lake Nam Co, the hydraulic interaction between lake and groundwater is still uncertain, as previous studies either neglected or ignored the influence of groundwater due to a lack of reliable data (Zhang et al., 2011). However, recent studies revealed a water imbalance, which was explained by lake water seepage with an estimated outflow of 1.9×10^9 m^3 and 1.5×10^9 m^3 during 1980–1984 and 1995–2009, respectively (Zhou et al., 2013; Du et al., 2018).

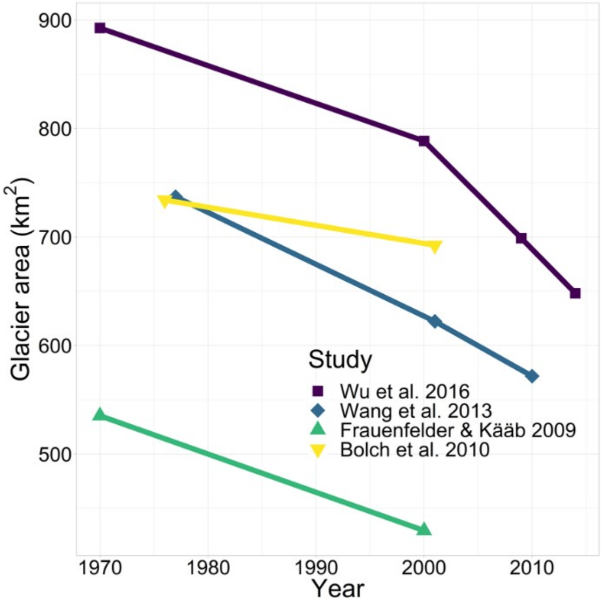

Figure 2. Glacier area reduction at south-western Nyainqêntanglha range since 1970 as evaluated in various studies.

2.3 Enhanced water availability controls changes in lake water chemistry

The maximum recorded depth of lake Nam Co is 122 m (Li et al., 2008a), with brackish water characterized by an alkaline pH of 7.8–9.5 and a conductivity of 1920 µS mm^{-1} (Keil et al., 2010). The chemical composition of a lake is essentially a function of its climate (which affects its hydrology) and the basin geology. Increased freshwater input from precipitation, melting glaciers and thawing permafrost alters the chemical composition of the lake water and enhances surface runoff, infiltration rates as well as subsurface flow. Together with the input of freshwater, streams transport dissolved organic matter (DOM) which is composed of a wide range of dissolved components and particles (≤ 0.45 µm), thus affecting the water chemistry in the lake (Spencer et al., 2014). Excessive landscape disturbance through removing vegetative cover causes higher rates of DOM leaching, more erosion and increasing water runoff velocity, resulting in additional input of minerals and nutrients into the lake. Since the process of DOM leaching and translocation itself is largely dependent on water and sediment cycles (Kaiser and Kalbitz, 2012), it represents both the seasonal and inter-annual variation in an ecosystem as well as its long-term trend. As the glaciers on the TP retreat, highly bioavailable DOM may provide additional nutrients to downstream environments and amplify the trend of eutrophication of lotic and lacustrine ecosystems. Furthermore, the rivers on the TP have been shown to transport dissolved organic carbon from thawing permafrost areas (Qu et al., 2017), which is likely rapidly degraded via microbial activity, resulting in CO_2 emissions, thus potentially producing a positive feedback on global warming. However, the research of DOM as an important allochthonous source of nutrients, and as a capture of bio- and geodiversity of its respective catchment area is largely lacking for High Asia. The concentration and ratios of different ions in the water have a regulatory impact on the structure of biotic communities (microbes, invertebrates and fish), that can best tolerate abiotic conditions (Wrozyna et al., 2012). In Nam Co, water conductivity has been regarded as the most important environmental factor for shaping communities such as archaea, bacteria, phytoplankton, and microinvertebrates (Hu et al., 2010; Wang et al., 2011). Studies demonstrated that ammonia-oxidizing archaea (autotrophic microorganisms) are key contributors to ammonia oxidation in deep and oligotrophic lakes (Callieri et al., 2016). This has implications for CO_2 fixation in the hypolimnion or the benthic zone, where there is insufficient irradiance to support photosynthesis, implying that archaea would perform the final step in the decay of organic matter via methanogenesis, resulting in carbon dioxide accumulation (e.g. when they decrease during winter). Although nitrification does not directly change the inventory of inorganic Nitrogen in freshwater ecosystems, it constitutes the only known biological source of nitrate and as such represents a critical link between mineralization of organic N and its eventual loss as N_2 by denitrification or anaerobic ammonia oxidation to the atmosphere (Herber et al., 2019). Ultimately, the changes in the communities of primary producers could alter the lake's trophic structure, which affects also the top predators of the ecosystem. The primary productivity, as an indicator of nutrient supply and a longer growing period associated with a shorter ice-cover duration, has increased markedly at Nam Co within the last 100 years (Lami et al., 2010). Wang et al. (2011) reported the increasing abundance of the diatom species *Stephanodiscus minutulus* during the last decades (ca. 1970–2001). This species is generally viewed as an indicator of water phosphorus enrichment, suggesting increasing inputs from the lake's catchment and stronger mixing in spring season. To predict future consequences of ongoing climate change, it is essential to understand the responses of biotic communities to hydrological variations. Thus, long-term monitoring is needed to adequately address the feedbacks of recent environmental changes, while climatic conditions of the past can be reconstructed through the study of organisms such as diatoms and ostracods that are sensitive to hydrologic and chemical variations (see section 3.2).

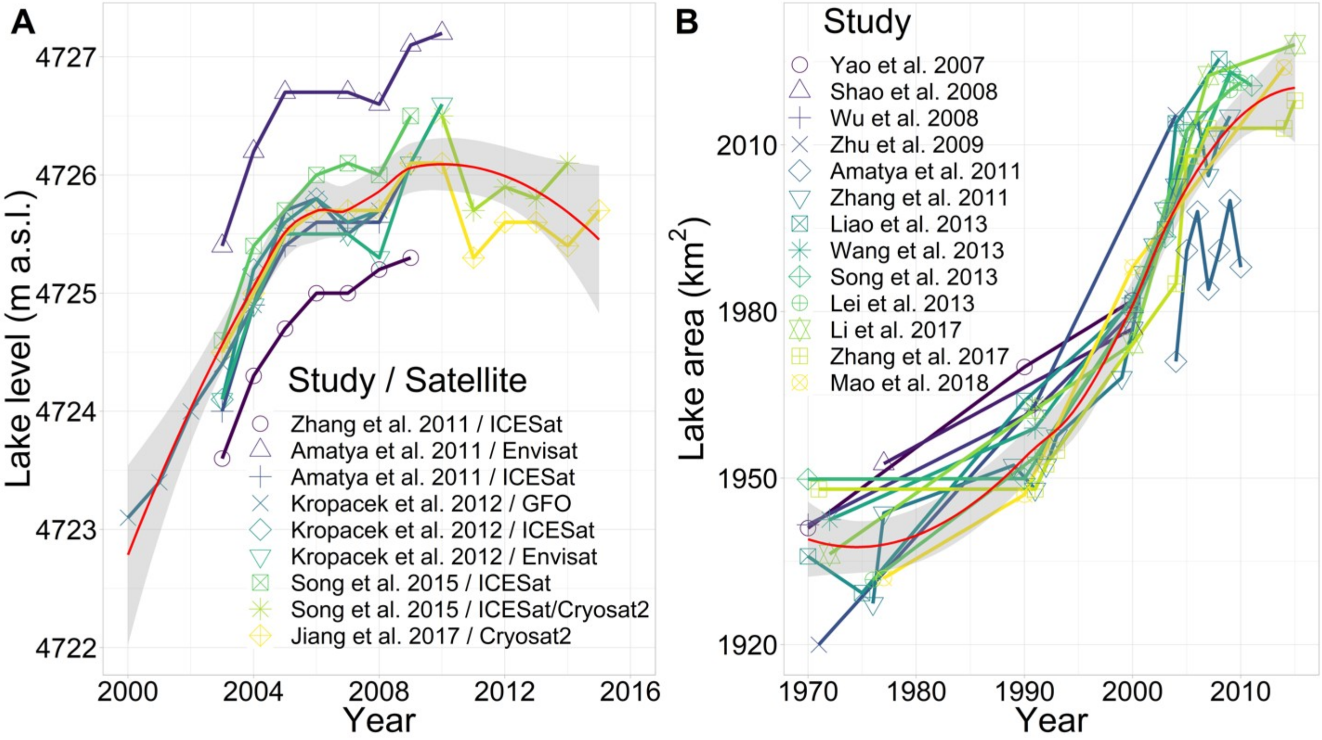

Figure 3. A) Lake level changes of Nam Co since 2000 (notable shift in the water balance in 2009); and B) changes of the lake surface area since 1970, as evaluated in previous studies. Overall increase rate of lake area is 2.1 km² yr^{-1}. Red lines denote LOESS curves with the 95 % confidence interval in gray.

2.4 Vegetation, soils and pasture degradation in the catchment

Lake Nam Co is located in the transitional zone between the central Tibetan *Kobresia pygmaea* pastures and the north western alpine steppe ecosystem (Miehe et al., 2019) (Fig. 4). Situated on the northern slope of the Nyainqêntanglha range, the vegetation pattern changes according to elevation, moisture availability and temperature. Grazing intensity and abundance of small rodents, such as the plateau pika (*Ochotona curzoniae*), may contribute to the shaping of the vegetation cover (Dorji et al., 2014; Miehe et al., 2014). The area close to the lake (< 4800 m) is covered mainly with alpine steppe vegetation consisting of *Artemisia, Stipa, Poa, Festuca* and *Carex* (Li, 2018; Nölling, 2006). Soils developed in the drier steppe areas consequentially tend to show lower organic carbon contents, naturally lowering their total C sink or source potential, as indicated by a study from Ohtsuka et al. (2008). Only one evaluable soil investigation exists from the area of Nam Co. Wang et al. (2009b) investigated two lake terrace sites, situated in the alpine steppe biome. According to their findings, the soils reflect the cold semiarid climate of the area, by showing low biologic activity, while the influence of physical weathering is dominant. The soils showed several decimeter thick layers of loess in which mainly the A-horizons were developed. Although only very sparse to moderate vegetation cover occurs, an almost 30 cm thick organic rich topsoil with granular structure was developed there (Wang et al., 2009b). Further organic-rich buried horizons were found and dated in both profiles, showing phases of climatic conditions enabling the buildup of organic material related to warm-wet periods in the past (before 2.4 and 1.6 cal ka BP) and interchanging with phases of erosion, leading to e.g.: sheet erosion, the formation of gullies and alluvial fans, supposedly during colder periods. These results fit well to climate reconstructions presented in part 3.3 of our review. In accordance with the World Reference Base for Soil Resources (WRB) classification, we propose that the soils described by Wang et al. (2009b) can be classified as Calcisols as there is evidence of carbonate translocation.

Higher up the slope (4800–5200 m), the alpine steppe is replaced by *Kobresia pygmea* pasture. Wang et al. (2007) and Kaiser et al. (2008) investigated the relationship between plant communities and development of soil types on the High Asian Plateau and for pasture soils in the wider area. Vegetation strongly controls the input of organic material into the soil, but beyond that also stabilizes fine materials (< 0.1 mm) and governs the degree of chemical weathering. The authors found soils with stronger signs of biologic activity and chemical weathering (e.g. Cambisols) associated with alpine pasture sites. *Kobresia* root mats are usually developed in up to 40 cm thick loess layers and form a distinctive felty horizon which protects against erosion. The genesis of this felty root mat is attributed to *Kobresia pygmaea,* since this shallow rooting, small plant allocates most of its biomass belowground and is able to reproduce vegetative, making it well adapted to the high grazing pressure (Miehe et al., 2008). The curious dominance of *K. pygmaea* is often linked to grazing: (i) *K. pygmaea* replaces taller plants at sites where grazing pressure is increased experimentally. (ii) Several enclosures show that other grasses and shrubs gain in dominance after grazing competition ceased (Miehe et al., 2008). Hence, the felty root-mat can be seen as an effect of an anthropozoogenic plagioclimax. At higher elevation (5200–5900 m), only alpine sparse vegetation associated with initial soil processes occurs (Ohtsuka et al., 2008).

Where water availability is abundant, alpine swamps with *Carex sagensis and Kobresia schoenoides* are formed, especially at source areas, along river banks and in waterlogged depressions, some of which can cover large areas (Li et al., 2011). Concerning soil development in alpine wetlands, the data base is sparse compared to the alpine pasture and steppe biome. It was pointed out for alpine pastures, that a strong relationship exists between plant communities and (top)soil genesis. This relationship probably also holds true for alpine wetlands, with the exception, that the influences of water logging and seasonal fluctuations and frost-melt cycles in the water table are likely to have an effect on soils. This can be expressed in terms of formation of gleyic features, frost turbations, heaves or other azonal features related to the soil forming effects of water (Chesworth et al., 2008). It still needs to be clarified, how these water-logged areas effect the cycling and processing of organic matter and nutrients. There is no evidence of tree species, only the evergreen shrubs of *Juniperus pingii var. wilsonii*, which are mainly found on the south-facing slopes of the northern Nam Co catchment, and shrubs of *Salix* spp. in the Niyaqu Valley in the eastern lake catchment (Li, 2018). Alpine steppe comprises more plant species compared to pasture and marsh ecosystems, which are predominantly covered with *Carex* spp. and *Kobresia spp.* (Miehe et al., 2011b). Alpine pastures are often described as "golf-course-like" (Miehe et al., 2014) with the intention of illustrating their unique plane surface. However, small scale structures such as thufa or hummocks are also present. The origin of these structures around Nam Co remains unclear; however, frost heave and permafrost degradation processes are considered to play a major role (Adamczyk, 2010). The landscape, generally dominated by endemic *Kobresia pygmea* sedges, harbors only a few other species (Miehe et al., 2019), but the additional microhabitats provided by thufa and hummocks enable rarer and low-competitive species to settle in niches in these heterogeneous structures (Vivian-Smith, 1997). Compared to the surroundings, the microtopography of thufa possesses different degrees of wetness, exposition and insulation, depth of soil material and type of topsoil. Local studies of the Nam Co area state that slightly degraded bare soil patches and gullies are often areas where plants have the chance to evade the suppression of the closed *Kobresia pygmea* root mat (Schlütz et al., 2007; Dorji et al., 2014). Thus, the genesis of thufa and mild, limited degradation processes are likely to increase species richness and diversity by cracking open the closed root mat of alpine pastures. These structures can also be formed by grass species that grown in tussocks (i.e. clumps, bunches or tufts), such as the endemic species *Stipa purpurea* (Liu et al., 2009) or *Kobresia tibetica* (Yu et al., 2010) and *Kobresia schoenoides* (Nölling, 2006).

The often cited degradation of alpine pastures is likely initiated by natural polygonal cracking (Miehe et al., 2019), which can occur through drying (Velde, 1999), and then tend to be amplified by livestock trampling and plateau pikas using the cracks as highways (Liu et al., 2017b; Hopping et al., 2016). Overgrazing in alpine pastures is one of the most frequently mentioned causes of pasture degradation (Unteregelsbacher et al., 2012; Harris, 2010; Miehe et al., 2008) as excessive trampling by livestock might aggravate the initial conditions of polygonal cracking (Miehe et al., 2019). This effect, however, seems to be limited to the direct vicinity of herder ´s settlements and camps (piosphere-centers), and many factors that are usually attributed to degradation rather proof to be environmentally controlled, especially in drier areas (Wang et al., 2018b). Some researchers argue that climate change is the dominant or even sole driver of degradation (Wang et al., 2007), although the effects of rising temperatures and increasing precipitation appear to be an intensifier rather than the cause of degradation (Zhou et al., 2005; Harris, 2010). In turn, both Wang et al. (2018b) and Cao et al. (2019) point out that a multitude of effects might be in play, with a locally differing magnitude or even reversion, while usually moderate grazing was not to be found to cause degradation. Certainly, there are more factors than just grazing-pressure, and there might be site-specific effects leading to non-equilibrium behavior of the study object, be it pasture or steppe (Wang and Wesche, 2016). Plot-level experiments from the Nam Co area found warming to have significant effects on the shallow rooted *Kobresia pygmaea* by reducing the number of flowers and delaying its reproductive phenology. These changes were provoked by simulating increasing precipitation by means of snow addition (Dorji et al., 2013) and also by maintaining a moderate level of grazing combined with snow addition (Dorji et al., 2018). This underlines the importance of climate forcing on the terrestrial systems in the Nam Co catchment. Grazing should not be seen as a disturbance but as an integral part of a non-steady state but plagioclimax environment. Currently there are no estimates of the extent of degraded land at Nam Co, but the degradation of wide areas of alpine pastures is not without consequences for the pastoralist communities. The severe degradation and sloughing off of the whole topsoil remove the basis for business and might lead to unknown consequences for the lake ecosystem by means of enhancing or terminating nutrient exchanges. The economic rationale of herders might be to increase the numbers of livestock as this represents a form of social security (Simpson et al., 1994). The bottom line is that conflicts arise as less land is available for grazing (Hopping et al., 2016).

The Chinese government has favored policies such as sedentariness and fostered the construction of stationary settlements, which have, in turn, created hotspots of overgrazing (Miehe et al., 2008). In these hotspots, large portions of the topsoil are lost by erosion and denudation, leaving only an area of humic material or subsoil, thus being called “black beach” (Miehe et al., 2008) or “black-soil patch” (Liu et al., 2017a). The remaining landscapes are usually dry, poor in plant cover and prone to further degradation. Increasing areas of bare soil patches enhance evapotranspiration, causing earlier cloud cover formation especially before noon. This may, in turn, lead to reduced radiation and temperature at the surface, thus hampering photosynthesis and consequently overall carbon sequestration (Babel et al., 2014). However, the evolution of grasslands on the TP has been accompanied by herbivore communities, thus, the plants have developed coping mechanisms to persist under continuous grazing pressure (Miehe et al., 2011a). According to the intermediate disturbance hypothesis, species diversity is higher under moderate disturbances, which suggests the positive effect of intermediate level of grazing pressure. Indeed, a plant clipping experiment to simulate grazing demonstrated that under the effect of climate warming, the grazing activities mitigated the negative effects of rising temperature by maintaining a higher number of plants (Klein et al., 2008). Many studies hold the traditional nomadic practice to be a sustainable one (Miehe et al., 2008; Babel et al., 2014; Hafner et al., 2012), but the current policy of removing pastoralist lifestyles from certain regions could potentially reduce overall species richness.

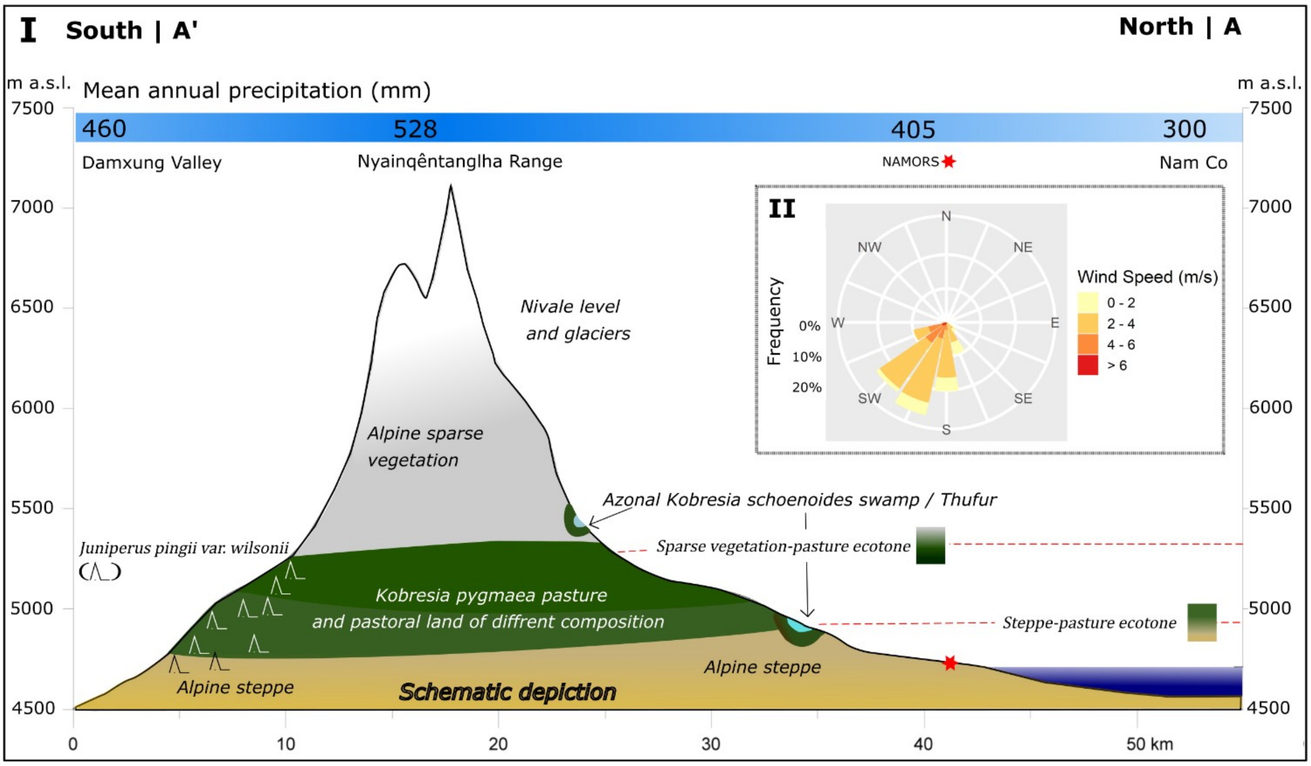

Figure 4. I: Cross-section from Damxung valley to lake Nam Co study area (A'-A) as shown in Fig. 1 (B). Schematic depiction of altitudinal dependent biomes and azonal landforms, changes in chroma denote height-dependent biome shifts. Approximate biome heights were gained from: satellite imagery (Sentinel 2B) and herewith derived vegetation indices, field excursion and literature review (Ohtsuka et al., 2008; Wang and Yi, 2011). II: Frequency, direction and velocity of mean daily wind measurements at the NAMORS (30°46'22" N, 90°57'47" E) between 2005–2015.

2.5 Effects on carbon cycling in alpine ecosystems

Changes in temperature and moisture have a significant effect on the biotic community structure with feedbacks on ecosystem productivity. Alpine meadows respond with increased plant productivity to warming, while productivity may be hampered in alpine steppe ecosystems (Ganjurjav et al., 2016). As soil moisture governs the community response to warming, negative effects of warming on the plant productivity likely occur due to limited water availability (Ganjurjav et al., 2016). Warming was also reported to have a negative effect on plant species diversity in both alpine meadow and steppe ecosystems(Klein et al., 2008; Ganjurjav et al., 2016). Possible explanations for a decline in plant species diversity include changes in small mammal activity, storage of belowground nutrient resources as well as water stress and microclimate in general (soil temperature and moisture) (Ganjurjav et al., 2016; Klein et al., 2008, 2004). Thus, climate change may reduce the habitat quality for the local populations of grazers and reduce well-being of the pastoralists by diminishing abundance of palatable and medicinal plant groups. The changes in the plant productivity levels as well as community changes affect the local carbon cycle. Alpine grassland root mats on the TP are estimated to store up to 10 kg of carbon (C) per square-meter (Li et al., 2008b), summing up to roughly 2.5 % of the global terrestrial carbon stocks (Wang et al., 2002). At Nam Co, the top soils contain an almost 30 cm thick organic rich layer (Wang et al., 2009b), thus representing considerable soil organic carbon (SOC) stocks. Due to higher plant productivity, alpine meadows in general represent a CO_2 sink, however, the interannual and seasonal uptake is highly variable (Kato et al., 2004; Kato et al., 2006; Gu et al., 2003). Like plant productivity, the CO_2 uptake depends on water availability and temperature which exhibit a diurnal, seasonal, and annual fluctuation. The overall great importance of water availability and temperature on ecosystem-atmosphere CO_2 exchange in the central Tibetan alpine *Kobresia* meadows was demonstrated in several studies through Eddy Covariance measurements (Zhang et al., 2018), chamber measurements (Zhang et al., 2018; Zhao et al., 2017), decomposition of cellulose cotton strips (Ohtsuka et al., 2008) and altitudinal transplantation experiments (Zhao et al., 2018). Similarly, carbon fluxes in alpine steppe are driven by precipitation and temperature on a daily to seasonal and annual time scale. The inter-annual flux variability follows the varying monsoonal precipitation, showing stronger tendencies to function as a C sink in wetter years and as a source in drier years (Wang et al., 2018; Zhu et al., 2015b). Soils that develop in the drier steppe areas tend to show lower organic carbon contents, therefore lowering the total C sink and source potential (Ohtsuka et al., 2008). Although the production of plant biomass may be hampered in steppes, the ecosystem may still act as a carbon sink through microbial CO_2 fixing activities as shown by a recent study on the TP that reported relatively high CO_2 fixation capacity (29 mg kg^{-1} soil d^{-1}, Zhao et al., 2018). Interestingly, this study also found that alpine steppe soils demonstrated significantly higher microbial CO_2 fixation capacity compared to meadow soils (29 *vs*. 18 mg kg^{-1} soil d^{-1}, respectively).

As a result of increasing precipitation and glacier runoff, wetlands in the Nam Co area are expanding, thus increasing emissions of CH_4, which is 28 times more climate active than CO_2 (Intergovernmental Panel on

Climate Change, 2014). A study conducted in the alpine wetlands around Nam Co reported, that CH_4 emissions have increased exponentially with increasing precipitation, especially when soil moisture exceeded 80 % (Wei et al., 2015). However, there was a large difference between swamp meadows and swamps (67 and 1444 µg CH_4 m^{-2} h^{-1}, respectively). Swamps are permanently inundated, while swamp meadows are usually seasonally inundated. Furthermore, SOC stocks are higher in swamps compared to swamp meadows (Wei et al., 2015). Large amounts of SOC in combination with anoxic conditions are the main precursors for methanogens activity, which results in increasing CH_4 emissions to the atmosphere (Kato et al., 2013). Thus, the saturated soils with high SOC content produce higher CH_4 emissions (Deng et al., 2013). Observations from 2008 to 2013 at Nam Co have shown, that alpine steppe and alpine meadows show annual uptake rates of 72 and 59 µg CH_4 m^{-2} h^{-1}, respectively (Wei et al., 2015); however, the corresponding emission rates are much higher. Generally, it is expected that the alpine wetland acts as a CH_4 source while the aerated soils of alpine steppe and alpine meadow act mainly as a CH_4 sink.

As the grasslands on the TP are widely used for yak and sheep grazing, carbon cycling is influenced particularly through human activities and the degree of degradation. The intensity of grazing has a significant effect on CO_2 fluxes, with moderate grazing enhancing belowground allocation of carbon (Hafner et al., 2012), while adversely affecting the C-sink potential through reduction of above- and belowground biomass at higher grazing intensities (Babel et al., 2014). Overgrazing, along with the increase of burrowing pikas in the Tibetan grasslands may increase the Nitrous Oxide (N_2O) emissions (Zhou et al., 2018), an important greenhouse gas with 297-times larger warming potential compared to CO_2 (IPCC, 2013). Despite several studies focusing on greenhouse gas emissions on the TP, the magnitude of the N_2O emissions in different ecosystems has not yet been estimated. Experimental studies on the eastern TP demonstrated that the rate of N_2O emission may increase with increasing soil temperature and soil moisture under a future climate change scenario (Yan et al., 2018; Yingfang et al., 2018). Expanding wetland areas provide anoxic conditions for the release of methane and, due to the greater temperature sensitivity of permafrost areas, subsurface SOC is at high risk of loss, which may decrease the carbon sequestration potential in the region (Li et al., 2018). Besides carbon cycling through decomposition processes, responses to changing temperature and precipitation depend on the composition of decomposer communities (Glassman et al., 2018). Thus, the conclusive effects and feedback mechanisms (i.e. positive *vs.* negative loop) on warming are complex and not always clear.

3 Paleoenvironments on the Tibetan Plateau and landscape evolution at Nam Co

3.1 Geodiversity and evolution of biodiversity

Topography, geological context, climate and their complex interplay are key determinants for the distribution of organisms. In general, the ecoregion can serve as a proxy for community- and species-level biodiversity, which best describe communities of mammals, birds and plants (Smith et al., 2018). The TP forms a distinctive zoographical region, an “ecological island” (Deng et al., 2019), characterized by fauna that is adapted to high altitudes, drought, low temperatures and low oxygen levels (He et al., 2016). The TP is forming a unique high-altitude biogeographical biota by harboring also many unique lineages of other organisms, with higher endemism of low dispersal species (Yang et al., 2009; Clewing et al., 2016). As mountain building has been directly associated with the development of biodiversity (Hoorn et al., 2013; Antonelli et al., 2018), the biodiversity hotspots are located especially in the south and south-east of the TP. There is also a pattern of increasing biodiversity from west to east, which correlates positively with increasing precipitation. In contrast, the harsh central areas of the TP show much lower richness, but nevertheless harbor various endemics (Päckert et al., 2015). Throughout the geological formation of the TP, the mountainous south-eastern parts have been hypothesized to serve as center of species diversification (Mosbrugger et al., 2018), although the core TP region is also suggested to represent a center of origin (Deng et al., 2011). The TP has been a source area for several mammalian lineages (Out-of-Tibet hypothesis; Deng et al., 2011), including the snow leopard and the arctic fox (Wang et al., 2015), as well as birds, such as redstarts (Voelker et al., 2015), and plants, such as *Gentiana* (Favre et al., 2015). These mountainous areas may also have acted as refugia, which preserved unique lineages over long periods (López-Pujol et al., 2011; Lei et al., 2014). Whether some endemic taxa represent relics of a formerly more diverse clade or have never extensively diversified, remains unclear (Päckert et al., 2015). Besides being a center of origin, the TP may represent a center of accumulation as proposed by the examples of *Saxifraga* (Ebersbach et al., 2017), warblers (Johansson et al., 2007) and hynobiid salamander (Zhang et al., 2006). Overall, the regional biota of the TP is comprised mainly of Palearctic and Oriental species, Nearctic species from the Bering land bridge, as well as species from speciation *in situ*, and postglacial recolonization from adjacent areas. The evolution of biodiversity on the TP has been affected by the combination of geological and climatic changes over the time of the uplift phases (Mosbrugger et al., 2018). Although many studies have associated recent *in situ* radiations to different uplift phases of the TP, Renner (2016) pointed out that the evidence for recent rapid uplift (9–8 or 3.6–2.6 Ma) remains doubtful and controversial. As proposed by the "mountain-geobiodiversity" hypothesis, the evolution of biodiversity on the TP is a result of an increasing local geodiversity in combination with rapid climatic oscillations and steep ecological gradients (Mosbrugger et al., 2018).

The combination of geological, climatic and ecological changes has left its footprint in the history of Nam Co. There are at least seven different levels of continuous terraces around Nam Co, with the highest being over 30 meters above current lake level, corresponding well with the elevation of the natural spillway in the northeast of Nam Co. Several authors claim the existence of a much larger fluvial lake system called "Old Qiangtang Lake", which covered an area of about 30,000–50,000 km^2 or more (Li et al., 1981; Zhu et al., 2002). The connections provided by a large lake allowed the gene flow between drainages, which is reflected, for example, by the closely related clades of schizothoracine fish (Cyprinidae, Osteichthyes) from Nam Co and the surrounding lakes, compared with more distant parts of the TP (He et al., 2016). In contrast, due to a vector-mediated passive dispersal across large areas, other aquatic taxa, such as freshwater snails, seem to have been less influenced by drainage histories (Oheimb et al., 2011). Higher lake terraces are older, suggesting a long-term reduction in lake level (Zhu et al., 2002). This may be associated with an evolution from wet to dry phase, which Li et al. (1981) connects to the gradual uplift of the plateau from early Pleistocene to the Holocene. However, there is an alternative suggestion to this interpretation of a rather modern uplift proposed by Renner (2016) who states that large parts of the TP had already reached average heights of 4,000 m and more during the mid-Eocene (~40 Ma ago). Recent findings of palm leave fossils on the central part of the TP, dated to ca. 25.5 ± 0.5 million years, do not suggest a presence of such a high plateau before the Neogene (Su et al., 2019). Thus, although it is suggested that the final large lake phase took place during ca. 40-25 cal ka BP (Lehmkuhl et al., 2002; Zhu et al., 2002), the complex relationship between evolution of the TP and the development and the temporal existence of "Old Qiangtang Lake" are not completely resolved.

3.2 Holocoene lake level changes and climate reconstruction based on aquatic bioindicators

Lake sediments contain important indicators, or proxies, that can be used to reconstruct limnological and (hydro-) climatic conditions over long time periods (Zhu et al., 2010a; Wrozyna et al., 2010). Widely used environmental indicators include communities of diatoms (Bacillariophyceae) and ostracods (Crustacea: Ostracoda) as they are abundant and usually preserve well in sediments (Kasper et al., 2013). For example, the investigations of Quaternary ostracods, modern assemblages, and stable isotopes from Nam Co and nearby water bodies represent the most detailed application of ostracod analysis in the south-central region of the TP (Mischke, 2012). Different approaches (stratigraphy, paleoecology, etc.) detected several climatic fluctuations between warmer/cooler and wetter/drier periods (Fig. 5). In general, higher lake levels based on aquatic fauna suggest a more humid environment during the early and middle Holocene, which displayed a shift pattern compared to the northern TP (Wünnemann et al., 2018). Together with the indicator species approach, and the application of transfer functions for Nam Co sediments different stages can be recognized. Stage I (8.4–6.8 cal ka BP): climate changed from warm-humid to cold-arid with water depth being much lower than today (Zhu et al., 2010a). Stage II (6.8–2.9 cal ka BP): environmental conditions returned to warm and humid (Zhu et al., 2010a). During 4–2 cal ka BP, lake water depth initially remained much shallower than today but then gradually increased due to high rates of precipitation (Frenzel et al., 2010). The presence of the diatom taxa *Stephanodiscus* in this stage indicated stronger monsoon activity and higher availability of nutrients (Kasper et al., 2013). Finally, stage III (2.9 cal ka BP to present): the climate again became warm-humid, with a cold-dry event between 1.7 and 1.5 cal ka BP (Zhu et al., 2010a). Between 2 and 1.2 cal ka BP, benthic diatoms, inferred a lower water level and drier climate (Kasper et al., 2013). Subsequently, wetter conditions and an increase in lake level was detected (1.2 cal ka BP until 250 cal BP), possibly corresponding to the Medieval Warm Period (MWP), with high planktonic diatom species and high ostracods diversity (Kasper et al., 2013). During the late Holocene, the minimum water level occurred throughout the Little Ice Age (LIA) (~1490 and 1760 AD) (Frenzel et al., 2010). However, the lake level increased towards the present, which is plausibly linked to the melting of the glacier due to the current warming.

Although a large number of studies describe profound hydrological changes and general climate fluctuations, there are several uncertainties regarding taxonomy, resolution and proxy sensitivities. For example, modern ostracod data detects several morphological variations, characterized by different nodding or shell sizes, which could lead to an erroneous ecological interpretation and later, vague paleoenvironmental conclusions in relation to salinity changes (Fürstenberg et al., 2015). In paleo-studies, different sedimentation rates and uncertainties in the core chronologies also cause a lack of correspondence between signals detected by different proxies (Wang et al., 2012). For this reason, it is surrogate to understand the precise causal relationships between a complex environmental gradient (e.g. water depth, water chemistry, temperature, etc.) and the response of bioindicators. Although ecological information is still poorly known for many species, ostracod and diatom assemblages represent reliable proxies to trace the climatic history of Nam Co.

Further emphasis should be placed on combining morphology and DNA analysis to corroborate the classification of the species already described. Furthermore, experiments with living individuals should be performed under controlled environmental variables to allow the setup of a transfer function that could be used to evaluate quantitative data for paleoreconstructions.

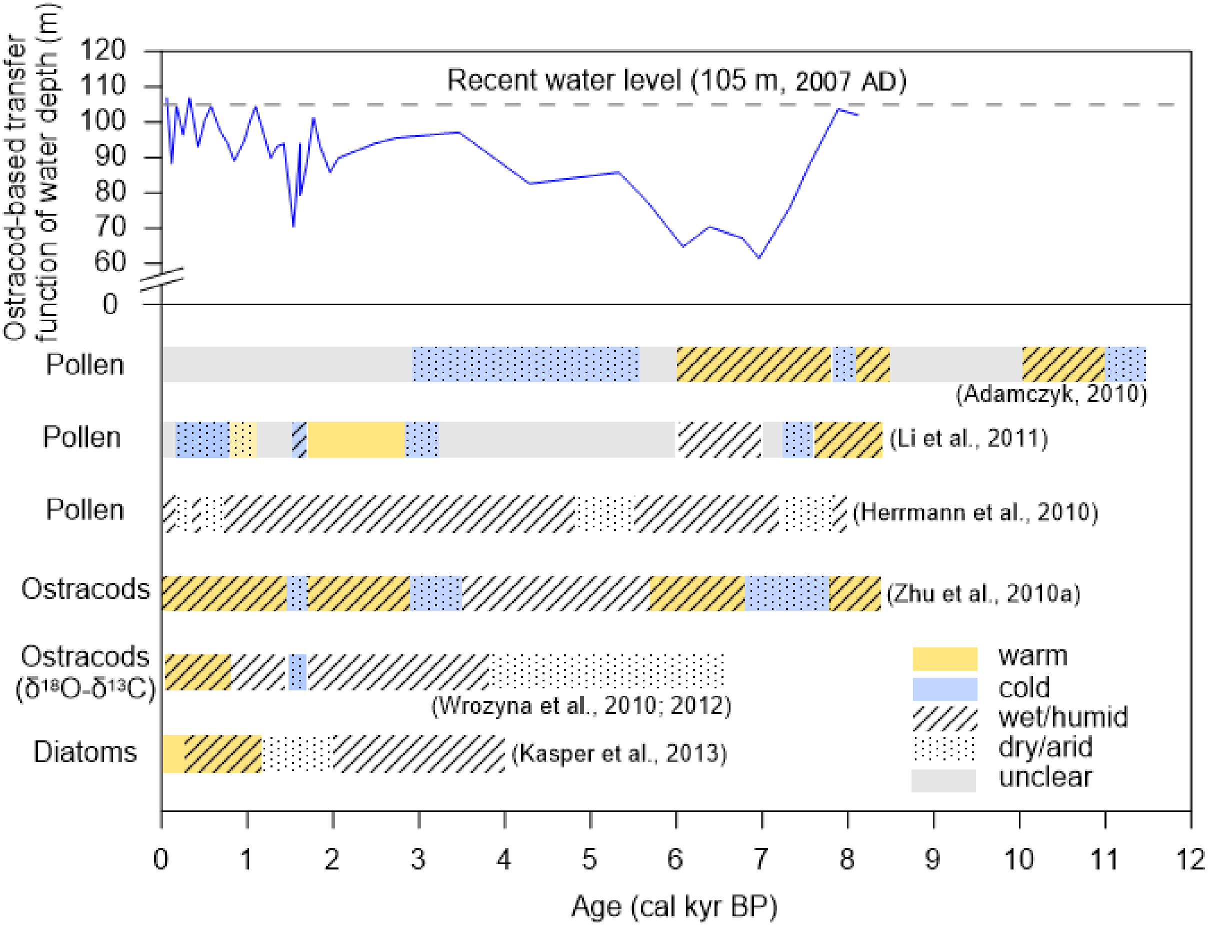

Figure 5. Comparisons of the reconstructed climate conditions based on fossils of pollen (Li et al., 2011; Adamczyk, 2010; Herrmann et al., 2010), Ostracods (Zhu et al., 2010a), Ostracod δ18O (Wrozyna et al., 2012; Wrozyna et al., 2010) and diatoms (Kasper et al., 2013) from sediment cores in, and at the shoreline of lake Nam Co. Ostracod-based water-depth transfer function (Zhu et al., 2010a); blue line) was used to indicate long-term hydrological changes and all reconstructed water-depth values were adjusted to the maximum water level of the lake according to the 45 m difference between this study site (60 m) and deepest site (105 m) of lake Nam Co. Main species also shown in different periods.

3.3 Holocene vegetation cover and climate reconstruction based on pollen records

The comparison of modern pollen assemblages with those from sediment cores allows the reconstruction of floristic diversity and distribution across various time scales. Vegetation patterns contribute to reconstruct past climate and also with assessing the degree of local human influence. Modern vegetation belts around Nam Co reveal that alpine steppe is containing mostly species of *Artemisia* (Asteraceae) and Poaceae, while alpine meadows and swamps are dominated by Cyperaceae (Li et al., 2011). The sedimentary pollen ratio of *Artemisia* to Cyperaceae (A/Cy) can, within certain limitations, be used to reconstruct past climates (Li et al., 2011; Li, 2018; Zhu et al., 2015a) provided that vegetation belts move with altitude during climate change. For example, when the climate is warmer and drier, alpine steppe reaches higher up the mountain, displacing alpine meadow into areas further away from the lake, leading to a higher input of *Artemisia* pollen into the nearby lake and consequently a higher A/Cy pollen ratio in the sediments. However, the A/Cy pollen ratio and abundance of tree-pollen originating from a short distance can be altered by human-driven change of plant composition, hence the beginning of pastoral economy might limit the explanatory power of pollen records (Adamczyk, 2010; Miehe et al., 2014). Pollen composition inferred from sediment cores reveals a downward shift of the altitudinal vegetation belts since 8.4 ka BP (Li et al., 2011). A major extension of alpine pasture and alpine sparse vegetation closer to the lake shore during the late Holocene is corroborated by a pollen-based climate reconstruction from a peat core near Nam Co (Herrmann et al., 2010) and two other pollen records from the eastern lake shore (Adamczyk, 2010). They found a trend of increasing temperatures from the late glacial until the early Holocene, accompanied by an extension of alpine steppe, tree and shrub vegetation. Already in this period, synanthrope taxa pollen are increasing in the data used by Adamczyk (2010) with the only small occurrence of e.g. *Plantago lanceolata* in the whole profile. This very early signal shows, that still much room exists for studies of pollen archives around Lake Nam Co with today much doubt persisting. Climate fluctuated between dry and humid from 8.5 to 4.8 ka BP, with an intense cold regression between 8.1to 7.8 ka BP. The onset of human activity at Lake Nam Co is dated to 5.6 ka BP according to synanthrope taxa proxies (Li et al., 2011; Herrmann et al., 2010). Between 4.8 and 0.7 ka BP, a relatively stable climate with predominantly humid conditions developed (Fig. 5), the vegetation pattern already showed trends of a human-made steppe biome, potentially a plagioclimax (Adamczyk, 2010). Since 0.7 ka BP, drier conditions prevailed.

Whether and to what extent the central Tibetan Plateau was forested, and what caused the forest decline is the subject of ongoing discussion (Miehe et al., 2006; Miehe et al., 2019). This matter is closely related to the prior discussed onset of more intense human activity in the area, since parts of the discussion involve a human-made forest clearing in combination with a natural forest decline. As stated, there are only occurrences of shrubs (*Juniperus pingii var. wilsonii* and *Salix* (Nölling, 2006)) in the Nam Co area. No remains and yet no reliable evidence of a once tree-rich vegetation can be found in the Nam Co catchment. According to locals, there exist several caves with potentially (pre-) historic tree depictions of unknown age. Unfortunately, there is no verification of their existence, nor any dating approach. Since the area of Damxung still does feature larger occurences of *Juniperus pingii var. wilsonii* and, around 4250 m a.s.l., also tree stands of *Juniperus tibetica* in enclosed areas, there is the potential to discuss, that these species have been more numerous in this area (i.e. last-tooth-theory). Miehe et al. (2019) show locations of forest relicts and give a drought line of 200-250 mm precipitation and elevations between 3600 and 4000 m a.s.l. as the upper tree line. Questions arise, whether there has been an expansion of *J. tibetica* into the Nam Co catchment in earlier times, which would be feasible within certain limitations according to the presented thresholds. Charred micro remains as a potential sign of fire driven forest decline, are missing in one of the profiles of Adamczyk (2010) but can be found throughout the Holocene until 1 cal ka BP (Herrmann et al., 2010). The authors attribute the size and shape of the charcoal remains to local, small-scale burning of wood and leaves, not showing signs of larger forest clearings. In addition to the burning of *Juniperus* trees for religious reasons (Miehe et al., 2006), trees and shrubs may have been burned for heating or clearing of pastures by nomads. Following the presumptuous argumentation of some authors, the trees were previously able to spread again due to sufficient precipitation provided by the summer monsoon. Furthermore, the occurrence of synanthropic taxa has been observed in the nearby Damxung valley since 8.5 cal ka BP, corroborating the strong anthropogenic influence on the formation and restructuring of the vegetation patterns in the area (Schlütz et al., 2007). The decrease of summer precipitation and temperature, in conjunction with ongoing human activity ultimately led to the total disappearance of trees and the formation of the alpine grasslands and steppe as we know them today (see section 2.4). Furthermore, the occurrence of synanthropic taxa has been observed in the nearby Damxung valley since 8.5 cal ka BP (Schlütz et al., 2007). This corroborates the strong anthropogenic influence on the formation and restructuring of vegetation patterns in the area but leaves a time gap of almost 3 ka between the evidence from Damxung valley and Nam Co. Hence, further research is needed to address the question of onset of human activity and degree of landscape modification.

4. Conclusions and perspectives

This literature review summarizes the manifold environmental changes affecting abiotic and biotic processes in the area caused by past and ongoing climate change. Ecosystems on the Tibetan Plateau experience an increase in air temperature roughly twice the global average. This has accelerated deglaciation of the Nyainqêntanglha range during the last decades, leading to substantial inflow of freshwater and various solutes resulting from weathering to the lake. The combined effects of overgrazing by livestock and warming accelerated degradation processes of the alpine grasslands further increase surface runoff in the catchment. Moreover, warmer and wetter climate as well as pasture degradation may turn alpine wetlands and steppe-pasture ecosystems into an overall source of methane and carbon dioxide, respectively. Based on the reviewed literature focusing on the catchment of Nam Co, we outline perspectives to improve the understanding of the close connections between geo- and biodiversity. (1) Permafrost areas act as buffers of the water budget, and influence the behavior of geomorphological processes and periglacial landforms. Although a significant warming and consequent decay of permafrost have been reported throughout the TP in recent decades, studies on permafrost in the Nam Co catchment and in the immediate Nyainqêntanglha range are missing. (2) The rising lake level trend, starting in late 1970, had a point of reflection around 2009, which indicates changes of variable precipitation and evaporation trends, reduced water inflow from already melted glaciers and additional ground water seepage out of the lake. Therefore, long term monitoring is necessary to calibrate and validate models properly, to achieve on a more accurate climate prognosis. (3) To improve climate modelling approaches, the dynamics of DOM, CO_2 and CH_4 fluxes need further clarification by in-depth analysis of the different biomes and in-situ observations. (4) The development of molecular methods for biomonitoring and water quality assessment has advanced greatly during last decade with the aim to provide clear monitoring standards. These offer time- and cost-effective approaches for complementary studies to tackle community shifts of various water quality indicator organisms. (5) Alongside the ‘traditional’ paleobioindicator analysis, DNA based taxa identification methods hold also a great potential for application in paleoecological studies to provide improved taxa differentiating accuracy. Various biological and geochemical proxies in Nam Co sediments have enabled the tracking of historical events and the reconstruction of past environments, which provide information about the magnitudes and directions of past climate change and thus a key to assess future changes. Both the formation of high elevation environments and pronounced past climate oscillations have contributed to the development of biota on the TP. Interdisciplinary research of the Nam Co catchment has provided vast insights into how warming trends may affect ecosystems from microbes to the top of the food chain. Recognizing the impacts of a warming climate is the base for establishing effective climate change adaptation strategies and actions in the TP region and in alpine regions in general.

Team list:

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Author contributions:

SA, MAR, JB, PEG, JK, WK, LK, PM, FN, ER, HT, TVT, YW contributed equally to the content of the Manuscript. AS conceived the idea and was responsible for funding acquisition. SA, JB, PM, FN structured the main text body; The following authors were in charge of the corresponding sections: JB, ER, TVT: *Glacier retreat and hydrological patterns of Nam Co*; PEG, LK: *Enhanced water availability controls changes in lake water chemistry*; PM, FN: *Vegetation, soils and pasture degradation in the catchment* & *Effects on carbon cycling in alpine ecosystems* & *Holocene vegetation cover and climate reconstruction based on pollen records*; SA, PEG, WK: *Geodiversity and evolution of biodiversity* & *Paleo-lake level changes and climate reconstruction*; The tables and figures were prepared as follows: ER: Fig. 1, JB and FN: Tab.1, Tab. 2, Fig. 2 and Fig. 3, PM: Fig. 4, WK: Fig. 5.

Competing interests:

The authors declare that they have no conflict of interest.

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Data availability:

As this manuscript is reviewing existing literature findings, there was no data analyzed that is not already published in the studies we cite. The dataset from which Table 1 was generated was provided by the Institute of Tibetan Plateau Research and is publicly available under https://data.tpdc.ac.cn/en/data/4deeb2b4-4fc1-4c7c-b0c6-6263a547d53f/ (Wang and Wu, 2018) and https://data.tpdc.ac.cn/en/data/3767cacc-96e3-48b2-b66c-dac92800ca69/ (Wang, 2019)

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Study II

The glacial – terrestrial – fluvial pathway: A multiparametrical analysis of spatiotemporal dissolved organic matter variation in three catchments of Lake Nam Co, Tibetan Plateau

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Highlights

- **First multiparametrical study of DOM source signatures from multiple catchments for a large central Tibetan lake watershed**
- **Lake and stream water DOM are chemically distinct with respect to their terrestrial DOM sources and impact of seasonality**
- **There is an ample impact of catchment properties on stream DOM, indicating different DOM sources**
- **The Indian summer monsoon greatly modifies stream DOM composition**
- **The DOM degradation cascade model and river continuum concept functionally represent catchment and lake biogeochemistry**

Abstract

The Tibetan Plateau (TP) is a sensitive alpine environment of global importance, being Asia's water tower, featuring vast ice masses and comprising the world's largest alpine grassland. It is under pressure by multiplied threats due to intensified land-use and pronounced global climate change. This study presents a multiparametrical approach, based on inorganic water chemistry parameters, dissolved organic carbon (DOC) concentrations, dissolved organic matter (DOM) characteristics (chromophoric DOM, PARAFAC-resolved fluorophore DOM and $\delta^{13}C$ of DOM), in river samples of three catchments of the Nam Co watershed and the lake itself. We aimed at testing, whether DOM has site and seasonal specific characteristics, making it a potential indicator for environmental change. Satellite based plant cover estimates were used to link the biogeochemical data to structure and degradation of vegetation zones in the catchments. Catchment streams showed site-specific DOM signatures inherited from glaciers, wetlands, groundwater, and pastures dominated by *Kobresia pygmaea*. Comparing stream and lake samples, we found processing and unification by vanishing of site-specific signatures. These findings are in line with the River Continuum Concept (RCC), stating that DOM diversity is largest in the headwaters of the catchments and heavily modified in terminal aquatic systems. Seasonality was characterized by minor influence of freshet but a very strong impact of the Indian summer monsoon on DOM composition, with more microbial DOM sources. The DOM of Lake Nam Co was chemically distinct from river water samples, with the lake being comparable to a quasi-marine environment concerning the degree of chemical modification and sources of DOM. Our findings showed good applicability of the RCC and the DDC to explain the fate of DOM in the Nam Co watershed, proving functionality of these concepts in the High Asian context. With this, we can better assess how human-induced changes will alter alpine ecosystems' processes and functions.

Keywords

Alpine pastures, dissolved organic carbon, PARAFAC, fluorescence, Tibetan Plateau, third pole environment

1 Introduction

Dissolved organic matter (DOM) is a manifold of organic substances ≤0.45 µm. Despite of its often low concentrations in surface waters, it is an important nutrient and energy source in aquatic and terrestrial ecosystems (Wymore et al., 2016). DOM participates in the global carbon cycle (Roulet and Moore, 2006), and much of an ecosystems carbon losses can take place via export of these diverse organic compounds (Lau, 2021). The often observed huge variability in dissolved organic carbon (DOC) concentration and DOM composition in surface waters reflects characteristics of and processes within the respective catchment (Jaffé et al., 2012; Kawahigashi et al., 2004; Singer et al., 2012).

DOM bridges glacial, terrestrial, lotic (riverine) and lacustrine ecosystems, and shows pronounced effects of seasonality in DOM composition and mobility. Further, DOM serves not only as indicator of environmental change, such as acidification (Han et al., 2022; Guggenberger, 1994), change of the soil thermal regime (Kawahigashi et al., 2004), and eutrophication (Görs et al., 2007), but also governs changes of biodiversity (Zhao et al., 2019), e.g. by delivery of large quantities of highly biodegradable compounds to an oligotrophic estuary (Görs et al., 2007).

The process of DOM formation largely dependents on the respective catchment area and therefore also inherits the biogeodiversity of catchments (Coch et al., 2019; Lafrenière and Sharp, 2004) as well as climatic processes (Song et al., 2020) and the impact of seasonality (Jennings et al., 2020). DOM is being processed and chemically altered during its fluvial pathway in streams (Riedel et al., 2016) and lakes (Massicotte and Frenette, 2013), thereby changing its signature and finally forming terminal, globally recurrent characteristics (Riedel et al., 2016). In this regard, Vannote et al. (1980), developed the river continuum concept (RCC) as a heuristic framework characterising the changes of stream morphology within the river course as a function of changing dynamics in water physics and biology. Later, Mosher et al. (2015) and Roebuck et al. (2020) have expanded this framework on the variability of DOM composition within the river course, stating that DOM diversity peaks in first order streams and then decreases with stream order (Mosher et al., 2015). In an extension of the RCC, Zark and Dittmar (2018) argue, that universal processes of DOM degradation in rivers and estuaries lead to successive losses of terrigenous DOM signatures and finally to a globally successive molecular conformity of DOM, as a result of a DOM degradation cascade (DDC). These concepts present a functional framework, allowing to predict influences on DOM in diverse watersheds and further to forecast the changes and processing of DOM in natural ecosystems.

The Tibetan Plateau (TP) is of global importance in multiple scales and processes. The TP comprises the largest ice mass outside of the Polar Regions (third pole environment) (Qiu, 2008; Yao et al., 2012). With a vast amount of freshwater resources stored in the frozen environment (Ding et al., 2006), it qualifies as a glacial hotspot. Studies underline the decisive role of High Asian watersheds for Asian lowlands and even for marine environments in South Asia (Xu et al., 2021). Goes et al. (2020) showed that ecological changes on the TP can prompt responses of marine environments and coastal shelves several thousand kilometres away of the actual source area. The TP also features the largest alpine grassland of the world (Miehe et al., 2019) and huge alpine wetlands (Zhang et al., 2020) with an active pastoral community (Gongbuzeren et al., 2018). The sensitive high-altitude ecosystems of the TP are under pressure by multiple threats such as intensified land-use and pronounced global warming (Harris, 2010). To study the biogeochemistry inside the TP is an essential component in elucidating the understanding of present day's worldwide challenges. Grassland degradation is one major threat for the Tibetan highlands (Harris, 2010). However, implications on DOM formation, quality, and fate are seldom addressed in studies dealing with the effects of grassland degradation (Anslan et al., 2020). But DOM has already proven a powerful marker for terrestrial degradation processes, and having a high potential for eutrophication of downstream ecosystems (Coch et al., 2019; Jennings et al., 2020; Lafrenière and Sharp, 2004). Via this marker function, DOM might help to upscale plot based investigations of grassland degradation to landscape level (Speetjens et al., 2020). In addition, alpine wetlands are considered the least studied terrestrial biome on the TP (Anslan et al., 2020), but degradation of wetlands is already underway (Zhang et al., 2020). This process can lead to the release of considerable amounts of high chromophoric DOM leading to several threats, e.g. to increased water temperatures (Gao, 2016) or hamper the larval growth of fish in lakes, due to reduced light penetration (Kritzberg et al., 2020). Hence, wetland degradation bears additional risks, but is easily monitorable via the DOM marker function as suggested by Li et al. (2021).

On the temporal scale, the rapid temperature rise on the TP (Hopping et al., 2018; Song et al., 2020) has led to massive changes especially in glaciated catchments (Bolch et al., 2010). Glaciers possess unique DOM signatures (Boix Canadell et al., 2019; Hood et al., 2009) with accelerating climate-driven export of DOM into downstream water bodies. Still, there is only limited knowledge on how the current glacier melting is influencing DOM and its impact on downstream catchments for High Asia. However, first works show that glacial DOM on the QTP can be more bioavailable than initially suspected (Spencer et al., 2014). Positive changes in precipitation (Zhang et al., 2017; Zhang et al., 2019) and increased water availability in conjunction with pronounced warming trends of 0.16 °C per decade (Yao et al., 2019), are further described to lead to earlier green-up dates of the alpine steppe and alpine pastures and a greening of the QTP (Zhong et al., 2019). It is unknown in detail, how this might affect the ecosystem, but Hopping et al. (2018) see ecosystem services at Nam Co on the QTP largely threatened. Spatiotemporal changes in the DOM composition of terminal aquatic systems, such as endorheic lakes, however can be utilised to monitor biogeoecological processes in glacial, terrestrial and aquatic ecosystems.

On the TP, endorheic basins provide excellent opportunities to use catchment-based studies to investigate into ecosystem processes as influenced by environmental change. Here, we focus on the oligotrophic, endorheic Lake Nam Co. This lake receives DOM input from its diverse watershed but also forms the terminus of potential DOM degradation cascade by loss of terrigenous DOM properties, as suggested by initial work of Li et al. (2021). The Nam Co watershed is spatially very diverse, and comprises glaciers and periglacial landforms, alpine steppe, alpine wetlands and *Kobresia pygmaea* alpine pasture/meadow biomes in different degree of plant cover and land degradation. Alpine pastures are predominantly developed in the mid and higher slopes as well as in depressions, and alpine steppe is mostly developed in the arid lake foreland. The streams of the catchment are fed by glacial melt, precipitation (Adnan et al., 2019) and springs of groundwater aquifers (Tran et al., 2021), depending on catchment properties, such as aspect and height. With that, the Lake Nam Co watershed is a perfectly suited natural laboratory to study controlling factors of the spatiotemporal variability of riverine DOM in this sensitive high mountain area.

Our objective was to identify the impact of small-scale catchment properties, referred to as endmembers and season on the origin and composition of riverine DOM. By testing mechanistic models of DOM dynamics (River Continuum Concept: RCC and DOM Degradation Cascade: DDC), we aimed at elucidating the mechanisms controlling DOM processing in the fluvial pathways to Nam Co Lake. We tested whether DOM characteristics are connected to landscape units in the catchments, with fundamental differences between alpine steppe, alpine pastures and alpine wetland riverine DOM. We particularly assumed that DOM composition is largely determined by catchment hydrology and the degree of green plant cover and that unique signatures of glacial meltwater, precipitation-fed catchments and groundwater fed areas can be discriminated. Further, we assessed the impact of the very pronounced seasonality on the TP on DOM composition. We hypothesized that 1) catchment biogeodiversity in the Nam Co watershed governs DOM and water chemistry. We expected that processing of DOM during the fluvial pathway undergoes a DOM degradation cascade, with chromophoric DOM and readily bioavailable compounds preferentially altered during the pathway. Consequently, we further hypothesized that 2) the endorheic Nam Co Lake forms the terminus of a DOM degradation cascade leading to transformed, and seasonally stable DOM signatures. Finally, we expected that 3) the processes of DOM transformation within the Nam Co watershed fits to the RCC and DDC theoretical frameworks.

We approached this by establishment of a DOM multiparameter approach employed for three catchments as well as the Lake Nam Co on the central TP. Multiparameter datasets are especially suited to understand complex environments (Kayler et al. 2019), in our case between the glacial, terrestrial, lotic (riverine) and lacustrine ecosystem of the Nam Co watershed. To link DOM characteristics to the vegetation and vegetation changes, we conducted a satellite-based (Landsat type) watershed-wide estimation of plant cover in 30 meter resolution for an observation period of 30 years (1990-2020).

2 Materials and Methods

2.1 Study area

The oligotrophic, endorheic Lake Nam Co (4726 m asl.), has a total size of more than 2000 km² and the whole watershed comprises 10789 km^2 (Figure 1). The Nyainqêntanglha mountain ridge with elevations up to 7000 m asl. forms the south south-eastern border of the Nam Co watershed. It comprises a well investigated glacial environment (Bolch et al., 2010; Buckel et al., 2020; Yu et al., 2021). The north of the watershed in contrary, is dominated by hilly uplands (Yu et al., 2021). Sparse alpine vegetation occurs in the glacial foreland at elevations > 5350 m asl, while beneath *Kobresia pygmaea (K. pygmaea)* pastures dominate until 4900 m asl.. At lower elevations the ecotone between *K. pygmaea* pastures and the alpine steppe biome stretches. The alpine steppe is fully developed in the lowlands of the lake shore and on lake terraces (Miehe et al., 2019). Alpine wetlands are developed in depressions (Anslan et al., 2020), and often formed from riverine influence. Degradation and aridity are visible along the south-north gradient of the watershed (Anslan et al., 2020), increasing towards the north.

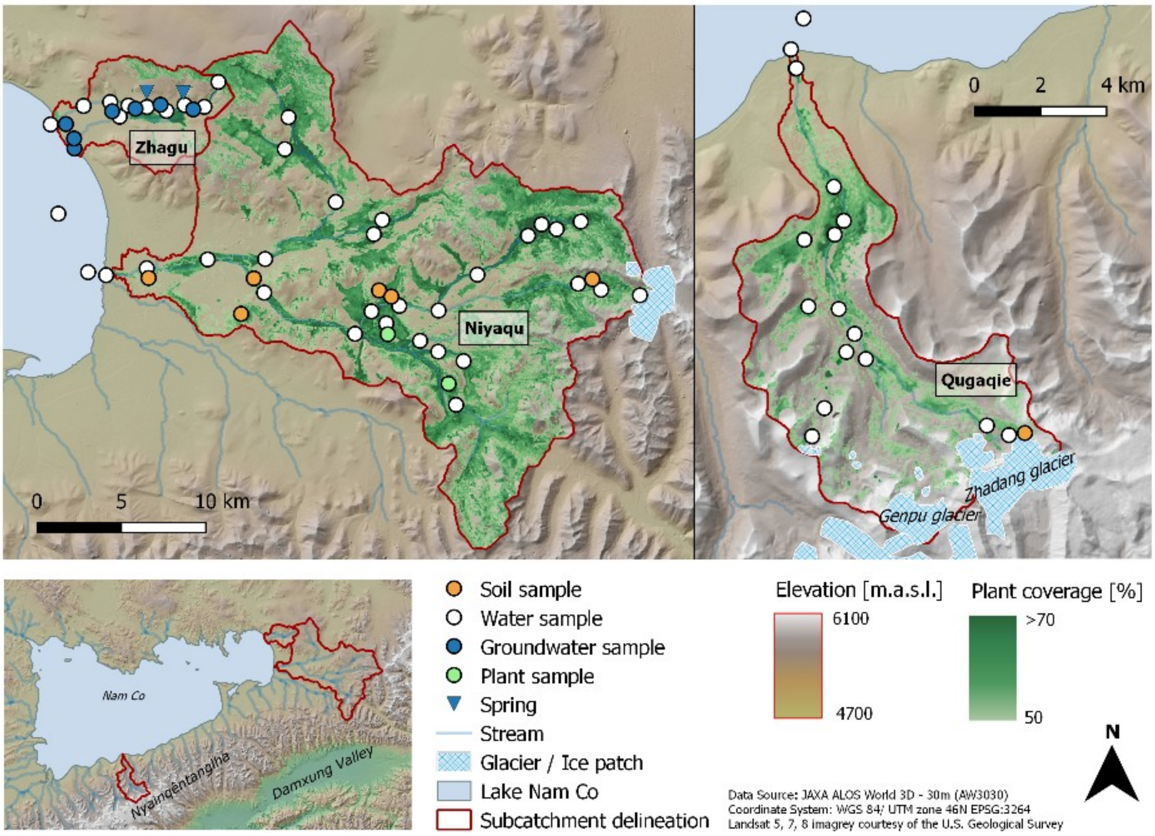

Figure 1: Overview of the investigated catchments including positions of sampling sites.

Mean annual temperature at Lake Nam Co is 0.6°C and mean annual precipitation amounts to 405 mm at the southern lake shore of the watershed compared to 300 mm at the watersheds northern margin (Anslan et al., 2020). Lake Nam Co's climate is biannually shaped. The winters are cold and dry with air temperature minima below -20°C between December and February. During winter, the Westerlies shift cold and dry air masses across the TP, which leads to only sparse precipitation events with an usual absence of a closed snow cover (Dorji et al., 2013). The Indian summer monsoon (ISM) has its usual onset around May to June and moves wet, moist and warm air masses from the Indian subcontinent towards the inner TP. 80% of the annual precipitation is falling during the monsoon months (until September), and the mean day-time air temperature in the monsoon season reaches 11°C (Chen et al., 2019). The ISM is the most influential hydrological and temperature control factor in our study area (Nieberding et al., 2021). Within the Nam Co watershed, three catchments were identified based to represent well the degree of plant cover, status of degradation, different landscape units, elevations and extent of glaciation of the whole Nam Co watershed. The study objects included the catchments of the rivers Niyaqu, east of Lake Nam Co, Zhagu north-east of the lake, and Qugaqie, south-west of Nam Co (Figure 1). Our dataset consists of samples obtained from three high Strahler number rivers and 12 lower Strahler number streams of these catchments and from the lake.

The Niyaqu catchment has the largest spatial extent with 406 km². A major river with two river arm systems drains the catchment. The southern one comprises glacial zones in the eastern outcrops of the Nyainqêntanglha and runs through an area of extended alpine pastures (Figure 2a) and alpine wetlands (Figure 2c). The northern river arm system is determined by the absence of glacial meltwater and a hilly upland relief with a transition between the alpine pasture and alpine steppe. The Niyaqu catchment is used for yak grazing throughout the year. Its valley course is mostly in west-east direction with a high altitudinal gradient between the lake shore and the eastern branch of Nyainqêntanglha (peaking at 5680 m asl. within Niyaqu catchment).

The Zhagu catchment is the smallest catchment under investigation (46 km²) and possesses only small altitudinal differences (highest point = 5230 m asl.). Two river systems drain the catchment to Lake Nam Co (Figure 2; B), but close to the Lake their stream beds were found empty during the periods of investigation. Apparently, episodic drainage events into the lake take place. The stream water is sourced by wells and precipitation (Tran et al., 2021). The catchment shows definite signs of degradation of the alpine pasture biome, by existence of barren soil and remaining isolated patches of alpine pasture turfs (so called pancake-land; Miehe et al., 2019). Animal husbandry is concentrated at the stream beds and at the flanks of hills.

The Qugaqie catchment comprises an area of 58 km². It is situated at the flank of the Nyainqêntanglha, which reaches elevations of up to 7000 m asl. Hence, this catchment exhibits the largest altitudinal gradient of our three study areas. One major stream drains the meltwater of both, the Genpu and Zhadang glaciers. The steep relief of the Qugaqie catchment provides a section through all vegetation zones. Sparsely vegetated glacial foreland defines large parts of the catchment at higher elevations (Figure 2; d). Alpine wetlands are developed in depressions or dammed by moraine structures. The dominant vegetation type is *K. pygmaea* pasture, which is grazed by yaks in the summer.

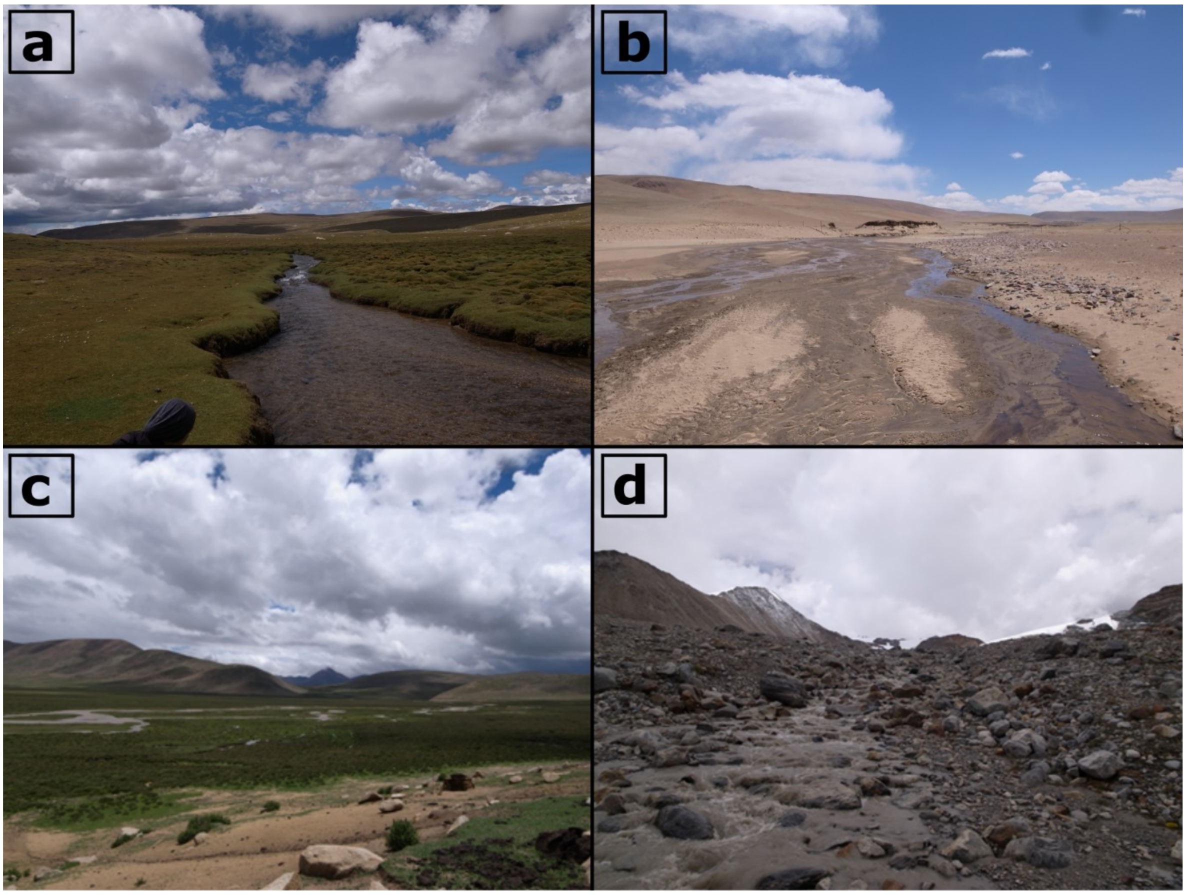

Figure 2: Landscape units of the sampling area. a: *Kobresia pygmaea* turfs in the Niyaqu catchment. b: Periodical stream in the Zhagu catchment alpine steppe. c: Alpine wetland in depression of the Niyaqu catchment, in the foreground degraded, south-exposed hillslope. d: Glacial foreland of Zhadang glacier, Qugaqie catchment.

2.2 Water sampling

Samplings of river and lake waters were conducted in three field campaigns in June/July 2018, May 2019, and September 2019, to cover seasonal differences. No sampling was possible in May 2019 in the Qugaqie catchment due to limited access. The sampling scheme was designed to cover the influence of potential endmembers, i.e.: landscape units, such as glacial effluents, alpine vegetation zones, wetlands and groundwater wells. Further samples were taken with the course of the streams to cover longitudinal shifts of DOM characteristics from the source until the river mouth (Figure 2). Additionally, lake samples were taken. Individual sample IDs and endmember affiliations of sampling points are depicted in the supplementary material (Figure S1).

River and lake water Samples were taken as mixed samples (1L was used from a 7L mixed sample) from the middle depth of the pelagial in a central position of the stream in polyethylene bottles (HDPE). Lake samples close to the shore were collected as mixed samples in the middle of the stream mouth from the pelagial, in 1 m depth and off-shore lake samples were taken about 200 m off shore from the studied catchments, as mixed samples at the surface and in 30 m depth using a submersible sampler. In addition, groundwater samples, used for δ ^{13}C of DOM, were obtained in May 2019 from piezometers of the Zhagu catchment as described by Tran et al. (2021).

Prior to sampling, bottles were rinsed with 10% HCl, washed with ultrapure water and dried. After sampling 350 µg L^{-1} $HgCl_2$ was added to the samples to inhibit biological degradation, following instructions published by VDLUFA (2012). All samples were frozen and kept at -21°C until analysis.

2.3 Sample analyses

Water samples were filtered using a 0.45µm polyethersulfone (PES) membrane (Supor, Pall, Port Washington, USA). pH was measured at 20°C using a potentiometric glass-electrode, calibrated with three standard solutions at pH 4.01, 7.00 and 10.01 (DIN 19266:2015-05), and electrical conductivity (EC) was determined using a conductivity sensor at 20°C, controlled with a standard solution (0.01 mol^{-1} KCl) at 1278 µS cm^{-1} (DIN EN 27888:1993-11). The concentration of the cations Na^+, Mg^{2+}, Ca^{2+}, K^+, Li^+, and NH_4^+ and the anions Cl^-, SO_4^{2-}, PO_4^{3-}, NO_2-, NO_3^-, F^-, Br^-, as well as oxalate were analysed by ion chromatography (Metrohm 930 Compact IC Flex, Herisau, Switzerland).

DOC concentrations were determined by high-temperature oxidation on 20 mL aqueous samples, acidified with 50 µL of 32% HCl on a total organic analyser (varioTOC Cube, Elementar GmbH, Langenselbold, Germany). Total dissolved carbon (TDC) samples were treated similarly, but no acid was added, and DIC was calculated as difference of TDC and DOC.

The $\delta^{13}C$ values of DOM samples from streams, the Nam Co Lake, and of groundwater samples (were measured in solution after acidification with HCl (32%) to pH 2 on an isoTOC cube (Elementar, Langenselbold, Germany) coupled with a continuous flow isotope ratio mass spectrometer (Elementar, Langenselbold, Germany; Federherr et al., 2014; Kirkels et al., 2014). Sample $\delta^{13}C$ signature was calculated as follows:

$\delta^{13}C_{sample}$	$\delta^{13}C$ of the sample [‰]
$\delta\,^{13}C_{mix}$	$\delta^{13}C$ of the mixture [‰]
$\delta\,^{13}C_{ref}$	$\delta^{13}C$ of the added reference standard [‰]
m_{ref}	C mass in the added reference standard [mg]
m_{sample}	C mass in the sample [mg]

$$\delta 13\, C_{sample} = \frac{\delta 13\, C_{mix} - \left(\delta 13 \frac{Cref * m_{ref}}{1000}\right)}{\frac{m_{sample}}{1000}}$$; with

In addition to the $\delta^{13}C$ of DOM, $\delta^{13}C$ was measured also in potential source materials of the riverine DOM. This includes three glacial sediment samples (Zhadang glacier) 15 topsoil samples (i.e. A horizons; IUSS Working Group WRB, 2015)), four root samples of the dominating terrestrial pasture plant *K. pygmaea* and four samples of a fully submerged plant of the genus *Elodea* from an alpine wetland. All samples were dried at 40°C until constant weight was reached. Samples were sieved to < 2 mm equivalent diameter and milled. Inorganic carbon in samples was estimated using a manometric method (van Lierop and MacKenzie, 1974). Inorganic carbonate was destroyed using the volatilization method following the descriptions of Hedges and Stern (1984) and Harris et al. (2001). $\delta^{13}C$ of the organic carbon in the samples was then determined using a isoprime cube elemental analyzer (Elementar, Langenselbold, Germany) coupled with an isoprime 100 isotope ratio mass spectrometer (Elementar, Langenselbold, Germany).

Ultraviolet absorbance properties of aqueous samples were determined using a Spectro Star Nano device (BMG Labtech, Ortenberg, Germany) with a 1mL Suprasil® quartz-glass cuvette in temperature-controlled conditions (20°C), scans were blank corrected. The UV/VIS absorbance at 254nm ($SUVA_{254}$) was normalised by the DOC content following Weishaar et al. (2003). Absolute absorbances at 254nm (A254) were taken uncorrected.

Fluorescence spectroscopy was measured using an Edinburgh F920 spectrometer (Edinburgh Instruments, Livingston, UK). Measurements were conducted in temperature-controlled conditions (20°C), in 90°mode in a QX class quartz Suprasil® 300 cell with a path length of 10mm. Excitation modes were set to 240-450nm with a 5nm step width and emission were set between 250-600nm with 2nm step width. Dwell time was defined as 0.1sec and slit width (Δλ) was set to 10nm in both the excitation and emission mode. An ultra-pure water Raman standard was used to avoid hysteresis effects.

Technically, fluorescence measurements as a set of excitation emission matrices (EEMs) data can be understood as a tensor-decomposition problem with an unknown number of fluorophores but following Beer's law. To retrieve information of single fluorophores in multiple EEMs, decomposition of tensors is done with the higher array principal component analysis (PCA)-like

PARAFAC algorithm. Unlike in PCA, real chemistry results, or components, can be obtained by PARAFAC, because of a non-rotational decomposition in the ordination space (Bro, 1997). Through the PARAFAC algorithm, fluorescence measurements became more unambiguously quantifiable and interpretability is improved (Fellman et al., 2010). Fluorescence spectroscopy is a valuable tool to determine the origin and potential fate of fluorescence DOM (FDOM) in biogeochemistry (Coble, 1996). FDOM represents the integral fraction of DOM that fluoresces. PARAFAC decomposition of fluorescence was conducted for EEMs with excitation > 280nm and with emission > 350nm; our data contained no information at lower wavelengths compared to blank samples. EEMs were corrected for a blank process sample to remove the water Raman signal, by removing 95% of the blank sample EEM from each sample. PARAFAC was run using the N-way toolbox (Andersson and Bro, 2000) in MATLAB R2017b (The MathWorks, Natick, USA). The dataset was found to be very diverse with large differences between sampling points. To prevent confounding the PARAFAC algorithm due to concentration induced mode shifts, we conducted PARAFAC in a twofold approach. First, a 4 component PARAFAC model was estimated and factor scores were summed for each sample. A threshold was determined to split the dataset into two more homogeneous datasets. The threshold was set at $5*10^5$ relative concentration, splitting the data in almost equally sized groups. Second, PARAFAC was then run for each group, i.e. low fluorescence response (below the threshold) and high fluorescence response (above the threshold). The Qugaqie samples were exempt from statistical analysis in the high fluorescence response dataset, because of small sample size (n=4) herein. Sample group affiliation is depicted in the supplementary material (Figure S2-S4). Residual mean EEMs of both models show mostly noise (supplementary material Figure S2/S3), pointing to a high quality modelling result (Stedmon and Bro, 2008).

2.4 Plant coverage estimates

Plant cover estimates (PCE) were calculated using pre-trained support vector machine regression (SVE) models published in Lehnert et al. (2015), which are fully validated against field samplings from the Tibetan Plateau (RMSE < 10 %). The models were applied to 30 years of multispectral satellite data acquired by Landsat 5, 7, and 8 to receive plant cover maps for the whole Nam Co watershed with 30m spatial resolution. Landsat scenes were downloaded from the United States Geological Survey website (http://earthexplorer.usgs.gov) and processed using an extended version of the 6S-code for atmospheric corrections (Curatola Fernández et al., 2015; Vermote et al., 1997) and the Minnaert model for topographic corrections (Riano et al., 2003). Obstructions (e.g.: clouds) were removed using the quality bands of the Landsat data. In total, 24 watershed-wide acquisitions were available for the plant cover assessment. Pixel-wise mean and standard deviations (SD) of plant coverage were calculated for the investigation period. The final product is in good conformity with the actual plant cover of our study area and was cross-verified by field investigation (Maurischat et al., unpublished). Non-parametric Mann-Kendall’s correlation tests were applied to those pixels where at least 7 valid PCEs were available (significance level of

$\alpha=0.05$). These tests were applied to detect changes of plant cover over time (decreasing or increasing). The term greening is used in case of a significant increase of plant cover. For the processing of the satellite data including the plant coverage estimates, R (The R project for statistical computing, v3.6.3, GNU free software) was used.

Differences in the mean plant cover of the investigated catchments Niyaqu, Qugaqie, and Zhagu were compared by the non-parametric Conover-Iman test (Conover, 1999) using the package conover.test (Dinno, 2017) with significance level of $\alpha=0.05$. Bonferroni post-hoc correction was applied using the R-base package (R Core Team, 2013).

2.5 Statistics

The dataset was grouped into three main effects: endmembers, site (i.e. location) and sampling seasons. Non-parametric tests and comparisons between and within the main effect groups site and season were conducted, no tests were conducted for endmembers, because of partly small sample sizes, here means were compared. When prerequisites for parametric tests were violated, we conduced non-parametric tests, the Mann-Whitney-U test (Birnbaum, 1956) for pairwise comparisons and Conover-Iman test (Conover, 1999) for multiple pairwise comparisons of repeated measurements. Bonferroni post-hoc correction was applied in the latter case. Significant effects were accepted on the level of $\alpha<0.05$. Multiparameter subsets were created by connecting the DOM and water chemistry dataset with the $\delta^{13}C$ of DOM measurements and the FDOM components to test the two independent main effect variables location and sampling season as well as potential endmembers as further explanatory variables. Multiple bivariate regressions and non-metric multidimensional scaling (NMDS) were performed for the multiparameter dataset. As proposed by Faith et al. (1987) and Anderson et al. (2006), dissimilarities in variable abundances can be used to compute the ecological distance or β-diversity between sampling sites. Here we apply the same distance approach to calculate the biogeochemical distance between sampling sites, endmembers and seasons. NMDS was performed on mean-centred and scaled data (Jolliffe, 2002), using the vegan package (Oksanen et al., 2020). For the first subset, including the high fluorescence response PARAFAC data, the Euclidean dissimilarity index and k=3 is used. The Manhattan index and k=3 is used for the low fluorescence response PARAFAC components and corresponding dataset, forming the second subset. Tables of scores, coefficients of determination (R^2) (Table S3-S4) and of loadings (Table S5) are provided in the supplementary material.

Statistics were computed using R language (The R project for statistical computing, v3.6.3, GNU free software), its base packages (R Core Team, 2013), the conceptual ‘tidyverse’ meta-package, as described by Wickham et al. (2019) and ‘Conover.test’ (Dinno, 2017) for the Conover-Iman test.

3 Results

3.1 Plant coverage estimates around Lake Nam Co

The mean plant cover in the Nam Co watershed area was 48.1 % for the composite of the investigated period, and within the tree catchments under investigation, PCE decreased in the order Niyaqu catchment (52.3%) > Qugaqie catchment (49.5 %) > Zhagu catchment (44.5 %) (Supplementary material Table S1). The vegetation coverage further showed an altitudinal and expositional dependent distribution with higher plant coverage in the south-west and east of the Nam Co watershed compared to the north and north-west (Figure 3a). Plant cover was connected with water availability and increased insulation (north exposition) during the dry season. Especially the north exposed valleys of the Nyainqentanglha showed an overall higher plant coverage driven by the glacial-melt driven water surplus (Song et al., 2014). The western catchments were characterized by high plant cover along streams and in depressions, while the south-exposed slopes in the northern Nam Co watershed exhibited low plant cover of 35 to 45%. The satellite based PCE data fit well to field surveys in the Qugaqie and Niyaqu catchment, which revealed the existence of extended wetlands with mires and submerged plants (*Elodea*) and of vast *K. pygmaea* pastures close to streams and on well insulated hillslopes mostly with an east-west valley course (Maurischat et al., unpublished). Alpine pastures are partly an azonal vegetation type in our study area, because *K. pygmaea* seamed the river banks almost until the lake.

During the 30-years investigation period, only a small number of pixels reveal a significant decrease in plant cover, whereas in large areas a pronounced greening can be observed (Figure 3). Especially the southern shoreline shows large areas of significant greening between 1990 and 2020, and also the eastern and north-eastern areas of the Nam Co watershed exhibit a significant greening trend.

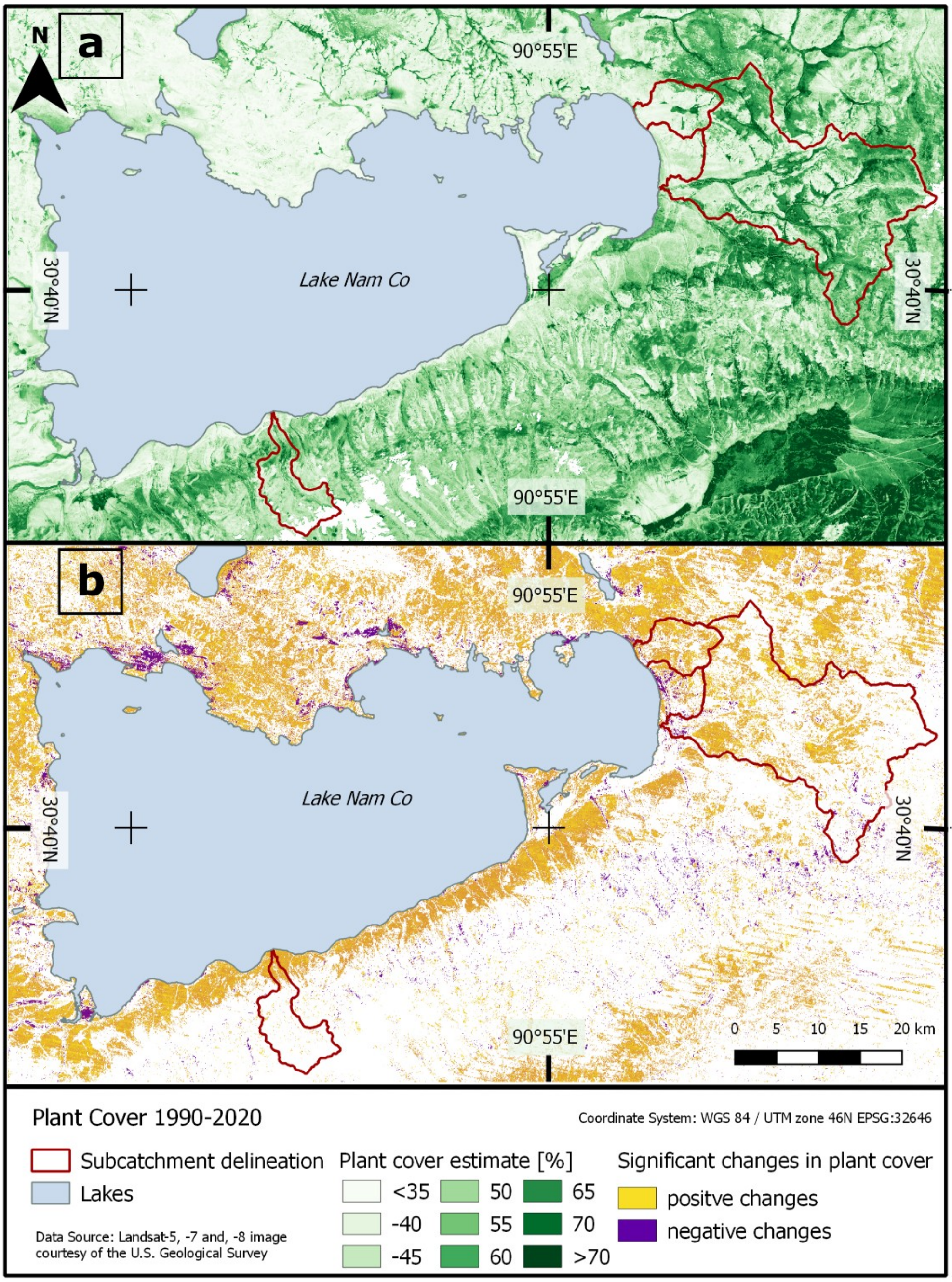

Figure 3: Mean plant cover estimate (PCE) for 1990-2020 in 30m resolution (a) and significant changes during the investigation period (b).

3.2 Solute composition in the catchments and Lake Nam Co

Water pH value clearly discriminated Lake Nam Co from the river waters in the catchments, Niyaqu, Qugaqie, Zhagu (Figure 4a, Table 1). Differences further existed between stream waters of Niyaqu and Qugaqie and between Qugaqie and Zhagu (Table 1). Samples from June/July 2018 showed higher pH values than those from May 2019 and September 2019. Glacial effluents had the lowest pH compared to other endmembers (Table 2). Mineral nitrogen varied as well in the catchments. Ammonium had smaller concentrations in the June/July 2018 sampling season than in the May 2019 sampling (Table 1), while the wetland water showed highest values compared to the other endmembers (Table 2). Water samples of the Zhagu catchment had highest nitrate concentrations, equally followed by Qugaqie and Niyaqu, while the lake had the lowest nitrate concentration (Figure 4c). Seasonal effects on nitrate were less pronounced, with May 2019 samples showing higher values compared to June/July 2018 and September 2019. Nitrate was found enriched in water directly sampled from springs, compared to other endmembers (Table 2)

The Niyaqu and Zhagu stream samples had larger calcium concentrations than the Qugaqie stream samples and the Lake Nam Co (Table 1). No effect of season was found for calcium. A large group of anions and cations showed a different pattern than calcium. Along with the EC and the DIC concentration, bromine, lithium, chlorine, fluorine, magnesium, potassium, sodium and sulphate showed highest concentrations for Lake Nam Co samples with differences compared to the three investigated catchments. The DIC concentrations discriminated between the river samples and Lake Nam Co water (Figure 4d, Table 1). Phosphate concentrations differed not only between the catchments and the lake, but differed also between the rivers, those of the Zhagu catchment had larger values than those of the Niyaqu and Qugaqie catchments.

Lake Nam Co water samples showed a smaller oxalate concentration than the river samples from the Niyaqu and Zhagu catchments. Smallest oxalate concentrations were measured in samples of the Qugaqie catchment, (Figure 4b, Table 1). In general, oxalate concentrations were observed increased in June/July 2018 than in May 2019, and September 2019. For endmembers, oxalate concentrations were found to be larger in wetland water compared to other sources (Table 2).

DOC concentrations in the water samples of the three catchments and Lake Nam Co varied between 0.1 and 23.2 mg L^{-1} (Figure 4e, Table 1). Mean DOC concentration were 4.3 mg C L^{-1} in water samples from Lake Nam Co and significantly higher compared to Niyaqu (2.2 mg C L^{-1}) and Qugaqie (2.3 mg C L^{-1}), while mean DOC concentrations of the Zhagu catchment (2.9 mg C L^{-1}) showed no statistical differences. For the endmember groups, wetland waters showed by far highest DOC concentrations (12.4 mg C L^{-1}) compared to other landscape units, while glacial waters showed the lowest concentrations (0.6 mg C L^{-1}) (Table 2).

Table 1: Solute composition for stream water samples from the Niyaqu, Qugaqie, and Zhagu catchments and from Lake Nam Co, and at different seasons, i.e. at freshet (June/July 2018 = JJ 2018), at Indian Summar Monsoon (May 2019), and at base flow (September 2019 = Sep 2019). Shown are arithmetic means and standard deviation (SD).

	Niyaqu		**Qugaqie**		**Zhagu**		**JJ 2018**		**May 2019**		**Sep 2019**		**Lake Nam Co**	
Variable	***Mean***	***SD (±)***	***Mean***	***SD (±)***	***Mean***	***SD (±)***	***Mean***	***SD (±)***	***Mean***	***SD (±)***	***Mean***	***SD (±)***	***Mean***	***SD (±)***
pH	8.23	0.49	7.11	0.65	8.06	0.38	9.41	0.13	8.24	0.94	7.90	0.41	7.88	0.70
EC [μ S cm^{-1}]	112.33	33.81	80.70	206.70	105.11	32.23	1457.5	301.38	207.68	376.64	136.05	214.57	190.17	377.92
NH_4^+ [mg L^{-1}]	0.00	0.03	0.00	0.00	0.01	0.01	0.01	0.01	0.00	0.01	0.00	0.01	0.00	0.04
NO_3^- [mg L^{-1}]	0.77	0.50	1.05	0.41	3.75	2.45	0.17	0.38	1.06	1.26	1.87	1.99	1.12	1.35
Ca^{2+} [mg L^{-1}]	9.43	4.10	3.94	1.41	8.06	2.63	2.15	0.52	6.74	4.02	7.55	3.78	8.33	4.51
Br^- [mg L^{-1}]	0.00	0.00	0.00	0.03	0.00	0.00	0.21	0.05	0.01	0.05	0.00	0.03	0.01	0.06
Li^+ [mg L^{-1}]	0.00	0.00	0.00	0.04	0.00	0.00	0.28	0.14	0.02	0.08	0.00	0.00	0.02	0.09
Cl^- [mg L^{-1}]	2.68	1.77	3.92	7.25	3.28	1.95	50.57	13.70	8.29	12.30	4.08	8.65	4.70	13.88
F^- [mg L^{-1}]	0.05	0.03	0.23	0.50	0.06	0.01	3.80	0.48	0.42	1.04	0.16	0.59	0.32	0.95
Mg^{2+} [mg L^{-1}]	1.88	0.80	1.53	5.99	1.49	0.48	52.17	9.93	5.49	12.64	3.08	9.74	4.97	13.87
K^+ [mg L^{-1}]	0.59	0.23	1.10	3.58	1.15	0.49	26.01	6.95	2.75	6.68	1.49	4.42	2.37	7.21
Na^+ [mg L^{-1}]	2.24	1.29	7.16	33.98	3.33	1.06	234.86	57.66	22.06	62.49	9.33	42.93	17.65	63.30
SO_4^{2-} [mg L^{-1}]	12.53	11.02	10.17	22.66	2.78	0.96	160.42	39.72	21.49	40.85	13.08	29.87	20.71	42.79
PO_4^{3-} [mg L^{-1}]	0.00	0.01	0.00	0.00	0.01	0.03	0.01	0.02	0.00	0.01	0.06	0.02	0.00	0.01
$(COO)_2^{2-}$ [mgL^{-1}]	0.03	0.02	0.01	0.01	0.04	0.03	0.01	0.01	0.03	0.02	0.02	0.03	0.02	0.02
DIC [mg L^{-1}]	7.60	2.32	4.54	18.75	6.26	2.68	141.63	51.13	16.41	34.80	8.24	13.84	16.92	44.47
DOC [mg L^{-1}]	2.22	2.26	2.28	5.18	2.94	2.09	4.31	3.21	3.10	4.27	1.87	2.21	2.30	2.40
$SUVA_{254}$ [L mg C^{-1} m^{-1}]	4.47	2.56	3.32	2.93	2.87	1.79	0.39	0.37	1.70	1.33	5.52	2.92	4.37	2.16

Table 2: Solute composition of different sample groups, including endmembers. Shown are arithmetic means and standard deviation (SD). Fluorescence components are in the unit of relative intensity (RI).

	Stream water		Glacial effluent		Spring water		Wetland water		Lake and brackish water	
Variable	***Mean***	***SD*** *(±)*	***Mean***	***SD*** *(±)*	***Mean***	***SD*** *(±)*	***Mean***	***SD*** *(±)*	***Mean***	***SD*** *(±)*
pH	7.99	0.63	7.29	0.91	8.05	0.56	8.14	0.27	8.68	0.88
EC [µ S cm^{-1}]	96.21	38.07	74.82	51.80	105.14	51.02	171.85	1.91	722.28	715.96
NH_4^+ [mg L^{-1}]	0.00	0.01	0.01	0.01	0.01	0.02	0.17	0.21	0.01	0.01
NO_3^- [mg L^{-1}]	1.35	1.53	0.88	0.60	3.37	2.62	0.04	0.01	0.61	0.50
Ca^{2+} [mg L^{-1}]	8.16	3.99	6.91	4.88	7.73	3.54	14.09	2.86	4.16	2.91
Br^- [mg L^{-1}]	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.11
Li^+ [mg L^{-1}]	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.13	0.17
Cl^- [mg L^{-1}]	3.02	1.90	2.00	1.42	2.42	1.95	2.10	1.47	24.54	25.96
F^- [mg L^{-1}]	0.08	0.05	0.04	0.02	0.06	0.01	0.24	0.01	1.83	1.86
Mg^{2+} [mg L^{-1}]	1.48	0.74	1.34	1.23	1.13	0.47	4.55	0.32	24.48	26.09
K^+ [mg L^{-1}]	0.67	0.39	0.46	0.25	0.68	0.38	1.30	0.21	12.39	13.45
Na^+ [mg L^{-1}]	2.25	1.27	0.65	0.35	2.58	1.16	4.72	0.59	110.23	122.48
SO_4^{2-} [mg L^{-1}]	9.64	10.00	12.29	10.98	1.98	0.80	2.95	0.04	79.39	80.00
PO_4^{3-} [mg L^{-1}]	0.00	0.02	0.00	0.00	0.01	0.04	0.04	0.06	0.01	0.02
$(COO)_2^{2-}$ [mgL^{-1}]	0.03	0.03	0.01	0.01	0.03	0.01	0.12	0.10	0.01	0.02
DIC [mg L^{-1}]	6.04	3.09	3.66	3.38	6.27	2.85	14.58	1.56	67.83	75.61
DOC [mg L^{-1}]	2.41	3.23	0.63	0.46	2.84	2.13	12.46	1.04	2.89	2.60
$SUVA_{254}$ [L mg C^{-1} m^{-1}]	3.94	2.60	4.90	2.79	3.45	2.39	2.70	0.23	2.01	2.58
Absorbance [254nm]	0.06	0.05	0.03	0.02	0.10	0.09	0.34	0.00	0.04	0.02
$\delta^{13}C$ of DOM [‰]	-24.85	3.48	-24.21	6.46	-24.22	4.81	-25.50	0.54	-23.21	3.84
FC 1 (high) [RI*10^6]	3.46	0.22	0.25	0.04	0.40	0.35	1.28	0.28	0.20	0.08
FC 2 (high) [RI*10^5]	2.05	1.44	1.57	0.21	2.87	2.63	6.04	1.38	1.15	0.39
FC 3 (high) [RI *10^5]	0.75	2.55	0.15	0.02	0.17	0.16	1.67	1.00	0.27	0.15
FC 4 (high) [RI *10^5]	1.29	1.07	1.04	0.14	1.75	1.74	5.00	1.46	0.90	0.32
FC 1 (low) [RI *10^4]	6.77	3.55	2.41	2.37	5.68	0.78			4.59	3.21
FC 2 (low) [RI *10^4]	4.96	3.26	1.37	1.69	3.68	1.36			3.31	2.25
FC 3 (low) [RI *10^4]	2.11	2.91	0.73	0.62	0.87	0.23			2.02	2.76
FC 4 (low)	1.53	0.86	2.31	5.54	1.38	0.10			1.01	0.72

[RI *10^{4}]			

The normalised ultraviolet absorbance of DOM at 254 nm (SUVA$_{254}$) revealed statistically differences between both, catchments and seasons (Figure 4f, Table 1). First, lake samples had lower absorbance values compared to the river water samples from all three catchments. Second, catchment SUVA$_{254}$ increased in the order Zhagu < Qugaqie < Niyaqu, but being statistically different between Zhagu and Niyaqu only. Samples taken in June/July 2018 had a lower SUVA$_{254}$ than the May and September 2019 samples, showing seasonal effects on SUVA$_{254}$ (Table 1). For waters from endmembers, SUVA$_{254}$ decreased in the order glacier > stream > spring > wetland > lake, while absolute absorbance at 254nm was by far highest in wetland waters, followed by spring water (Table 2).

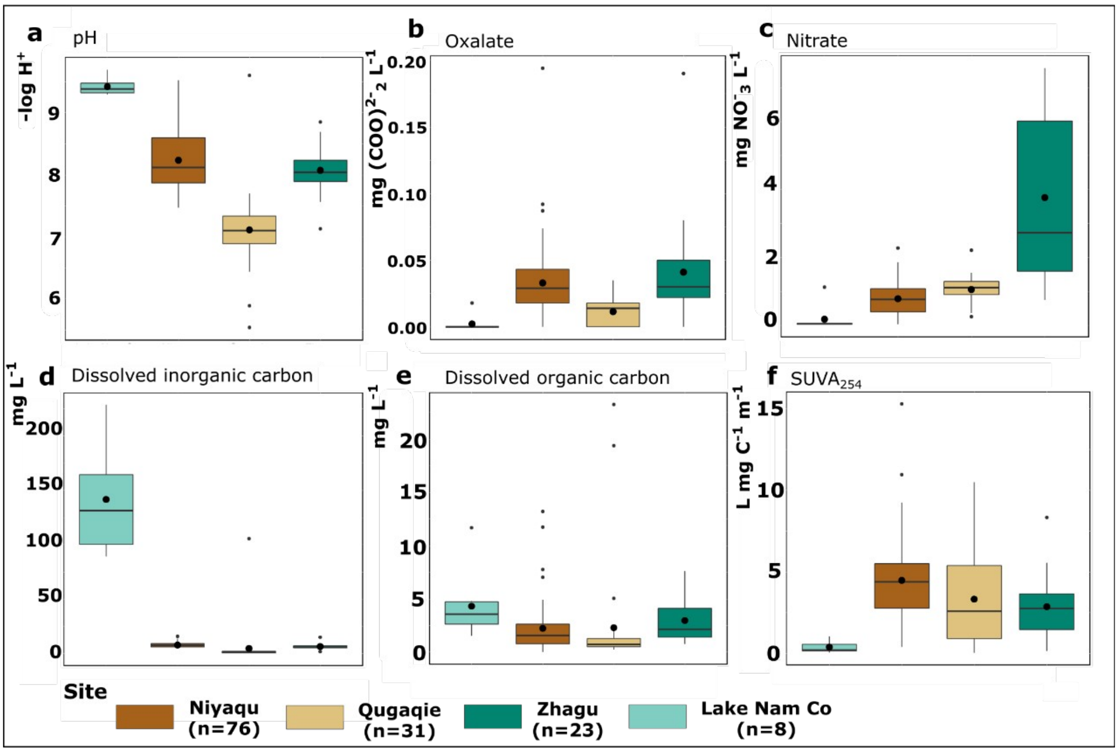

Figure 4: pH (a), oxalate (b), nitrate (c), DIC (d), and DOC (e) concentrations, and SUVA$_{254}$ values (f) of stream waters from Niyaqu, Qugaqie, Zhagu catchments and Lake Nam Co. Large black dots show the mean, horizontal lines show the median and whiskers are defined as respective quartile (I/III) +/- 1.5*inter-quartile range, outliers are shown as small black dots.

δ^{13}C values of DOM in soil and groundwater tended to be less negative as compared to those in plants (Figure 5). In contrast, organic materials in glacial sediments were much more enriched in ^{13}C compared to other samples. Consequently, the river samples of the Qugaqie catchment, receiving meltwater of the Zhadang glacier, were characterised by less negative δ^{13}C values of DOM. Also DOM from brackish water at the mouths of the rivers and from Lake Nam Co samples was enriched in ^{13}C (Table 2). Water samples of endmembers showed large differences, wetland showed the most depleted δ^{13}C of DOM, followed by glaciers and springs. δ^{13}C values discriminate lake samples from those of soils, plants, and groundwater as well as from water samples from the Zhagu and Niyaqu catchments, which form a first statistical group, except for the June/July 2018 samples (Figure 5). Riverine DOM from the Qugaqie catchment, glacial sediments and lake DOM belong to a second statistical group. With respect to strong seasonality, the δ^{13}C of DOM of the riverine samples taken in June/July 2018 were different from those of May 2019 and September 2019 (Figure 5).

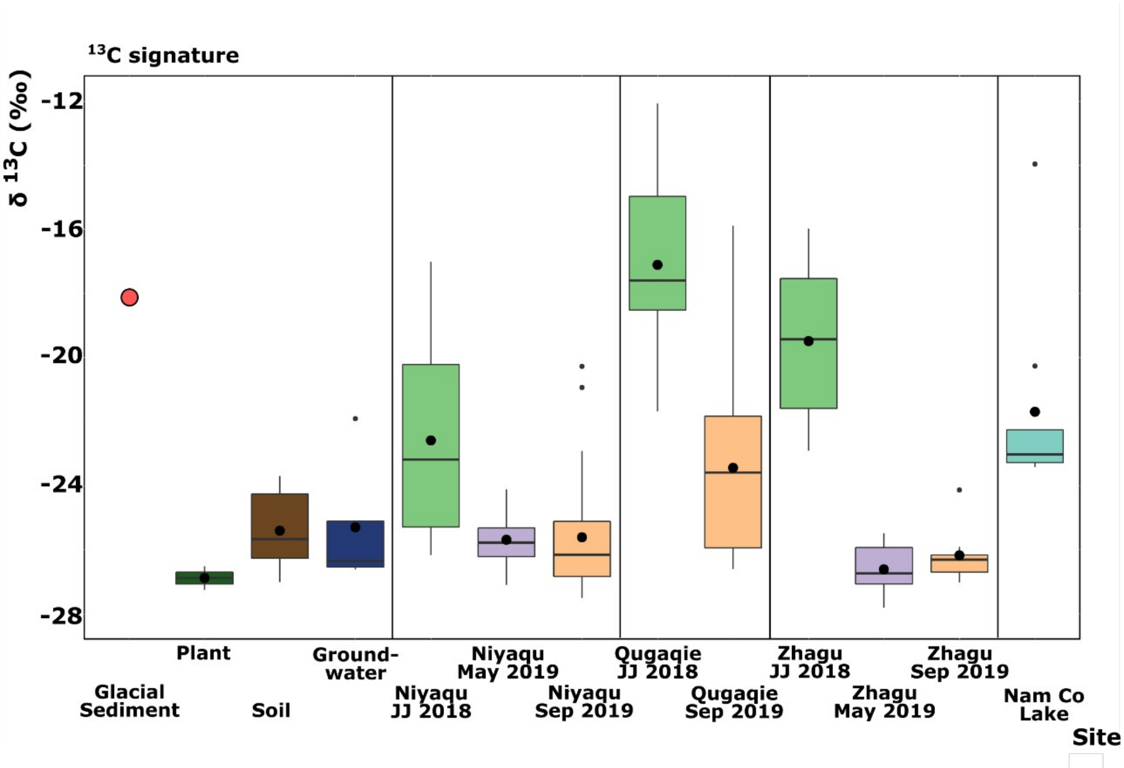

Figure 5: δ^{13}C natural abundance signatures of stream and lake DOM of the Nam Co watershed, along with its potential source material: glacial sediments (expressed as dot / n=3 with standard deviation < size of the symbol), plants, topsoils, and groundwater. Boxplots depictions is as follows: large black dots show the mean, horizontal lines show the median, and whiskers are defined as respective quartile (I/III) +/- 1.5*inter-quartile range, outliers are shown as small black dots. Abbreviations are: JJ= June/July, Sep= September; no sampling was possible in May 2019 in the Qugaqie catchment.

All variables were tested for rank-correlation using Spearman's ρ, all correlations were positive. Magnesium concentrations and EC had a high Spearman correlation ($R^2 = 0.82$), as well as DIC and EC ($R^2 = 0.62$), magnesium and DIC concentrations ($R^2 = 0.57$), and potassium and sodium concentrations ($R^2 = 0.51$). Further, negative correlation existed between δ^{13}C of DOM and SUVA$_{254}$ ($R^2 = 0.64$) and positive correlation between the DOC concentration and SUVA$_{254}$ ($R^2 = 0.32$).

3.3 PARAFAC resolved fluorescence spectroscopy EEMs

Two PARAFAC models were produced from the fluorescence EEM spectra, one for the high fluorescence response subset and one for the low fluorescence response subset. The high fluorescence response PARAFAC model consists of four components, and the number of samples described by this model was n=78. The components in the excitation mode were split between one early peak (below 280nm), one medium peak (320nm) and one late peaking component (360nm), while the fourth component has an early and a smaller late excitation phase and is regarded to describe an energy transmission effect which can occur in environmental samples (Bevilacqua et al., 2020). The emission mode is characterised by an early component peaking at below 350nm, one medium component at 425nm and two later reactions at 465nm and 500nm, respectively (Figure 6a). The sample factor loadings reveal a unique contribution of all modelled components for the samples. The component loadings are statistically different from each other (Pearson correlation coefficient).The low fluorescence response PARAFAC model revealed a four-component solution to describe the set of n= 61 samples. The excitation-emission spectra of the components and the component loadings are presented in Figure 6b. The excitation spectra of components are split in two early components, peaking at below 280nm, with an emission spectrum peak at 350nm and 420nm, respectively. Further, the model results showed a mid-range component with an excitation peak at 330nm and an emission spectrum at 430nm and a late component with excitation peaking at 390nm and emission at 475nm. The factor loadings of the low fluorescence response model show a rank shift between the dark-blue (3) and the purple component (4) for some samples. This serves as an indicator of composition changes driven by site-influences on fluorophore concentrations (Stubbins et. al., 2014). These two components are statistical related to each other. All other components of the low fluorescence response model are not related. Both models, of the low and high fluorescence response are in conjunction when excitation-emission spectra are compared to each other.

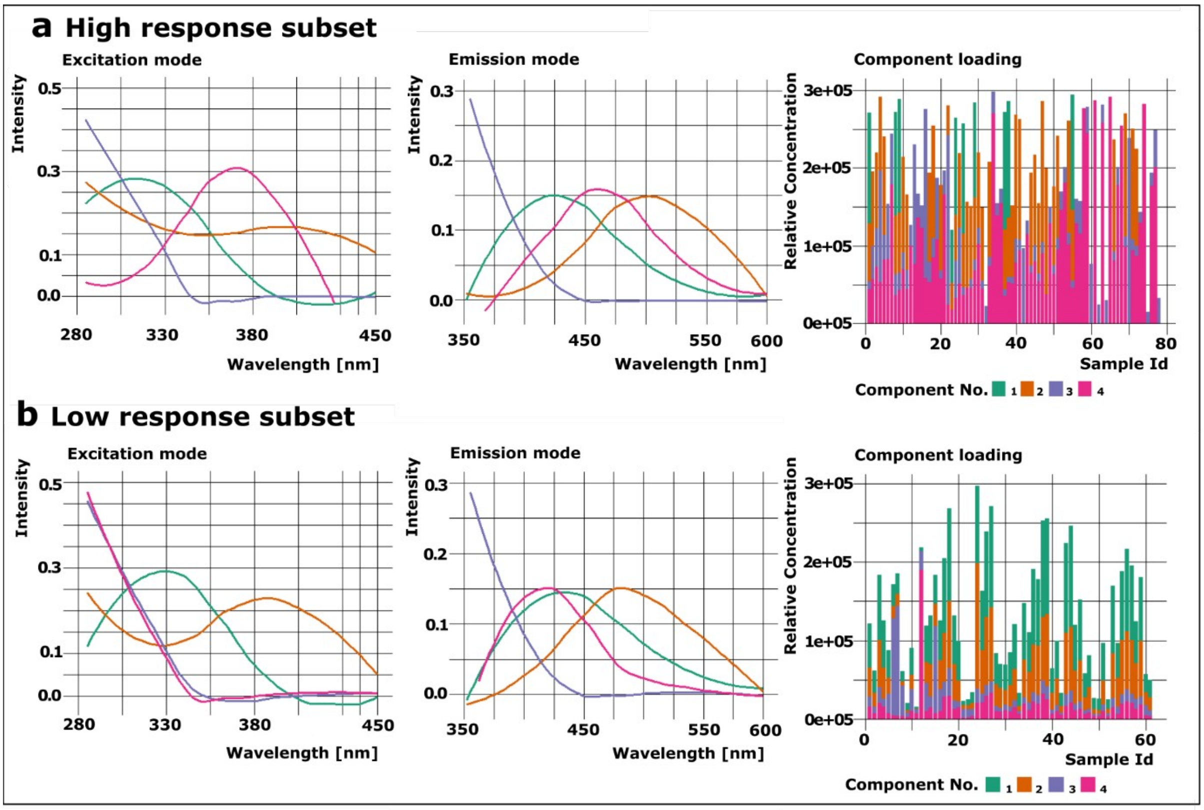

Figure 6: Excitation, emission, and factor loading modes of the four component PARAFAC models for high (a) and low (b) fluorescence response samples, respectively.

The PARAFAC models were used to test spatio-temporal FDOM variability in the dataset. Factor loading sums (Figure 7) were compared for group means, while individual FDOM-components were taken as dependent variables and analysed in NMDS ordination.

The comparison of the two model outputs shows differences in the composition of the high and low fluorescence response subset. Most samples from the Qugaqie catchment and all samples of the Nam Co Lake are grouped in the low fluorescence response subset (sample group affiliation is depicted in the supplementary material; Figure S4-S6). Concluding, the Qugaqie and all Lake Nam Co samples differ from the majority of Zhagu and Niyaqu samples. In the high fluorescence response model, no statistical effect between sites is visible. This indicates that these samples have a common FDOM composition and distribution (Figure 7a).When assessing the low fluorescence response subset (Figure 7c), the FDOM components between sites show a differentiated picture: Lake samples show lower FDOM concentration compared to the samples of the Zhagu catchment, while samples from Niyaqu and Qugaqie take a middle position.

For endmembers, no samples are in the low response subgroup for wetland waters, showing that this endmember is comparably rich in fluorophores. Consequently, in the high fluorescence subgroup wetlands show high concentrations for all FDOM components except for the first component, which has highest abundance in stream samples. Lake, brackish and glacial waters are overall comparably low in FDOM, while samples taken from springs have high abundance for the second component of the high response subset (Table 2). The low subset samples from stream, lake and brackish endmembers show high relative intensities over all components. Waters from springs show a rank shift, with FC 4 contributing more to the overall fluorescence than FC 3, it should be noted that the number of the FDOM component by definition represents its concentration rank for the whole sample set. This can point at compositional differences in spring FDOM. Glacial effluents have comparably lower values for FC 1-3, but highest values for FC 4, also associated with a rank shift, indicating a specific FDOM signature in glacial waters (Table 2).

With respect to seasons, the subsets show smaller differences. The high fluorescence response model shows higher FDOM concentrations for samples taken in September 2019 than for the June/July 2018 and May 2019 samples (Figure 7b).

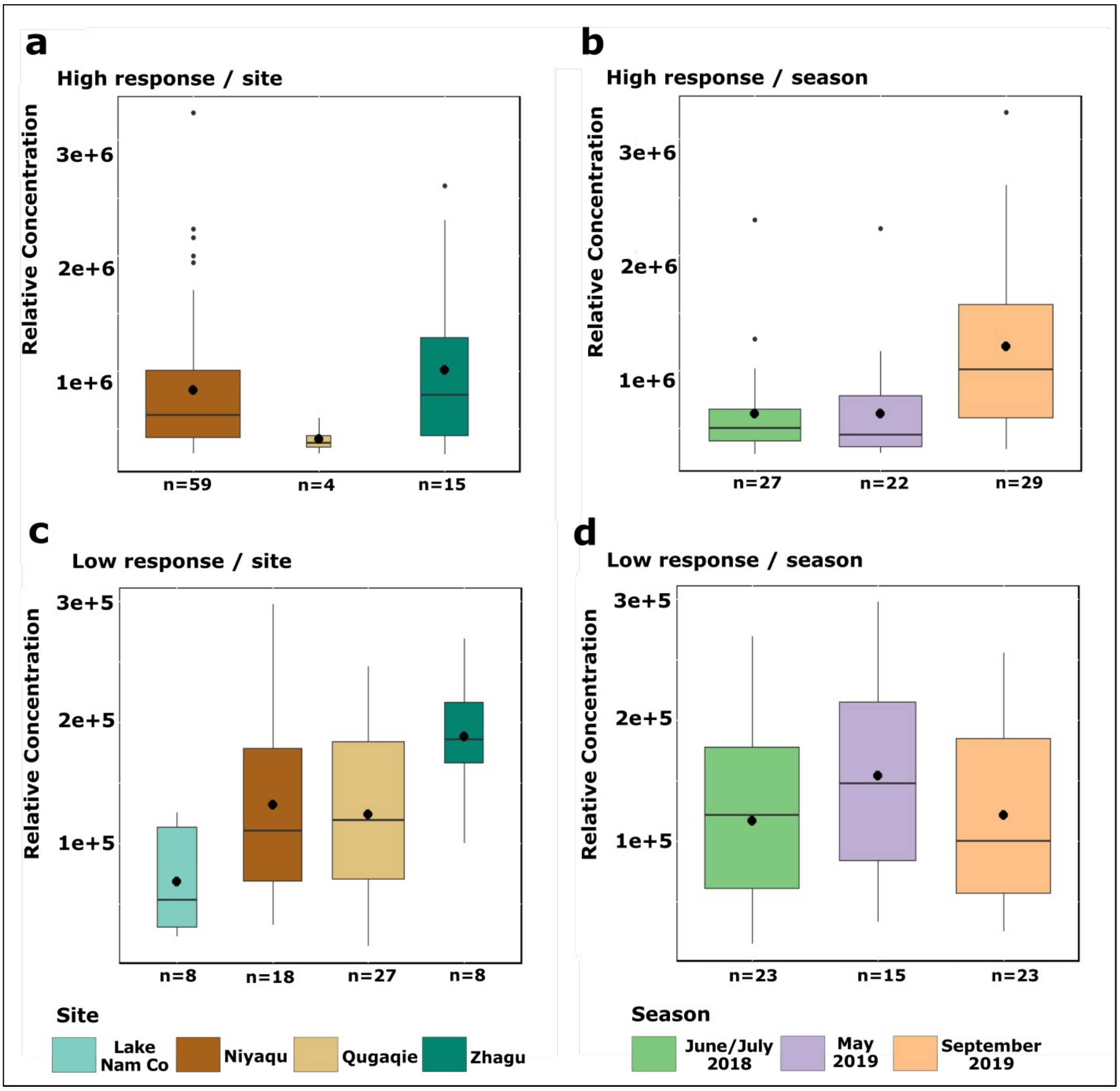

Figure 7: Sum of factor loadings of the PARAFAC models for the high fluorescence response subgroup sites: a and seasons: b and for the low fluorescence response subgroup sites: c and seasons: d. Large black dots show the mean, horizontal lines show the median, boxplot width is adjusted to sample size and whiskers are defined as respective quartile (I/III) +/- 1.5*inter-quartile range, outliers are shown as small black dots. Note the scale offset between high and low samples.

3.4 Bivariate regression and NMDS ordination of the PARAFAC resolved FDOM components with the combined DOM dataset

The FDOM components were tested for bivariate regression with all variables of the DOM and water chemistry dataset, in order to better understand FDOM chemistry and behaviour. For the high fluorescence response PARAFAC components, all here reported relations are positive (supplementary material Table S2). The first component (FC 1) shows relationships to DIC, DOC, sodium, chloride, and magnesium, FC 2 has weak relationships with DIC, DOC, chloride, sodium and, magnesium, while the third PARAFAC component (FC 3) is correlated to potassium, nitrate and negatively correlated to $\delta^{13}C$ of DOM, with weak and very weak fit, respectively. The fourth component (FC 4) shows a weak regression trend with DIC, DOC, chloride, and magnesium. FDOM component 1, 2 and 4 represent plant and soil derived fluorophores, as they are connected to solutes likely originating from the terrestrial domain. Component FC 3 likely represents a microbial FDOM component through its relation with mineralized nitrogen and less depleted signatures of $\delta^{13}C$ of DOM.

For the low fluorescence response PARAFAC subset, a positive relationship was identified between FC 1 and the $\delta^{13}C$ ratio of DOM, and FC 2 reveals positive correlations with $\delta^{13}C$ of DOM and calcium. The third PARAFAC component shows a negative correlation with $SUVA_{254}$ and a positive correlation with $\delta^{13}C$ of DOM, while FC 4 is weakly positive correlated with ammonium and $SUVA_{254}$. Hence, the FDOM components 1, 2 and 3 are linked to microbial-like fluorophores, as is indicated by the positive correlation with ^{13}C and the negative correlation with $SUVA_{254}$, while FC 4 is positively correlated to $SUVA_{254}$ and likely a plant- or soil-borne fluorophore.

The first NMDS ordination including the high fluorescence PARAFAC components of water samples is displayed in Figure 8 a,b,c for the three independent variables (site, sample category and season). Dimension 1 of this ordination divides samples with high pH, high DIC and high EC readings. Three FDOM components (FC 1, FC 2 and FC 4) and the DOC concentration define this dimension as well. The opposite direction of dimension 1 is driven by $\delta^{13}C$ of DOM. The second dimension for the first subset differentiates between $SUVA_{254}$ and sulphate on the positive side and high concentrations of nitrate, oxalate, phosphate, potassium, DOC and FC 3 (FDOM) on the negative side. When comparing the three catchments sites (Figure 8a) and sample categories (Figure 8b), samples from wetlands are strongly clustered in the positive direction of dimension 1. In contrast, samples from the Qugaqie catchment and samples from locations close to source areas (springs and glaciers) of the Zhagu and Niyaqu catchment, are scoring in the negative direction. At the second dimension, a separation of samples from the Niyaqu in the positive and the Zhagu catchments in the negative direction is apparent. For seasons, a scattering in ordination is visible (Figure 8c). The June/July 2018 samples mostly locate in the negative directions of both dimensions, while the September 2019 samples show a loading on the positive direction of the second dimension. Samples obtained in May 2019 season cluster in the core of the ordination space.

The second NMDS ordination presents the low response fluorescence FDOM components (Figure 8 d, e, f). The first dimension is explained by the DIC content, the EC readings, and most of the anions and cations. The positive direction of dimension 1 is driven by a larger marker group for influence of salts and carbonates EC, DIC concentration, bromine, lithium, chlorine, fluorine, magnesium, potassium, sodium and sulphate concentration. While $SUVA_{254}$ and FDOM component FC 4, load on the negative direction. $\delta^{13}C$ of DOM, ammonium and FDOM component FC 4 load on the positive direction of the second dimension, while pH, nitrate, as well as oxalate, calcium, phosphate and two FDOM components (FC 1, FC 2) load the negative direction. When examining sample scores for sites, it becomes apparent, that Lake Nam Co samples are separated from stream samples on the first dimension (the Qugaqie sample is close to the terminus), while the second dimension mostly resolves site effects between the catchments. The Qugaqie and Niyaqu samples are scoring in the positive direction of dimension 2, especially samples that have direct influence of glacial effluents. The majority of the Zhagu samples score in the negative ordination space, with samples close to springs showing the most extreme values. The Lake Nam Co samples are not seasonally clustered, while the stream water samples of September 2019 score more in the positive space of dimension 2 compared to the May 2019 samples, and June/July 2018 samples are spread in the ordination space.

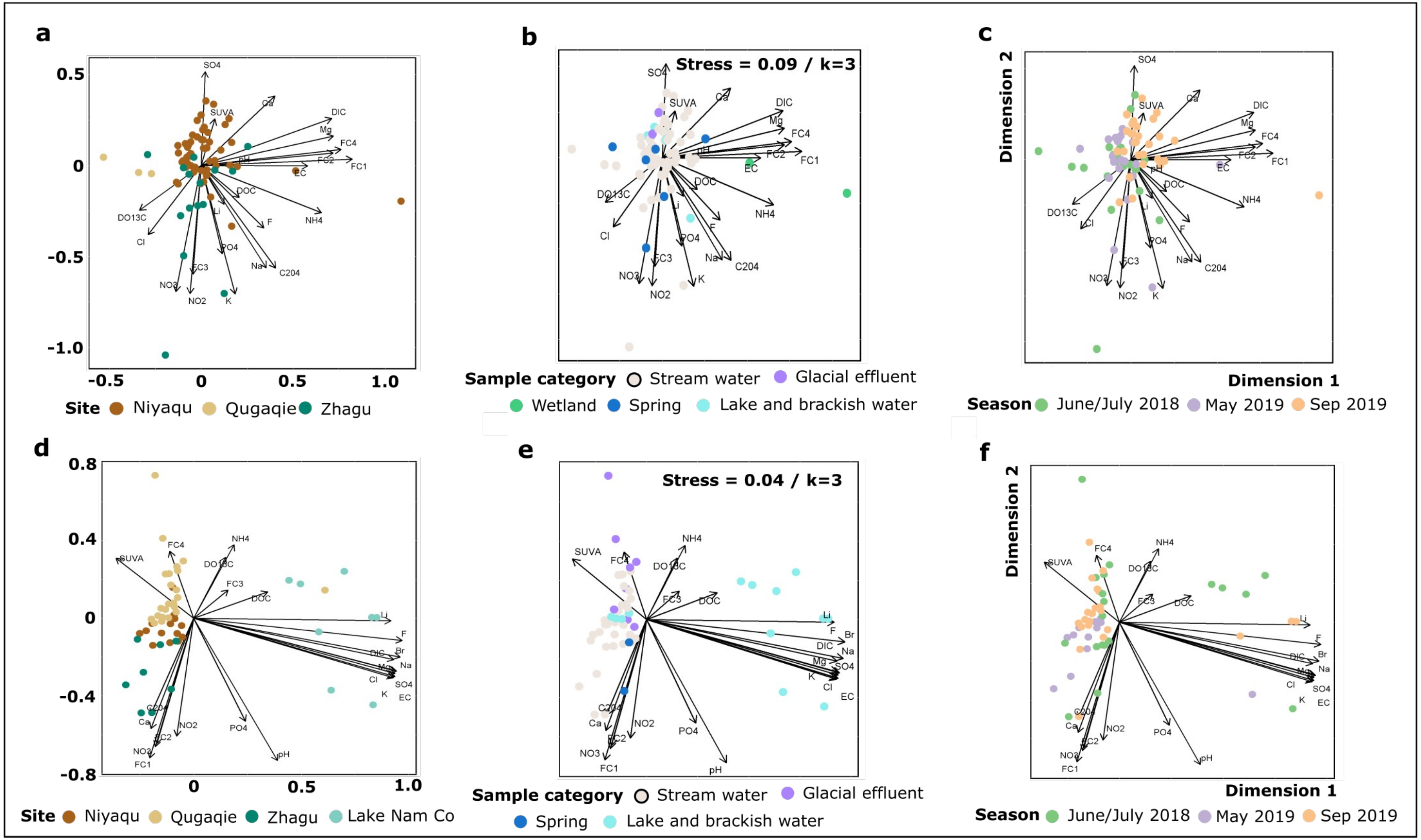

Figure 8: Non-metric multidimensional scaling (NMDS) ordination plots for the multiparameter dataset including the high fluorescence response PARAFAC model, between sampling site a), sample category b), and sampling season c). NMDS ordination plots for the subset including the low fluorescence response PARAFAC model, between sampling site d), sample category e), and sampling season f). Note: NMDS dimensions are scaled to increase readability while loading vector length and direction are retained.

4 Discussion

4.1 Plant Cover

All three catchments revealed features of degradation but with different extent (Maurischat et al., unpublished; Figure 2b,c). The plant cover estimate (PCE) showed significantly lower plant cover in Zhagu and Qugaqie compared to the Niyaqu catchment. For the Zhagu catchment, this is likely due to lack of water, caused by the absence of glacial meltwater and increasing aridity due to the lack of orographic barriers in the northern margins of the watershed and lowest MAP. Further, intense animal husbandry in the remaining productive areas increases degradative features such as patchy *K. pygmaea* sod and sparse vegetation cover. The Qugaqie catchment, which is used as a summer pasture (Tibetan Pastoralists, 2019), is to a large extent covered with productive alpine pastures at lower altitudes. At the sub-nival to nival altitudinal zone, however, there is only sparse vegetation, leading to smaller watershed-wide mean PCE values than in the Niyaqu catchment. Hence, at the Qugaqie catchment, the low plant coverage is not an indication of pasture degradation, but a consequence of its high elevation and corresponding vegetation zones.

Hopping et al. (2018) reported degraded pastures close to the southern shoreline of Lake Nam Co, as has been frequently reported for *K. pygmaea* pastures at TP scale (Damm, 1998; Qiu, 2016; Lehnert et al., 2016; Liu et al., 2018., Harris, 2010). In contrast, our satellite based PCE modelling revealed that only small patches were experiencing a decrease of plant coverage during the last 30 years. The PCE data confirmed a greening of the three catchment areas, in line with Zhang et al. (2017) and Zhong et al. (2019). However, this should not be understood as definitive evidence for the absence of degradation. Multispectral satellite data are feasible to detect changes in green cover, such as the sloughing off of pasture patches, but are limited to track changes in species composition which, according to Miehe et al. (2008) and Schleuss et al. (2015), can also be important degradation indicators. The large extent of plant cover increase in the Nam Co watershed is found mostly in areas with relatively low plant cover 30 years ago, which largely belong to the alpine steppe (Anslan et al., 2020), and most likely due to the reported water surplus in the last decades (Zhang et al., 2019; Li et al., 2020; Zhang et al., 2017).

Greening and changes in plant cover can massively influence the formation of DOM (Singh et al., 2017). Our data show that high plant cover prevails along the course of the streams, according to field observations (Maurischat, unpublished) being dominated by *K. pygmaea*. The potential direct influence of vegetation types is therefore mainly by alpine pasture.

4.2 Site-specific water DOM signatures

The DOM signature in streams can vary strongly depending on the different origin of DOM (Jaffé et al., 2012). Our data show, that water biogeochemistry and DOM composition in rivers of the three catchments and Lake Nam Co show unique signatures. They further discriminate between different landscape units, thus reflecting source areas such as wetlands, or glacial ecosystems. In the following we discuss endmembers and landscape unit features from the three investigated catchments of the Nam Co watershed.

***K. pygmaea* pastures**

Plant cover estimations show that the majority of stream water samples are under the direct influence of *K. pygmaea*, even if embedded in an alpine steppe surrounding. As outlined before, the alpine pasture in the Qugaqie and Niyaqu catchment extends along the stream path almost to the river terminus at Lake Nam Co. The majority of stream samples from Niyaqu and Qugaqie cluster together in a dimension influenced by high $SUVA_{254}$ (Figure 8d), coinciding with a high PCE in the surrounding (Figure 3). The majority of stream samples are under the influence of *K. pygmaea* pastures, as is visible by the PCE (Figure 1). We identified a positive relationship between PCE and $SUVA_{254}$ signatures together with a depletion of $\delta^{13}C$ of DOM and low DOC concentrations in the rivers. The $\delta^{13}C$ ratios of DOM of samples belonging to the stream sample category (Table 2) are closest to those of soil samples (Figure 5), underlining the direct relationship and likely input of topsoil material into the streams. To further identify the DOM composition and sources, FDOM is a feasible tool. It differentiates a microbial or protein-like DOM source and a broad range of phenolic components that form a group, *per definition* addressed as humic-like substances, such as lignin, polyphenols and tannins, as well as resynthesized compounds, rich in phenols, derived from the formation of soil organic matter in soils (Fellman et al., 2010), these substances are addressed here as phenol-rich FDOM, in order to account for the plant-borne and soil-borne source. Phenol-rich FDOM components (high subset: FC 1, FC 2, FC 4; low subset: FC 4) in stream samples of Niyaqu and Qugaqie (Figure 8a) indicate a prominent influence and steady input of plant- and soil-derived materials and suggest a negligible share of *in-situ* DOM production (e.g., by bacteria) in the river. The latter is likely due to the short residence time of DOM in the fast-flowing rivers.

River water DOM usually contains large proportions of plant and soil derived components (Coch et al., 2019; Zhou et al., 2019). In permafrost systems, a particularly high proportion of plant and soil derived DOM is connected to high DOC concentrations at times of snowmelt when there is only microbial processing in the still frozen soil in spring. Our results differ with this respect: The prevailing *K. pygmaea* pasture is well understood to retain large amounts of water, nutrients and carbon in their root turfs (Kaiser et al., 2008, Schleuss et al., 2015), being likely responsible for the low DOC concentrations in river water samples. Further, the depleted $\delta^{13}C$ values and high $SUVA_{254}$ in these samples suggest a limited microbial processing of the DOM before entering the stream. Rather, a direct washing out of plant material into streams is probable. We conclude that alpine pastures lead to a unique DOM signature, controlled by the characteristics of the firm *K. pygmaea* root mat and the shallow soils in our study area.

Glacial ecosystems

Glacial meltwater plays a key role for Nam Co's water budget (Song et al., 2014). The Niyaqu and especially the Qugaqie catchment are largely influenced by glacial meltwater (Gao et al., 2015). DOM from water samples taken at the glacial termini show unique DOM signatures. These waters showed low DOC concentrations, high $SUVA_{254}$, despite of sparse plant cover, and were richer in ^{13}C compared to DOM sampled further downstream. This is accompanied by high concentrations of ammonium and the microbial-like FDOM component FC 4 (low fluorescence response dataset). Also Spencer et al. (2014) and Li et al. (2021) reported that Tibetan glacier derived DOM is chemically distinct from stream and lake DOM. Glacial environments represent a unique environment with organic matter production by photoautotrophic microorganisms living in the glacier (Anesio et al., 2009), producing organic material which is highly enriched in ^{13}C as is contained in the sampled glacial sediments (Figure 5). The good water solubility of this material (Dubnick et al., 2010) explains the enrichment of microbial FDOM and of $\delta^{13}C$ in DOM. The high nitrogen in DOM is likely a release successive to microbial nitrogen fixation in the glacial environment (Telling et al., 2011). DOM with glacial influence is hence to a large part characterised by autochthonous DOM signatures.

The elevated values of $SUVA_{254}$ proxy on the other hand hint at a second source. Spencer et al. (2014) and Wang et al. (2019) reported high concentrations of black carbon, derived by organic matter combustion and atmospheric deposition, on Tibetan glaciers. Takeuchi (2002) identified cryconite, a powdery mixture of sediment particles, dust and microbes, blown onto the glacier from surrounding mountains or deposited by precipitation (Hodson et al., 2008), to contribute to dark-coloured organic substances in the cryosphere of Tibet. With this respect, the high $SUVA_{254}$ can be explained by further contribution of a dust borne, black carbon-like, chromophoric constituent. In conclusion, glacial DOM is characterized by a shared contribution of autochtonous organic materials derived from the microbiome of the glacier and of an allochtonous source, likely cryconite / black carbon type materials originating from atmospheric deposition. Stream DOM samples in the Qugaqie catchment are largely controlled by endmember DOM signatures from the glacier (Figure 8d, e). Adnan et al. (2019) pointed out that 80% of Qugaqie's water discharge originates from glacial meltwater, making this large influence probable.

Groundwater springs and intense pastoral activity

The Zhagu catchment comprises the lowest mean plant cover and pronounced pasture degradation. A groundwater aquifer supplies several first order streams with water (Tran et al., 2021), and the catchment was arheic during all sampling campaigns. Our data show a clear relation of high DOC and oxalate concentration with nitrate, phosphate, and potassium, mostly at sampling sites of the Zhagu catchment close to springs. The spring endmember further exhibited high concentration in microbial FDOM (FC 1, FC 2, FC 3 - low subset), and had a $\delta^{13}C$ ratio of

DOM (mean -24.2 ‰) that is richer in ^{13}C compared to that of groundwater samples (mean -27.2 ‰) and plants (mean - 26.8 ‰). Pastoral activity may have led to the high concentration of nutrients and DOC, as well as to the input of organic matter rich in ^{13}C in spring DOM. These markers suggest eutrophication and can be interpreted as the result of a strong local influence of yak dung, as was visible around springs (Supplementary materials, Figure S7, S8). According to Du et al. (2021), yak faeces are a relevant source for organic carbon and leaching of inorganic nitrogen on the TP. As the $\delta^{13}C$ composition of ungulate faeces fractionates the isotopic signature of the plant fodder source (Ma et al., 2013), this may also explain the $\delta^{13}C$ of DOM in Zhagu stream samples. In the ordination plots (Figure 8a, d), microbial FDOM components (FC 3 –high subset, FC 1 and FC 2 – low subset) are connected to spring DOM, indicating that its properties are potentially also influenced by microbial re-utilisation as possible in yak faeces. We conclude that the Zhagu catchment DOM is largely influenced by its groundwater source, which is affected by the degraded pastures and eutrophication by yak faeces in the remaining pastures.

Alpine wetlands

Wetland water samples are chemically distinct from all other sources, indicated by their large offset to other samples in the ordination space (Figure 8b). They have high DOC concentrations, along with $\delta^{13}C$ values resembling those of topsoils (Figure 5; Ah horizons are depicted in Figure 1) high oxalate concentrations in oxalate and high absolute absorbance of UV light (A254; Table 2). Shatilla and Carey (2019) reported high DOC concentrations of alpine wetlands of the Alaskan Yukon and high aromaticity of the DOM, assigning this wetland DOM to be primarily plant-derived. In the ordination plots (Figure 8b) the wetland waters are characterized by strong loading with FDOM components (FC 1, FC 2 and, FC 4), which can be identified as phenol-rich FDOM. Further, wetlands had high magnesium, ammonium, and DIC concentrations and highest EC of all catchment water samples. The ion enrichment likely is caused by the basin topography of the wetland, having an accumulation effect from the surroundings. The high ammonium concentrations further hint at *in-situ* primary production coupled with rapid degradation and subsequent release of mineral nitrogen, correspondingly, the microbial FDOM component FC 3 is as well elevated compared to other endmembers.

4.3 Unification of DOM signatures along the stream path

While unique individual endmember fingerprints largely define DOM signatures in the three catchments, aquatic systems are more than transport modules, but a mechanistic reactor, exposing DOM to biogeochemical processing during the fluvial pathway (Zark and Dittmar, 2018). Both conceptual models, the RCC (Mosher et al., 2015; Vannote et al., 1980) and the DDC (Zark and Dittmar, 2018) underline this by highlighting processing and unification of DOM. Such a unification was observed in samples of the Qugaqie catchment, where the initial glacial signal with high $SUVA_{254}$ and high ammonium gradually vanishes during the flow path (Figure 8e). Ammonium is presumably oxidized to nitrate and/or immobilized by riverine microorganisms (Aufdenkampe et al., 2001; Singer et al. 2012) This unification is likely due to alteration of DOM by photooxidation. Further, continuous inputs of plant- and soil-borne DOM from alpine pastures can explain the more depleted ^{13}C of DOM, the steady input source hampers unification processes as are reported for larger river systems worldwide (Riedel et al., 2016). The DDC states that DOM unification processes are in force globally (Kellermann et al., 2018), leading to homologous DOM signatures. Such processing was clearly identified for large rivers in the Yukon area (Shatilla and Carey, 2019) and the Ob-Irtysh system (Perminova et al. 2019). But those large streams are flowing slowly, enabling long time for microbial and photooxidative DOM processing. In contrast, the streams investigated here are small, quickly flowing with a short mean residence time of DOM before entering Lake Nam Co. Further, the high turbidity and sediment load in these rivers may shield DOM from sunlight and by this hamper unification through photooxidation. However, Ni and Li (2019) emphasized that especially chromophoric DOM as indicated by $SUVA_{254}$ is prone to fast photodegradation, likely resulting in decreased $SUVA_{254}$ indices, which we identified during the flowpath at Qugaqie. But for both other catchments, the relatively low stream order, in combination with the diverse source structure and the potential permanent inputs from alpine pastures hinder the identification of unification processes during the stream path. According to Roebuck et al. (2020), steady inputs of DOM in small watersheds render the identification of transformation processes more complex.

4.4 Season specific DOM composition

Seasonality is a major aspect, when assessing DOM properties on ecosystem scale. Particularly, freshet was found to be a major driver of DOC export in small watersheds, e.g. in permafrost ecosystems (Guggenberger et al., 2008) and in glaciated boreal watersheds of the Arctic (Lafrenière and Sharp, 2004). During freshet, in all three catchments a relative high mineral nitrogen load went along with lower DOC concentrations and more plant derived DOM than during other seasons of the year. Here, results from our study on the TP are exceptional: In fact a relative enrichment of plant derived material was reported at freshet for Siberian rivers (Prokushkin et al., 2011; Amon et al., 2012), the Yukon (Shatilla and Carey, 2019) and Arctic watersheds (Lafrenière and Sharp, 2004), but in contrast, this usually went along with higher DOC concentrations, unlike in this study. While during freshet the high contribution of plant derived DOM is due to mobilization and lateral runoff of DOM in the organic surface soil with snow melt (Guggenberger et al., 2008; Guo and Macdonald, 2006), high mineral nitrogen concentrations are likely a result of a mineralization pulse of lysed microbial cells after thawing (Austnes and Vestgarden, 2008). In our case study on the TP, the effect of freshet was smaller compared to boreal and tundra biomes. In contrast to these biomes, winter at the Lake Nam Co watershed is characterised by only small precipitation causing an absence of snow cover. Dorji et al. (2013) reported only episodical snow events in the Nam Co catchment. Nieberding et al. (2020) found, that plant primary production is rather hampered by low water availability than by deep temperature. This likely results in physiological reactions of drought resistance in *K. pygmaea* (Li et al., 2014) and alpine steppe flora (Huang et al., 2021), hampering DOM formation. Freshet effects in the investigated catchments are thus likely influenced by low water supply from the lacking snow cover, and limited accumulation of plant derived materials with associated DOM production.

During the onset of the Indian summer monsoon, DOM was strongly enriched in ^{13}C (up to +7 ‰) as compared to freshet and baseflow. This enrichment was accompanied by a shift in the composition of DOM at unchanged DOC concentrations. In June/July 2018, a coupled decrease of SUVA$_{254}$ and phenol-rich FDOM components along with less depleted ^{13}C of DOM corroborates a suggested shift towards a more microbial DOM source (Hernes et al., 2009) during the ISM. Leaching of microbial derived DOM is especially strong at high soil water contents (Caillon and Schelker, 2020) along with higher soil temperatures (Han et al., 2022), as it is the case during the ISM. Besides, we found a rerouting of draining systems in the investigated catchments. Wadi-like structures with episodic water flow were active in the Niyaqu and Zhagu catchment in June/July 2018. From these structures accumulated microbial–derived compounds, such as biofilms from small ponds and yak faeces can be washed out in great quantities. Hence, our data show a pronounced effect of the Indian summer monsoon on DOM composition and processing.

During the baseflow in September, δ^{13}C of DOM decreased again, concurrently with increased SUVA$_{254}$ and more phenol-rich FDOM: FC 1, FC 2 and FC 4 (high subset), especially in the samples of the Niyaqu catchment. This indicates, that the DOM composition after the monsoon influence went back to a pre-monsoon stage, with DOM inputs mainly derived from soils and plants.

Our results suggest unique features of DOM composition and processing with respect to seasonality. Freshet plays a smaller role, compared to boreal and tundra biomes, where this is an important factor to govern changes in DOM composition. The ISM in contrast has large effects on DOM composition for small watersheds of the TP, with a high contribution of microbial-derived DOM.

4.5 Unique DOM and biogeochemical signatures in Lake Nam Co

Terminal systems such as oceans or endorheic lakes play an important role for the cycling of DOM, as they form environments with unique DOM signatures due to processing and transformation (Goodman et al., 2011; Zark and Dittmar, 2018). In accordance with Zhang et al. (2008), the Lake Nam Co water was characterized by higher concentrations in inorganic ions including DIC compared to the water chemistry in catchments. Following Fujinami and Yasunari (2001), we attribute the increased concentration of ions to an evapoconcentration effect. Zhang et al. (2008) further showed, that Nam Co's ion chemistry is relatively unaffected by the ion signatures of inflowing streams, which we can corroborate. Nam Co is further largely unaffected by seasonality, echoing results of Kai et al. (2019), who also showed seasonal changes in DOC concentrations in Lake Nam Co to be much smaller than variations in the surrounding streams. This probably is due to the large water volume of the lake and the intense water mixing. The DOC concentration in the lake was higher than in the rivers of the catchments, except for sampling sites of Zhagu directly influenced by groundwater. $SUVA_{254}$ and the phenol-rich FDOM component of the low subset (FC 4) were depleted in lake water samples, while lake water DOM was enriched in ^{13}C during freshet and baseflow conditions with up to +5 ‰, as compared to stream DOM. Our data thus corroborate a finding from the Rocky Mountains that lake DOM and stream DOM are chemically distinct (Goodman et al., 2011). This is visible in the ordination distance between catchment sites and the lake (Figure 8d). A fluorescence component suggesting a microbial-derived DOM source (FC 3, low response dataset) was more abundant in lake water samples than in stream DOM and most endmember DOM. This gives hints for microbial activity in the lake. Intense photooxidation of the remaining chromophoric DOM in the lake water column further reduces $SUVA_{254}$, as is visible by its 90% loss compared to stream DOM (Table 1, Table 2). In addition, the higher ^{13}C of DOM in combination with protein-like FDOM suggests a change in the DOM source towards autochtonous algae-derived DOM. Following Helms et al. (2014) and Spencer et al. (2009), DOM enriched in ^{13}C along with low aromaticity indices are distinct features of terminal DOM signatures in endorheic lakes or oceans, due to microbial and photooxidative processing of riverine DOM and autochthonous microbial/algal DOM production. Hence, Lake Nam Co represents a functional terminal aquatic environment according the RCC framework.

5 Conclusions

We established a multiparameter dataset of DOM and biogeochemical water signatures (EC, pH, cations and anions, DIC, $SUVA_{254}$, PARAFAC-resolved FDOM, DOC, $\delta^{13}C$ of DOM) for three diverse catchments and for the Lake Nam Co to investigate the impact of landscape units on the source and processing of DOM along the course of the stream. The stream samples arestrongly influenced by terrestrial signatures and have site-specific, unique DOM signatures inherited from glaciers, alpine wetlands, groundwater sources and an operational signature that we attribute to the *K. pygmaea* pasture, as its abundance was positively correlated with mean plant cover of the catchments. A high site specificity of DOM in smallest stream orders diminishes with the DOM passage in the streams and further on to the lake. Lake water DOM is almost completely independent of inflowing streams due to photooxidative degradation of primarily phenol-rich organic matter and microbial processing of biolabile riverine DOM, along with pronounced autochtonous formation of algae- and microbial- derived DOM. When implementing the RCC and DDC heuristic frameworks to the QTP case study, Lake Nam Co functions as a quasi-marine terminal environment. While site-specific influences derived from endmembers are largest, in the headwaters of streams. Both findings are in-line with the RCC and DDC and manifest that these frameworks can be regarded as functional models in this high-altitude context.

Seasonal DOM variations at freshet were found less pronounced compared to studies of boreal and tundra biomes. We attribute this to the usual absence of snow cover during winter, hampering snow-melt driven DOM flush. In contrast, we identified a monsoonal control of DOC concentrations and DOM signatures, with a pronounced shift towards strong mobilization of microbial DOM sources. Reported changes in monsoon onset and intensity could hence greatly influence the input of DOM and DOC into the lake ecosystem.

Future changes of ecosystems may have pronounced impact on the DOC, inorganic solute concentrations and DOM composition in rivers entering Lake Nam Co. A glacier retreat likely will reduce the input of black carbon type organic matter into the hydrosphere of the Nam Co watershed. At the other hand, our PCE data indicate a fostered growth of alpine steppe and a greening of the lake shoreline. This may lead to higher DOC concentrations and to a higher proportion of plant derived, chromophoric DOM in stream water and finally in Lake Nam Co. To safeguard lake water quality, grazing control remains crucial. Soil degradation associated with overgrazing and intensive faeces production may lead to higher proportions of microbial-derived DOM and mineral nitrogen input to the streams, bearing the risk of lake eutrophication.

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Study II – Supplementary materials

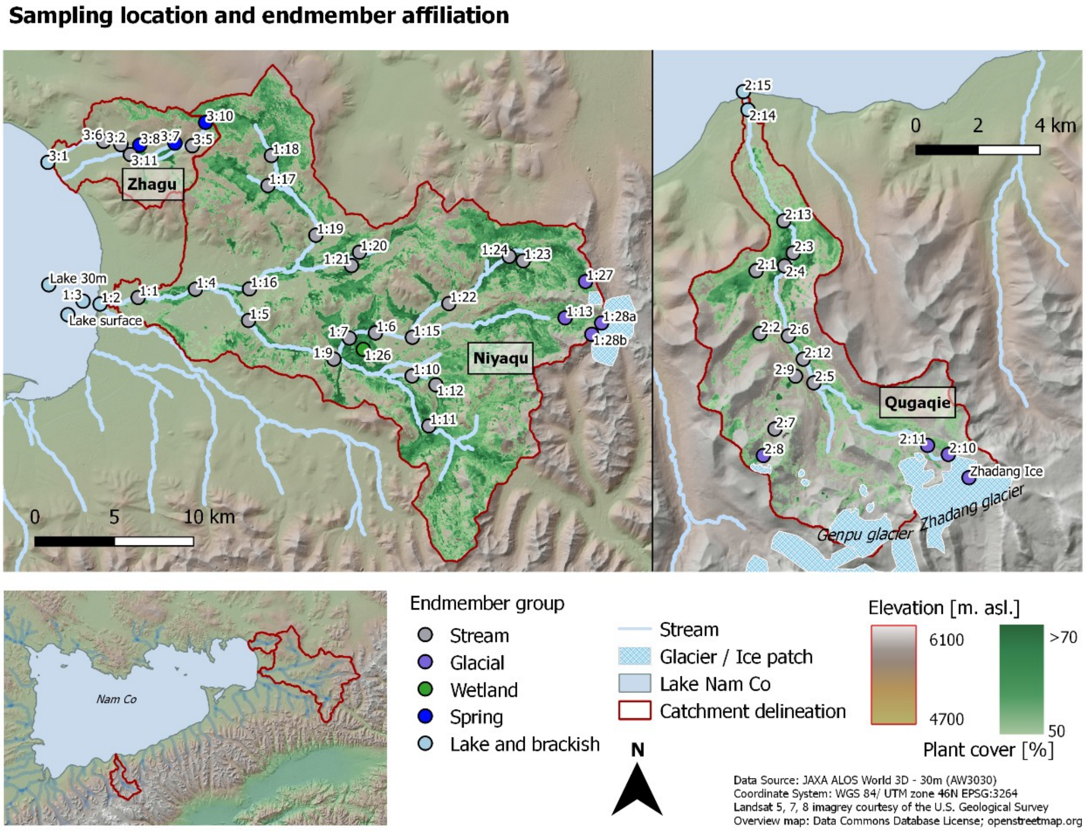


Figure S1: Endmember affiliation and sample Id for stream sampling points

Table S1: Statistical report for the plant cover estimate resolved on the three investigated subcatchments

Catchment	Mean plant cover [%] (1990-2020)	Relative standard deviation (RSD) [%]	Difference rank mean (Conover-Iman test) P value α=0.05 *, 0.01**, 0.001***		
			Niyaqu	Qugaqie	Zhagu
Niyaqu	52,3	9,49			
Qugaqie	49,5	6,44	52,6***		
Zhagu	44,5	8,15	198,7***	112,35***	

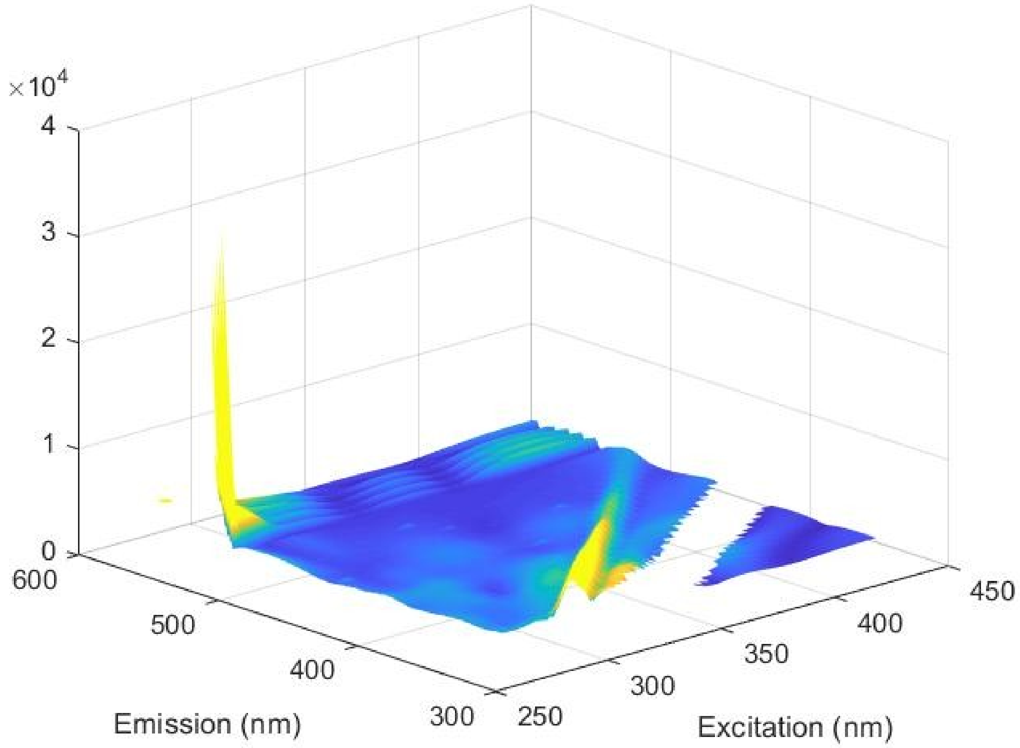


Figure S2: Mean residual EEM of the high fluorescence response sample set

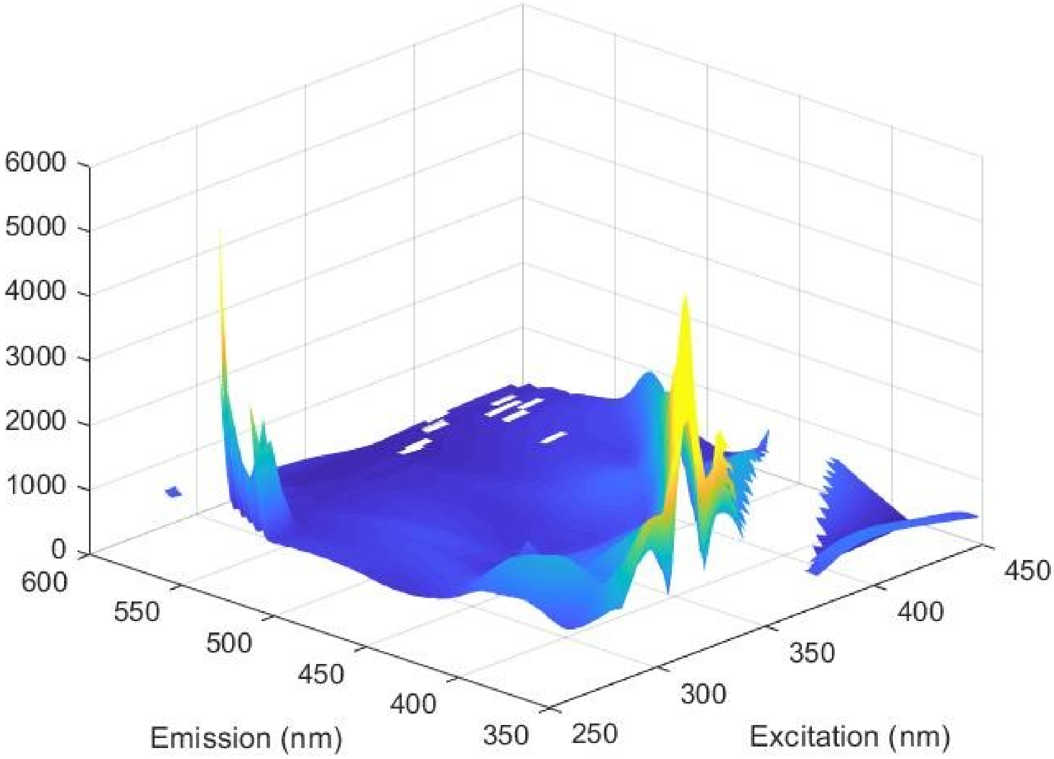


Figure S3: Mean residual EEM of the low fluorescence response sample set

Table S2: Bivariate regression R^2 and significance of the PARAFAC resolved components (high) with the multiparameter dataset. Significance indicators: ‘*’ 0.001; ‘**’ 0.01; ‘*’ 0.05; ‘.’ >0.05, NA = not calculated**

Tested Variable	FC1		FC2		FC3		FC4		Subgroup
	R^2	p	R^2	p	R^2	p	R^2	p	
DOC	0.23	***	0.16	***	0.01	.	0.15	***	High
DIC	0.34	***	0.27	***	0.00	.	0.31	***	High
SUVA$_{254}$	0.00	.	0.00	.	0.06	*	0.00	.	High
pH	0.00	.	0.00	.	0.00	.	0.00	.	High
EC	0.06	*	0.05	*	0.00	.	0.05	*	High
DO^{13}C	0.05	.	0.06	*	0.08	*	0.07	*	High
Na	0.14	***	0.10	**	0.04	.	0.10	**	High
K	0.01	.	0.00	.	0.17	***	0.00	.	High
F	0.17	***	0.09	**	0.03	.	0.11	**	High
Br	NA	NA	NA	NA	NA	NA	NA	NA	High
Li	0.00	.	0.00	.	0.00	.	0.00	.	High
NH$_4$	0.21	***	0.11	**	0.00	.	0.17	***	High
Cl	0.19	***	0.23	***	0.04	.	0.25	***	High
SO$_4$	0.04	.	0.05	*	0.00	.	0.05	.	High
PO$_4$	0.00	.	0.00	.	0.08	**	0.00	.	High
NO$_2$	0.01	.	0.01	.	0.11	**	0.02	.	High
NO$_3$	0.06	*	0.04	.	0.08	*	0.05	.	High
C$_2$O$_4$	0.11	**	0.05	*	0.03	.	0.05	.	High
Mg	0.21	***	0.10	**	0.00	.	0.12	**	High
Ca	0.08	*	0.05	*	0.00	.	0.04	.	High

Table S2 (continued): Bivariate regression R^2 and significance of the PARAFAC resolved components (low) with the multiparameter dataset. Significance indicators: ‘*’ 0.001; ‘**’ 0.01; ‘*’ 0.05; ‘.’ >0.05, NA = not calculated**

Tested Variable	FC1		FC2		FC3		FC4		Subgroup
	R^2	p	R^2	p	R^2	p	R^2	p	
DOC	0.02	.	0.02	.	0.09	*	0.00	.	Low
DIC	0.10	*	0.06	.	0.02	.	0.03	.	Low
SUVA$_{254}$	0.00	.	0.00	.	0.13	**	0.01	.	Low
pH	0.00	.	0.00	.	0.03	.	0.02	.	Low
EC	0.11	*	0.06	.	0.03	.	0.03	.	Low
DO^{13}C	0.14	**	0.14	**	0.11	*	0.06	.	Low
Na	0.11	*	0.07	.	0.03	.	0.03	.	Low
K	0.10	*	0.06	.	0.03	.	0.02	.	Low
F	0.12	**	0.08	*	0.03	.	0.03	.	Low
Br	0.12	*	0.07	*	0.03	.	0.03	.	Low
Li	0.11	*	0.07	*	0.04	.	0.03	.	Low
NH$_4$	0.10	*	0.05	.	0.00	.	0.24	***	Low
Cl	0.11	*	0.07	*	0.04	.	0.02	.	Low
SO$_4$	0.11	*	0.07	.	0.03	.	0.03	.	Low
PO$_4$	0.00	.	0.00	.	0.00	.	0.00	.	Low
NO$_2$	0.05	.	0.02	.	0.00	.	0.00	.	Low
NO$_3$	0.14	**	0.07	*	0.00	.	0.01	.	Low
C$_2$O$_4$	0.21	***	0.15	**	0.02	.	0.04	.	Low

Mg	0.12	*	0.07	*	0.01	.	0.03	.	Low
Ca	0.08	*	0.07	*	0.04	.	0.00	.	Low

Table S3: Parameter loading per NMDS dimension for the high fluorescence response PARAFAC components and the multiparamter dataset

Loadings for high fluorescence response PARAFAC and the multiparamter dataset					
Variable	Vectors		Coefficient of Determination	Corrected p-value	Significance P value α=0.05 *, 0.01**, 0.001***
	NMDS1	NMDS2	r2	Pr(>r)	
DOC	0.76097	-0.64878	0.0736	0.089	.
DIC	0.93972	0.34195	0.5715	0.001	***
SUVA	0.27869	0.96038	0.0710	0.082	.
pH	0.99985	-0.01737	0.0379	0.244	
EC	0.99999	-0.00344	0.3364	0.001	***
$DO^{13}C$	-0.80637	-0.59141	0.1737	0.005	**
Na	0.53210	-0.84668	0.4408	0.001	***
K	0.25314	-0.96743	0.5348	0.001	***
F	0.70391	-0.71029	0.2333	0.005	**
Li	0.50367	-0.86390	0.0611	0.107	
NH4	0.92930	-0.36933	0.4958	0.002	**
Cl	-0.60552	-0.79583	0.2290	0.001	***
SO4	0.04107	0.99916	0.2671	0.002	**
PO4	0.22957	-0.97329	0.2492	0.002	**
NO2	-0.08651	-0.99625	0.5004	0.001	***
NO3	-0.19473	-0.98086	0.4996	0.001	***
C204	0.58379	-0.81191	0.4824	0.001	***
Mg	0.97562	0.21947	0.5437	0.001	***
Ca	0.72398	0.68982	0.3071	0.001	***
FC1	0.99913	0.04172	0.6770	0.001	***
FC2	0.99543	0.09546	0.5219	0.001	***
FC3	-0.0745	-0.99722	0.3596	0.018	*
FC4	0.99347	0.11410	0.5880	0.001	***
Permutations free; Number of permutations 999					

Table S4: Parameter loading per NMDS dimension for the low fluorescence response PARAFAC components and the multiparamter dataset

Loadings for low fluorescence response PARAFAC and the multiparamter dataset					
Variable	Vectors		Coefficient of Determination	Corrected	Significance P value α=0.05 *, 0.01**, 0.001***
	NMDS1	NMDS2	r2	Pr(>r)	
DOC	0.93089	-0.36531	0.1377	0.043	*
DIC	0.97433	0.22512	0.9121	0.001	***
SUVA	-0.75736	-0.65300	0.2277	0.001	***
pH	0.46986	0.88274	0.6871	0.001	***
EC	0.95196	0.30621	0.9679	0.001	***
$DO^{13}C$	0.42994	-0.90286	0.1206	0.043	*
Na	0.96035	0.27879	0.9656	0.001	***

K	0.95205	0.30596	0.9596	0.001	***
F	0.99304	0.11779	0.9544	0.001	***
Br	0.97894	0.20414	0.9644	0.001	***
Li	0.99991	0.01365	0.8434	0.001	***
NH4	0.44866	-0.89370	0.1768	0.005	**
Cl	0.94944	0.31396	0.9557	0.001	***
SO4	0.95680	0.29075	0.9587	0.001	***
PO4	0.41517	0.90974	0.3371	0.002	**
NO2	-0.12896	0.99165	0.3741	0.001	***
NO3	-0.25981	0.96566	0.4684	0.001	***
C2O4	-0.37397	0.92744	0.2830	0.002	**
Mg	0.96056	0.27808	0.9436	0.001	***
Ca	-0.33142	0.94348	0.3607	0.001	***
FC1	-0.27432	0.96164	0.5574	0.001	***
FC2	-0.25780	0.96620	0.4439	0.001	***
FC3	0.72346	-0.69037	0.0473	0.285	.
FC4	-0.30704	-0.95170	0.1307	0.063	.
Permutations free; Number of permutations 999					

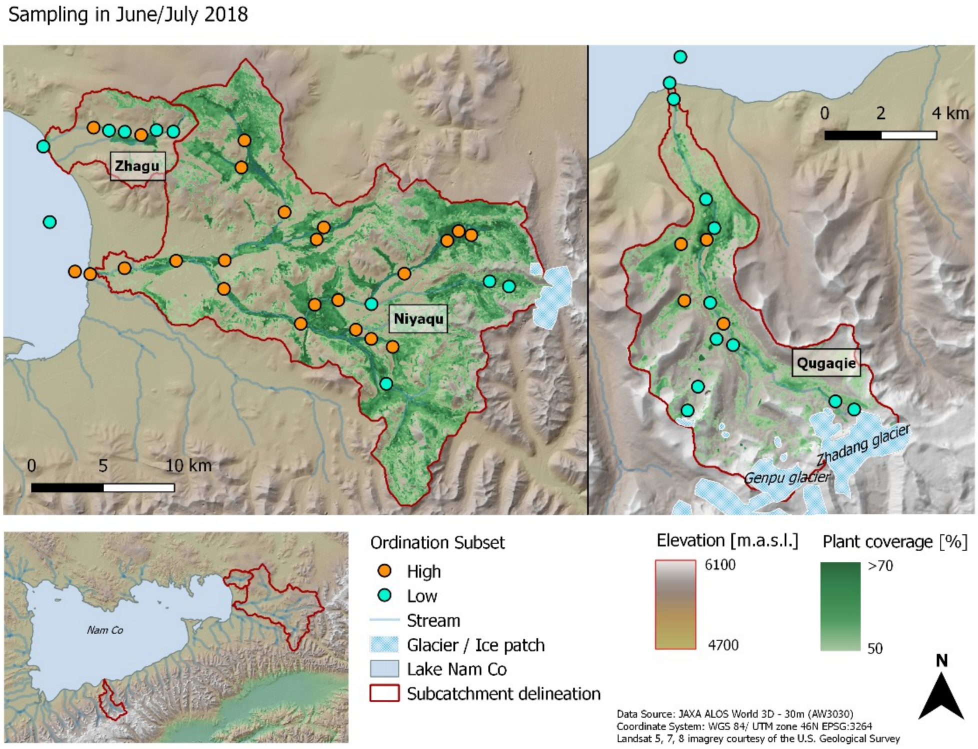
Table S5: Individual stream sample loadings per NMDS dimension. JJ 2018 =June/July 2018, Sep 2019= September 2019

Sample	Season	Group	NMDS1	NMDS2
1_1	JJ 2018	HIGH	-1,096	0,159
1_2	JJ 2018	HIGH	-0,706	0,365
1_3	JJ 2018	HIGH	1,754	-2,583
1_4	JJ 2018	HIGH	-0,560	0,307
1_5	JJ 2018	HIGH	-0,456	-0,311
1_6	JJ 2018	HIGH	-0,833	0,052
1_7	JJ 2018	HIGH	0,878	-0,068
1_8	JJ 2018	HIGH	-1,342	0,443
1_9	JJ 2018	HIGH	-0,898	0,769
1_10	JJ 2018	HIGH	0,038	1,789
1_11	JJ 2018	LOW	-2,595	-2,415
1_12	JJ 2018	HIGH	0,353	2,279
1_14	JJ 2018	LOW	-5,489	-3,138
1_15	JJ 2018	LOW	-3,621	-2,977
1_16	JJ 2018	HIGH	0,003	-0,221
1_17	JJ 2018	HIGH	-0,379	-0,169
1_18	JJ 2018	HIGH	0,298	-0,405
1_19	JJ 2018	HIGH	0,333	-0,417
1_20	JJ 2018	HIGH	-1,507	-0,586
1_21	JJ 2018	HIGH	-0,907	-0,069
1_22	JJ 2018	HIGH	-0,591	0,427
1_23	JJ 2018	HIGH	0,509	-1,305
1_24	JJ 2018	HIGH	-0,930	0,173
1_1	May 2019	HIGH	-0,718	0,790
1_2	May 2019	HIGH	-0,461	1,159
1_3	May 2019	HIGH	-0,902	0,863

1_4	May 2019	LOW	-12,538	-1,959
1_5	May 2019	LOW	-7,459	-1,911
1_6	May 2019	LOW	-9,683	-0,686
1_7	May 2019	LOW	-11,060	-1,624
1_9	May 2019	LOW	-4,520	0,445
1_13	May 2019	LOW	-4,423	-0,048
1_14	May 2019	HIGH	0,154	0,696
1_15	May 2019	LOW	-5,398	-0,720
1_16	May 2019	HIGH	-0,352	-0,070
1_17	May 2019	HIGH	0,340	-0,183
1_18	May 2019	HIGH	0,140	-0,635
1_19	May 2019	HIGH	-0,053	-0,693
1_20	May 2019	HIGH	-1,391	-0,662
1_21	May 2019	HIGH	-1,116	0,164
1_22	May 2019	HIGH	-1,340	0,813
1_23	May 2019	HIGH	-0,596	0,010
1_24	May 2019	HIGH	-0,773	0,726
1_26	May 2019	HIGH	5,605	-0,336
1_27	May 2019	HIGH	-0,576	0,921
1_1	Sep 19	HIGH	-0,187	1,038
1_2	Sep 19	HIGH	0,162	0,934
1_3	Sep 19	HIGH	0,279	0,612
1_4	Sep 19	HIGH	0,523	0,843
1_5	Sep 19	HIGH	0,067	0,956
1_6	Sep 19	HIGH	0,152	1,297
1_7	Sep 19	HIGH	0,439	1,197
1_9	Sep 19	HIGH	0,816	2,224
1_10	Sep 19	HIGH	1,699	1,712
1_11	Sep 19	LOW	-9,491	-3,486
1_12	Sep 19	HIGH	1,476	1,471
1_13	Sep 19	LOW	-2,816	-0,966
1_14	Sep 19	HIGH	1,835	0,834
1_15	Sep 19	LOW	-3,860	-1,952
1_17	Sep 19	HIGH	2,112	-0,028
1_18	Sep 19	HIGH	0,818	0,160
1_19	Sep 19	HIGH	1,711	-0,004
1_20	Sep 19	HIGH	0,504	0,180
1_21	Sep 19	HIGH	-0,820	0,343
1_22	Sep 19	HIGH	0,359	0,643
1_23	Sep 19	HIGH	1,131	0,664
1_24	Sep 19	HIGH	0,228	1,427
1_26	Sep 19	HIGH	11,967	-1,179
1_27	Sep 19	HIGH	-0,164	1,711
1_28	Sep 19	LOW	-4,863	3,959
2_1	JJ 2018	HIGH	-3,730	-0,159
2_3	JJ 2018	LOW	-3,698	2,662
2_4	JJ 2018	HIGH	-2,964	-0,187
2_7	JJ 2018	LOW	-3,716	6,131
2_9	JJ 2018	LOW	-4,025	3,648

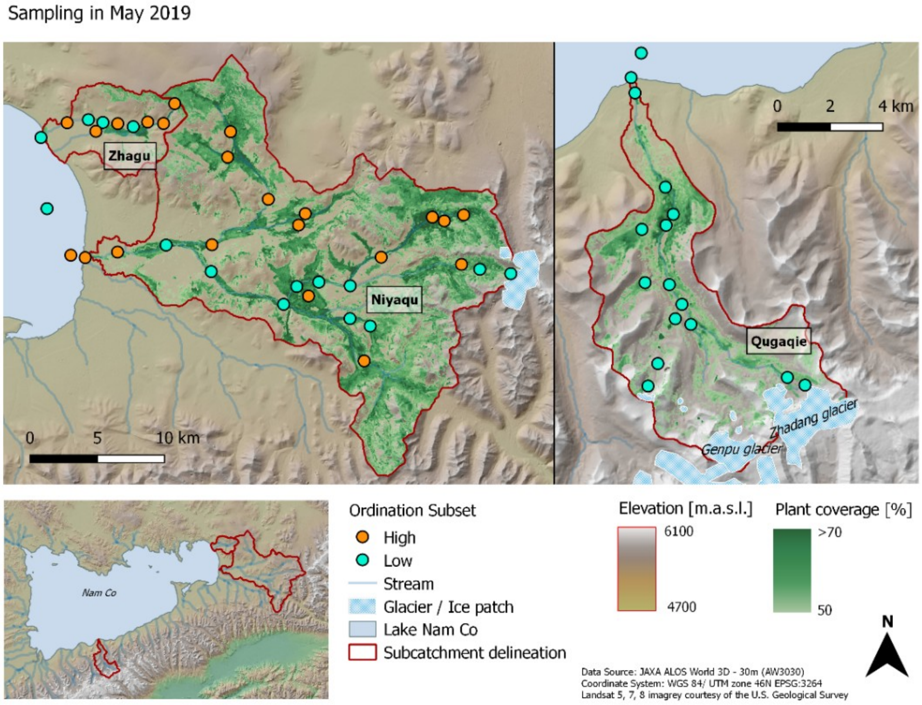
2_10	JJ 2018	LOW	-2,386	7,363
2_11	JJ 2018	LOW	-8,981	18,383
2_12	JJ 2018	HIGH	-5,814	1,205
2_13	JJ 2018	LOW	-5,671	5,728
2_14	JJ 2018	LOW	-4,094	0,810
2_15	JJ 2018	LOW	30,539	3,605
2_14	May 2019	LOW	-5,833	0,065
2_1	Sep 19	LOW	-9,165	-0,623
2_2	Sep 19	LOW	-9,826	-0,124
2_3	Sep 19	LOW	-5,769	1,852
2_4	Sep 19	LOW	-6,574	1,111
2_5	Sep 19	LOW	-5,299	4,263
2_7	Sep 19	LOW	-6,569	5,676
2_8	Sep 19	LOW	-7,199	10,269
2_9	Sep 19	LOW	-4,698	1,948
2_10	Sep 19	LOW	-3,869	6,632
2_11	Sep 19	LOW	-7,588	1,272
2_12	Sep 19	LOW	-4,466	1,473
2_13	Sep 19	LOW	-7,427	-0,391
2_14	Sep 19	LOW	-8,274	0,325
2_15	Sep 19	LOW	-7,177	0,117
3_1	JJ 2018	LOW	41,756	-11,130
3_2	JJ 2018	LOW	-12,189	-12,171
3_3	JJ 2018	HIGH	-1,300	-1,882
3_5	JJ 2018	LOW	-13,092	-2,706
3_6	JJ 2018	HIGH	-2,666	-7,044
3_7	JJ 2018	LOW	-4,054	-2,940
3_8	JJ 2018	LOW	-5,247	-9,130
3_1	May 2019	LOW	32,026	-9,280
3_2	May 2019	LOW	-11,695	-6,961
3_3	May 2019	LOW	-7,755	-3,410
3_5	May 2019	HIGH	-0,715	-0,315
3_6	May 2019	LOW	-15,806	-8,573
3_7	May 2019	HIGH	-1,044	-0,049
3_8	May 2019	HIGH	-1,111	-3,696
3_9	May 2019	HIGH	1,272	-4,917
3_10	May 2019	HIGH	-3,040	0,550
3_11	May 2019	HIGH	-0,257	-1,534
3_1	Sep 19	LOW	29,122	-1,775
3_2	Sep 19	HIGH	-0,728	-1,620
3_3	Sep 19	HIGH	-0,085	-0,679
3_5	Sep 19	HIGH	0,808	-0,198
3_6	Sep 19	LOW	-9,742	-12,137
3_7	Sep 19	HIGH	2,779	0,702
3_8	Sep 19	HIGH	0,108	-1,513
3_10	Sep 19	HIGH	-0,407	0,360
3_11	Sep 19	HIGH	1,865	-0,255
Lake 30m	JJ 2018	LOW	35,092	6,023
Lake 5m	JJ 2018	LOW	24,826	4,401

L1_1	JJ 2018	LOW	22,209	4,871
Lake SC1	Sep 19	LOW	42,642	0,087
Lake SC2	Sep 19	LOW	41,482	0,112



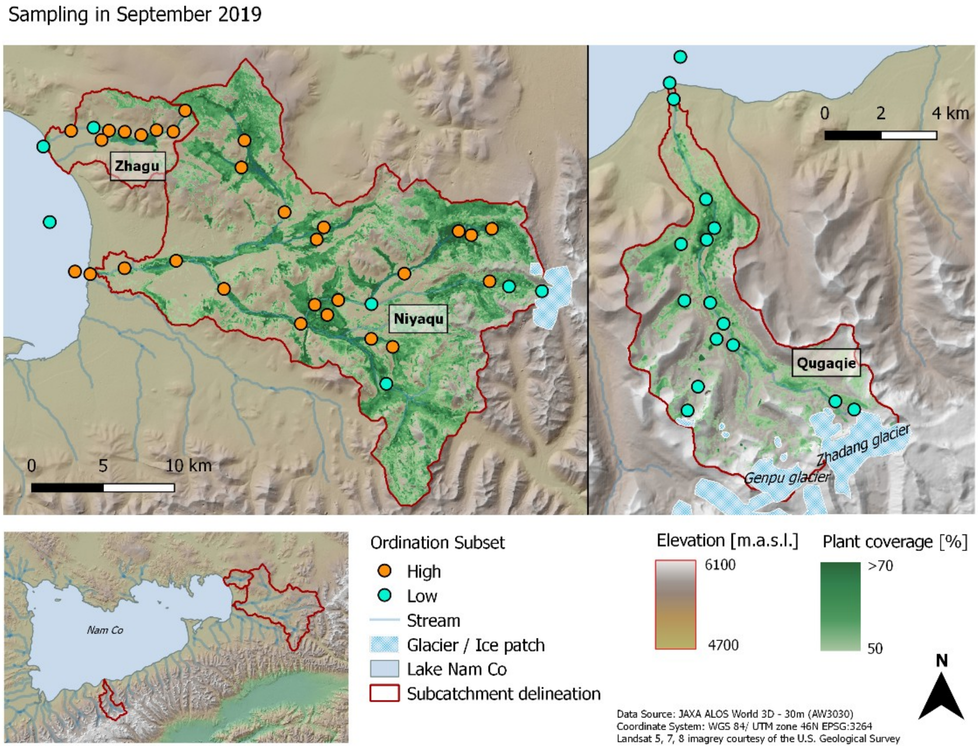
Data Source: JAXA ALOS World 3D - 30m (AW3030)
Coordinate System: WGS 84/ UTM zone 46N EPSG:3264
Landsat 5, 7, 8 imagrey courtesy of the U.S. Geological Survey

Figure S4: Ordination subset group affiliation per stream sample in the June/July 2018 sampling season



Data Source: JAXA ALOS World 3D - 30m (AW3030)
Coordinate System: WGS 84/ UTM zone 46N EPSG:3264
Landsat 5, 7, 8 imagrey courtesy of the U.S. Geological Survey

Figure S5: Ordination subset group affiliation per stream sample in the May 2019 sampling season



Data Source: JAXA ALOS World 3D - 30m (AW3030)
Coordinate System: WGS 84/ UTM zone 46N EPSG:3264
Landsat 5, 7, 8 imagrey courtesy of the U.S. Geological Survey

Figure S6: Ordination subset group affiliation per stream sample in the September 2019 sampling season

Figure S7: Topview of a Zhagu source site (Sample point 3_8)

Figure S8: Detail of the downstream direction of sampling site 3_8 in the Zhagu catchment. Note: the trampling damage in the foreground

Study III

Dissolved organic matter sources and processing in the endorheic Lake Nam Co watershed (Tibet) as assessed by ultra-high resolution Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS)

Contribution: I designed the experimental setup, carried out the field work and analysis. I wrote the original draft and was greatly involved in reviewing and editing of the manuscript.

Published: in preparation for publication

Dissolved organic matter sources and processing in the endorheic Lake Nam Co watershed (Tibet) as assessed by ultra-high resolution Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS)

Technical Report

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Study III

Abstract

The aim of this study was the in-depth characterisation of solid phase extracted (SPE) dissolved organic matter (DOM) obtained from three catchments of the Tibetan endorheic Lake Nam Co and the lake itself in order to get insight into the processes of DOM transformation along the flow path to the lake. In three catchments, we sampled river waters spanning different vegetation zones, including *Kobresia pygmaea* dominated alpine pastures. Further three endmembers, the Zhadang glacier of the Nyainqentanglha mountain ridge, an upland spring and an alpine wetland were sampled. DOM signatures of streams, the brackish mixing zone and of the oligosaline, oligotrophic Lake Nam Co were sampled as well. Ultra-high resolution Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS) is utilised on 48 DOM samples. We aim to identify DOM chemical signatures of contributing sources and to facilitate an understanding of DOM processing. Non-metric multidimensional scaling (NMDS) was used to identify the biogeochemical distance between samples of this diverse dataset.

Our data show that the DOM molecular composition differed between the three endmembers in the three catchments as well as between stream samples, the brackish mixing zone and the lake itself. Glacial meltwater shows signs of autochthonous DOM production in the glacial ecosystem alongside with allochthonous, dust-borne DOM sources. Glacial-fed streams are largely influenced by the glacial endmember, releasing both, fresh autochthonous and allochthonous, aromatic plant- and soil derived DOM. The spring water endmember showed background DOM signatures inherited from the direct contact with the microbial biome of the shallow upper aquifers, and the surrounding soils, while wetlands and streams are characterised mostly by inputs from vascular plants and soils. The brackish zone in the lake showed an intermixing of terrestrial-borne DOM signatures with lake water DOM, while Lake Nam Co exhibited most distinct DOM characteristics suggesting an intense microbial processing as well as photobleaching of inflowing stream DOM, alongside with an autochthonous microbial DOM source.

In conclusion, our study reveals that DOM characteristics are largely influenced by local endmembers such as glaciers, springs and wetlands. In the case of the glacial dominated catchment, the unique glacial signature is translocated with the stream towards the lake. Streams of catchments with less or no glacial influence show mostly plant- and soil borne DOM sources, while the large endorheic Lake Nam Co shows a terminal DOM signature suggested also for the open ocean. This diversity enables DOM to function as a monitoring agent in catchments and the lake and thus be a helpful tool to safeguard sensitive, multiply threatened High Asian ecosystems.

Introduction

The Tibetan Plateau (TP) comprises not only the largest connected alpine pasture system of the world (Miehe et al., 2019) linked to a rich cultural and historical heritage (Gongbuzeren et al., 2018). It also is known as Asia's water tower (Bandyopadhyay, 2013), forming the source area of several large river systems, providing freshwater resources to billions of people in east and south-east Asia. Large quantities of the water storage are located in the ice masses of the Himalaya and the TP, forming the largest frozen freshwater reservoir outside the Polar Regions. This third pole environment is well investigated since several decades (Qiu, 2008; Yao et al., 2012), revealing that High Asia's ecosystem services are threatened (Hopping et al., 2018) by climate change (Yao et al., 2019) and the intensification and changes in land-use patterns (Harris, 2010). The quality of water resources is of utmost importance for the TP and for societies downstream in Asia. DOM is a viable tool to investigate and detect changes in water quality (Li et al., 2021). Consequently, the analysis of DOM dynamics on the Tibetan Plateau (TP) is an important cornerstone to safeguard this remote and sensitive high-altitude ecosystem.

DOM can connect fluvial ecosystems over several hundreds of kilometres (Seidel et al., 2015), or connect terrestrial and aquatic biomes, for example upland pastures with lacustrine environments (Goodman et al., 2011). Changes in catchment properties might therefore trigger adverse effects in distanced and fragile ecosystems (Wilson and Xenopoulos, 2009), showing that action and response of land-use changes or climatic change might be temporally and spatially detached (Goes et al., 2020; Roulet and Moore, 2006). The molecular diversity and complexity of dissolved organic matter calls for the employment of advanced techniques (Fellman et al., 2010; Nebbioso and Piccolo, 2013). Since the mid-1990s Fourier transform mass spectrometry has begun to develop with accelerated pace (Hawkes et al., 2020). This has led to the development of electrospray ionization Fourier transform ion cyclotron resonance mass spectrometry (ESI FT-ICR MS), an ultra-high resolution mass spectrometry method that allows to identify thousands of molecular formulae in samples of natural organic matter, beforehand only characterised by spotlighting proxy-methods or laborious laboratory setups. This offers a unique opportunity to deepen the insights into DOM characteristics, dynamics and fate, especially in coupled multi-methodological approaches such as presented by Stubbins et al. (2014) and Herzsprung et al. (2017). Employing ultra-high resolution mass spectrometry might render the notoriously complicated task of interrelated critical zone assays, as described by Kayler et al. (2019) to ease. Current advancements have made sample preparation (Dittmar et al., 2008), analytical treatment (Merder et al., 2020) and data evaluation (Bramer et al., 2020) of FT-ICR MS applications much easier and by this more widely applied (Minor et al., 2014).

Here, we used FT-ICR MS to gain information about characteristics and processing of stream DOM of different sites, endmembers, such as glaciers, springs and alpine wetlands and DOM of lake Nam Co on the TP. As a study site, we have chosen the Nam Co catchment which is located in the southern branch of the TP (Figure 1) with an area of 10,776 km². To the south east, the Nyainqentanglha mountain ridge, with elevations of up to 7162 m. asl. delimits the catchment, while the north and western margin of the catchment are stretching in an increasingly arid zone. The Nam Co catchment lies in the transition zone of the alpine pasture dominated by the plagioclimax sedge *Kobresia pygmaea (K. pygmaea)* (Miehe et al., 2008) and the alpine steppe, expanding towards the north of the TP. The catchments unique positioning in the transition zone is expressed between the south, comprising alpine pasture with a more humid, glacial-influenced high-mountain ecosystem contrasting to the hilly northern margin of the catchment, with more arid climate and alpine steppe dominated vegetation, where clear signs of pasture degradation (Maurischat et al., in prep.) are visible. The climate is biannually shaped with cold and dry winters and warm and moist summers influenced by the Indian summer monsoon (Anslan et al., 2020). The endorheic Lake Nam Co forms the terminal aquatic basin of the catchment. Lake Nam Co and its catchment are relatively well investigated with respect to the status and dynamics of its alpine vegetation and the processes in the glacial sphere (e.g. Anslan et al., 2020; Bolch et al., 2010; Gao et al., 2015; Wang et al., 2020; Kai et al., 2019; Adnan et al., 2019a), while DOM characteristics and especially its transformation were only investigated to a limited extent by studies of Spencer et al. (2014) and Li et al. (2021). This leaves open questions such as: how diverse DOM signatures are influenced by the diverse biotic and abiotic inventory of the catchment, and which processing DOM undergoes during the fluvial pathway and in the lake. Its variability makes the Nam Co catchment a functional sentinel for changes and a natural laboratory, suited to test whether DOM characteristics can be employed as a precursor for changes in this sensitive alpine environment, representing vast parts of the TP. We hypothesized that 1) investigated catchments of Nam Co differ with respect to the chemical composition of DOM. 2) Investigated endmembers possess unique DOM signatures compared to the DOM of streams and 3) DOM signatures of lake water are chemically distinct compared to the DOM of different sources in the catchments and further suggest a processing of DOM between catchments and lake.

Materials & Methods

Site description

The Nam Co catchment has a total extend of 10789 km^2. In general, two main climate zones can be distinguished: The south of the catchment is delimited by Nyainqentanglha mountain ridge with highest elevations of more than 7000 m. asl. Higher elevations of the mountain ridge are glaciated (Bolch et al., 2010), glacial meltwater is the dominating water source of the streams draining the catchment here (Adnan et al., 2019b). Sparse vegetation dominates the glacial zone, while *K. pygmaea* pastures are developed at lower elevations. Close to the lake shore and lake terraces, alpine steppe vegetation is developed (Nieberding et al., 2021). The southern part of the catchment is characterised by higher precipitation compared to the north. Up to 530 mm are measured in the Nyainqentanglha mountains (Anslan et al., 2020).

The northern margin of the catchment is dominated by hilly uplands with a less steep relief (Yu et al., 2021), here lower annual precipitation of around 300 mm is reported (Anslan et al., 2020). Alpine pasture is developed on north-exposed hill-flanks and in the valley bottoms, while alpine steppe grows on less well insulated south exposed flanks and in the upland, as well as at the lake shoreline (Maurischat et al., in prep.). A degradation and aridity gradient is visible between the southern and the northern border of the catchment (Anslan et al., 2020).

Climate of the Nam Co catchment is developed biannual. The summer months are dominated by the Indian Summer Monsoon which usually lasts from May until late August/ early September (Chen et al., 2019). During this phase, warm and moist air is transported from the south towards the TP. More than 80% of the annual precipitation falls during the monsoon season and the mean day-time air temperature increases to 11°C (Chen et al., 2019). During winter and spring, the Westerlies dominate the climate of the Nam Co catchment. The climate is comparably dry and cold with minima of around -20°C between December and February and only few precipitation events (Nieberding et al., 2021).

The endorheic Lake Nam Co with an elevation of 4726 m. asl. has a total size of 2000 km^2. The lake water is oligotrophic and saline (Keil et al., 2010). The dimictic Lake Nam Co has a depth of 99 m, is well supplied with oxygen and has a clear water column (Wang et al., 2020).

Three catchments within the Nam Co catchment were chosen to best represent the diversity of natural variance in the catchment. The selected catchments differ with respect to plant cover, dominating vegetation type, steepness and exposition, aridity and glaciation.

The Niyaqu catchment in the east of the Nam Co catchment has a total area of 406 km^2. Two streams drain the catchment, of which the southern stream receives glacial meltwater from a comparably small glacial outcrop of the Nyainqentanglha located 700 m above the lake level. This river runs through extensive alpine pastures and feeds a large alpine wetland located in a depression (Figure 1). The northern stream drains a hilly upland area in the transition of alpine steppe and alpine pasture. Most parts of the Niyaqu catchment are hilly, permanent settlements are located close to the lake shore and herding of yak is conducted throughout the year.

At the southern margin of the Nam Co catchment, the Qugaqie catchment represents the high-altitude zone associated with the Nyainqentanglha mountain ridge. The comparably small (58 km²) catchment is characterised by its steep relief and a valley course in south-north direction (Keil et al., 2010). The altitudinal difference between the lake and the highest point of the catchment is more than 2200 m. Due to harsh winter conditions, this catchment is used as summer pasture. One major streams drains the catchment and passes through different vegetation zones, including *K. pygmaea* pastures and alpine steppe in the lower realms of the catchment.

The more arid north of the Nam Co catchment is represented by the Zhagu catchment located in the far north-east. The smallest investigated catchment has a size of 46 km² and is mostly characterised by hilly upland relief (Keil et al., 2010). Consequentially, there is no active glacial influence and only a small altitudinal gradient in this catchment, with the highest elevation at 5230 m. asl. Two small streams drain the catchment, both fed by springs. During our investigation the catchment was arheic. This catchment shows clear signs of degradation of alpine *K. pygmaea* pastures by remaining sods of turf. Barren land and sparse alpine steppe vegetation is developed in the valley bottom. The hilly upland and the stream beds are used for animal husbandry.

Water samples from streams of three catchments and the Lake Nam Co were taken during a field campaign in September 2019. Sampling locations were chosen, to follow the stream path until the stream mouth into Lake Nam Co. Further, special attention was given to endmembers such as glaciers, springs and alpine wetlands which were also sampled directly. In order to cover both, the transformation of DOM signatures along the stream path, as well as the DOM characteristics of potential endmembers.

We identified three endmember groups (glacial effluents, springs and alpine wetlands) and three additional sampling units (streams, brackish water and lake water), resulting in six sample categories (Figure 1). Glacial effluents were drawn directly at or close to the glacial terminus, while spring water samples were drawn directly or close to springs. Alpine wetlands samples are taken from the standing water column of areas characterised by bogs and aquatic plants. Stream samples are characterised by fast flowing water in the stream bed, embedded in the azonal extensions of the *K. pygmaea* biome (Figure 1) along the stream path. Brackish water samples were taken in the mixing zone of stream and lake water at the stream mouth at the shoreline of Lake Nam Co. Samples taken off the shore in the lake are supposed to characterize DOM of the terminal aquatic basin.

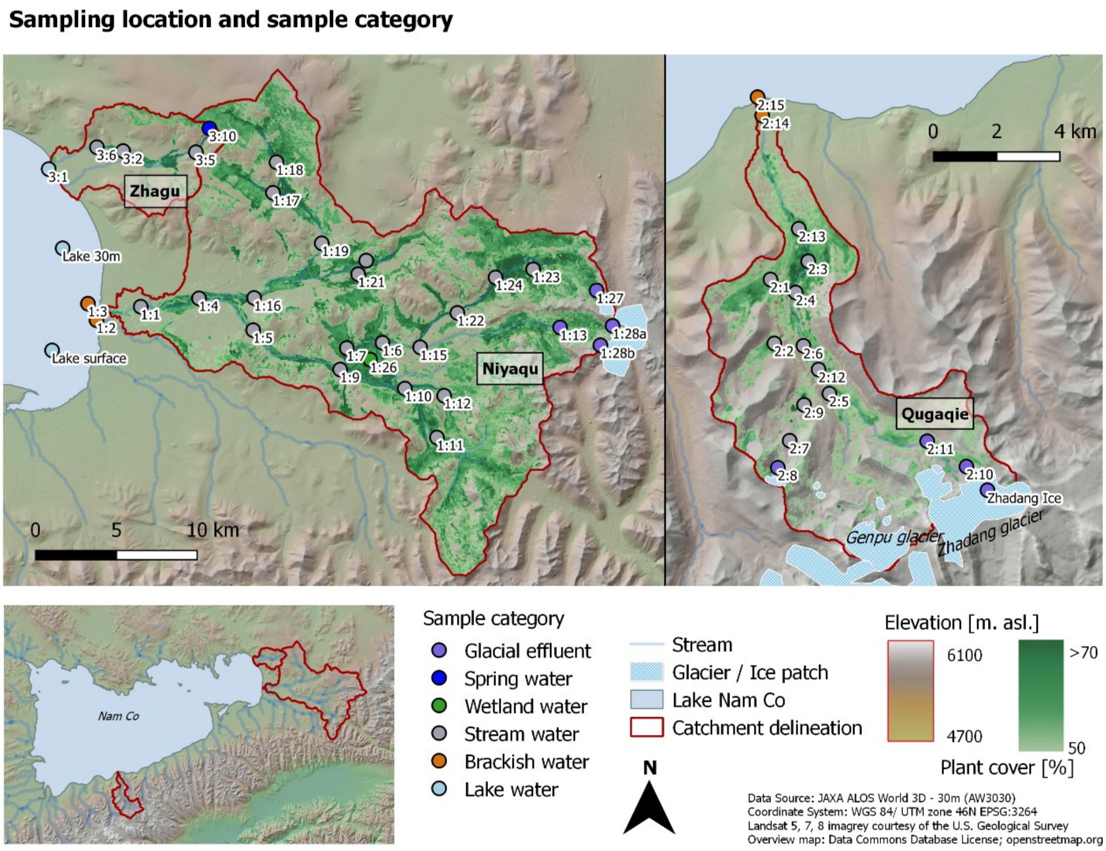

Figure 1: Overview map of the investigated catchments and sampling locations with sample category affiliation. Plant cover estimations are taken from Maurischat et al. (in prep.) and represent the extend of K. pygmaea pastures.

Water Sampling

Samples were taken from the middle of the stream profile by the use of a telescopic sampling device. Lake waters were sampled from the lake surface and from 30 m depth by use of submersible sampling device. All water samples were taken as seven subsamples with a volume of one litre each, mixing these and taking 500 mL of the latter, discarding the rest. Samples were stored in pre-cleaned high-density polyethylene bottles (Rotilabo, Carl Roth, Karlsruhe, Germany). They were directly filtered on-site by the commonly accepted 0.45 µm threshold of DOM using a polyethersulfone (PES) membrane (Supor, Pall, Port Washington, USA) and a portable electric pump. Samples were then acidified to pH 2 by use of hydrochloric acid (32%), controlled with litmus paper, transported to the NAMORS (Nam Co Monitoring and Research Station for Multisphere Interactions) research station and frozen at -21°C until later analysis.

Solid Phase Extraction

Solid phase extracted DOM (SPE-DOM) was separated following Dittmar et al. (2008). For that, DOM samples were thawed and, if necessary, re-acidified to pH 2 using 32 % HCL (Rotipuran p.a., Carl Roth, Karlsruhe, Germany). Initial dissolved organic carbon (DOC) content was measured from 20 mL of aqueous sample by high-temperature oxidation on a total organic analyser (varioTOC Cube, Elementar, Langenselbold, Germany). DOM samples were then diluted with ultrapure water to a concentration of 1.5 mg L^{-1} DOC, and 250 mL of diluted sample was used for extraction, not exceeding the threshold of 2 mg C per g absorber for SPE cartridges. Cartridges with 100 mg of styrene divinyl benzene polymer (PPL) resin (Bond Elut, Agilent Technologies, Santa Clara, USA) were used for extraction. Cartridges were rinsed with 3 mL of ultrapure water and soaked with 3 mL of analytical grade (≥ 99.9%) methanol (Chromasolv, Honeywell, Seelze, Germany) overnight, prior to DOM extraction. 250 mL of diluted DOC sample were passed over the absorber at a flow rate of ~5 mL min^{-1} in temperature controlled laboratory environment, ensuring reproducible retention and comparable carbon load per cartridge. 20 mL 0.01 M HCl was used to rinse inorganic compounds from cartridges after passing the sample. The resin cartridges were then dried under a constant flow of ultrapure N_2 (N_2 5.0, Linde, Dublin, Ireland) until dried. Finally, two cartridge volumes of methanol were used to elute the sample from the PPL columns. The samples were transferred in cauterized brown glass flasks (Neochrom, Neolab Migge, Heidelberg, Germany) sealed with polytetrafluorethylen caps (Neochrom, Neolab Migge, Heidelberg, Germany) and stored at -18°C until further analysis. Extraction efficiency was assessed by drying 0.2 mL of SPE-DOM under an N_2 atmosphere and resolving the aliquot in 20 mL of ultrapure water. The samples were then analysed for the total C content by high-temperature oxidation (varioTOC Cube, Elementar, Langenselbold, Germany) and the volumetric proportion of used DOM to extracted SPE-DOM was calculated. As pointed out by Spencer et al. (2014), low initial DOC concentrations, as occurred in some of our samples, can hamper an accurate calculation of extraction efficiency, hence process standards were used to ascertain that a functional extraction happened. Blank samples with ultrapure water instead of sample material did undergo the whole extraction process to receive information about process-induced contamination in SPE-DOM samples. Reference material (Suwannee River / IHSS) (Green et al., 2015) was SPE extracted and compared to routine assays, to ensure validity of results.

Fourier Transform Ion Cyclotron Resonance Mass Spectrometry

SPE-DOM methanol isolate samples were diluted in 1:1 methanol/ultrapure water to a final concentration of 5 mg C L^{-1} for analysis. Samples were then analysed in a Solarix XR 15 Tesla FT-ICR MS (Bruker Daltonik, Bremen, Germany). Duplicate sample electrospray ionization (ESI) was carried out in negative mode at a flow rate of 122 µL h^{-1}. 200 broadband scans (masses of 92.14 to 2000 Da) were acquired per sample, accumulation time was 0.2 sec per scan. Technical detection limits published by Riedel and Dittmar (2014) were used. Data processing followed the descriptions published by Merder et al. (2020). Mass accuracies were within an error of ≤ 0.2 ppm. Signal to noise ratios > 4 were accepted to bear meaningful chemical information (Koch et al., 2007) and masses were saved. Contaminants were deleted, when identified as such with the contaminant reference mass list. Detection limits for peaks were normalised to sample peak intensities. Overall peak intensities were scaled to the local sample maxima by using the sum of peaks. Isotope (^{13}C, ^{18}O, ^{15}N, ^{34}S) mass effects were corrected to the most abundant form of the respective isotopologue. Peaks with molar ratios of O:C = 0, O:C ≥ 1, and H:C > 2.5 were removed. Internal calibration standards from the Hawaii DOM ocean time-series (HOT) reference mass list (Repeta, 2015) were used to assign *m/z* peaks to molecular formulae. Duplicate samples were normalised by adding up molecular intensities per formulae and then dividing by the number of replicates, molecular formula were retained only, when present in both duplicate samples. The Hawaiian deep sea NELHA standard (Green et al., 2014) was used as an in-line calibration to measure machine drift.

Molecular formulae were further assigned to molecular compound classes. The modified aromaticity index (Ai_{mod}) and double bond equivalents index (DBE), both representing aromaticity in DOM, were calculated for each formula as proposed by Koch and Dittmar (2006) and Koch and Dittmar (2016). The Ai_{mod} was calculated as:

$$Ai_{mod} = (1+C - 0.5O - S - 0.5H) / (C - 0.5O - S - N - P) \quad [1]$$

Ai_{mod} indices > 0.5 were assigned as aromatic, while indices ≥ 0.67 were considered as condensed aromatic structures. DBE was calculated with:

$$DBE = 1 + \frac{1}{2}(2C - H + N + P) \quad [2]$$

The original compound classification was suggested by Šantl-Temkiv et al. (2013), compound class names were modified according to the descriptive classes used by ICBM-OCEAN (Merder et al., 2020) (Table 1). We indicated here, that FT-ICR MS is a mass based analysis not allowing for discrete, but a statistical assessment of molecular formulae allowing for suggestions of structural composition (Leyva et al., 2020).

Table1: Assigned descriptive compound classes for FT-ICR MS formulae (Merder et al. 2020)

Descriptive compound class	**Definition**
Aromatic O-rich (AROR)	$Ai_{mod} > 0.5$ AND $O/C > 0.5$
Aromatic O-poor (AROP)	$Ai_{mod} > 0.5$ AND $O/C \leq 0.5$
Highly unsaturated O-rich (HUROR)	$Ai_{mod} < 0.5$ AND $H/C <1.5$ AND $O/C > 0.5$
Highly unsaturated O-poor (HUROP)	$Ai_{mod} < 0.5$ AND $H/C <1.5$ AND $O/C \leq 0.5$
Unsaturated O-rich (UROR)	$H/C \geq 1.5$ AND $H/C \leq 2$ AND $O/C > 0.5$
Unsaturated O-poor (UROP)	$H/C \geq 1.5$ AND $H/C \leq 2$ AND $O/C \leq 0.5$
Unsaturated with nitrogen (USN)	$H/C \geq$ AND $H/C \leq 2$ AND $N > 0$
Saturated (SAT) = SOR + SOP	DBE = 0
Saturated O-rich (SOR)	DBE = 0 AND $O/C > 0.5$
Saturated O-poor (SOP	DBE = 0 AND $O/C \leq 0.5$

Statistics

Molecular formulae in each sample were intensity weighted by normalising with the sum of total signal intensity. Formula intensities were then rescaled between 0 and 1 and expressed as percent. Van Krevelen diagrams (Merder et al., 2020) were produced for samples, blanks and reference standards (Supplementary materials, Figure S1). The analysis was conducted by grouping with two independent factors. 1) The three catchments: Niyaqu, Qugaqie, Zhagu and Lake Nam Co and 2) sample categories: including the endmembers (glacial effluents, spring and wetland) and stream water, brackish water and lake water. Two endmember groups, spring and wetland, as well as one site group: Lake Nam Co were exempted from statistical analysis due to small sample size. Bar plots for the relative contribution of compound classes were produced, showing mean and standard deviation (as error bar) for all independent factor groups. Further, means and standard deviation were calculated for Ai_{mod}, number of formulae with Ai_{mod} >0.5, number of formulae with Ai_{mod} >0.67, mean DBE, and number of formulae containing the heteroatoms nitrogen (N), phosphorous (P) and sulphur (S), as well as the total number of assigned formulae.

Data were tested for Gaussian distribution and homoscedasticity by application of Kolmogorov-Smirnoff test and Levene test (Brown and Forsythe, 1974). Due to violations of normal distribution in combination with different sample sizes per group, parametrical test are considered unstable (Bortz and Schuster, 2010). Multiple pairwise comparisons were conducted using Kruskal Wallis test and

Mann Whitney test (Birnbaum, 1956) for pairwise comparisons with Bonferroni post-hoc correction for multiple testing. Significance levels (α) of 0.05 were set as threshold (Supplementary materials, Table S1 and Table S2).

Non-metric multidimensional scaling (NMDS) was used as dimensionality reduction and ordination approach (Anderson et al., 2006; Faith et al., 1987). For that, data for NMDS were scaled and mean-centred (Jolliffe, 2002). NMDS is an established ordination technique also in the field of FT-ICR MS (Derrien et al., 2017; Lu et al., 2015). Here, dissimilarities in samples, represented by compound classes, indices and formula counts were used to calculate the ordination distance between samples. NMDS was conducted for both independent factors (site and sample category). The Euclidean dissimilarity index and k=3 were used for NMDS. Scores, R^2-coefficients of determination, and loading tables are provided in the supplementary materials (Table S3-S4). R (The R project for statistical computing, v3.6.3, GNU free software) was used to calculate statistics. The R base packages (R Core Team, 2013) and 'tidyverse' (Wickham et al., 2019) were used for data organisation and pre-processing, as well as non-parametric statistics. 'ggplot2' (Wickham, 2009) was used for graphical illustration and 'vegan' (Oksanen et al., 2020) was used for ordination with NMDS.

Results

Sample Treatment

Extraction efficiency was calculated per sample and was over all samples 61.4 in mean ± 18.6 % standard deviation (Supplementary materials, Table S3). Extraction efficiency was variable between samples, although all treatments were carried out in control laboratory environment. Spencer et al. (2014) report that extraction efficiency could not be calculated for their High Asian sample set, due to low initial DOC concentrations in samples. This hints at the fact that technical limitations concerning analysis of liquid phase samples with high-temperature oxidation in TOC analysers might be responsible for variable extraction efficiencies rather than malfunctions of the robust and well-established solid phase extraction routine. The Suwannee River / IHSS process standard and the mean efficiency of our SPE extraction is within the expectancy range of 60% from published data (Dittmar et al., 2008; Green et al., 2014), making a functional extraction process most probable.

Group Counts and Statistics

The total number of 48 samples (excluding 6 control and standard samples) was grouped by two independent factors. Site, describes the sampling location of the sample, either one of the three investigated catchments Niyaqu, Qugaqie, Zhagu, or the Nam Co Lake. The independent factor sample category, groups the samples in six heuristic classes: the three endmembers (glacial, spring and wetland waters) and stream, brackish and lake water. The total number of assigned formulae decreased in the order Qugaqie > Zhagu > Niyaqu > Lake Nam Co for water samples from sites, and Lake Nam Co had only half the number of assigned formulae compared to the catchments (Table 2). For the heuristic classes, water samples from wetland and brackish environments have highest numbers of assigned formulae, followed by glacial effluents, spring and stream water, while lake water samples shows the least molecular diversity.

The heteroatoms (N, S, P) have differing abundances. Lake Nam Co had only half as many N containing molecular structures compared to the three catchments, but relatively more N-heteroatoms than the three catchments (Table 2). Sulphur containing formulae were uniformly distributed between the four site groups. Phosphorous, on the opposite, was fewest encountered in lake water, while significantly more P-containing molecules were found in samples from the Qugaqie catchment compared to the Niyaqu and Zhagu catchment when assessing absolute and relative values (Table 2, Table S1).

For the heuristic sample category, N-containing molecules were most abundant in wetland water and brackish water samples. Spring, stream and glacial effluents formed a group of medium N distribution and lake samples had the lowest count of formulae associated with N. For S, brackish and wetland samples again shows the highest count, followed by the other four groups with comparable amounts. Molecular formulae containing P were least abundant in samples from spring water, followed by the order lake < wetland < stream < glacial < brackish water (Table 2).

Concerning the mean Ai_{mod}, mean DBE and count of aromatic ($Ai_{mod} > 0.5$), and condensed aromatic ($Ai_{mod} \geq 0.67$) formulae, similar differences are visible for samples from the three catchments and Lake Nam Co, suggesting a positive correlation of these variables. DOM of Lake Nam Co shows by far least values for all four variables, while samples from Niyaqu, Qugaqie and Zhagu exhibit no distinct differences (Table 2). When samples are grouped to sample categories, lake water samples show lowest aromaticity indices (Ai_{mod}, DBE) and low proportions of aromatic or aromatic condensed structures. Glacial effluents show second lowest aromaticity indices, followed by stream, spring and wetland DOM, the latter three forming a uniform group. Brackish water shows the highest DBE and largest numbers in assigned aromatic and condensed aromatic formulae, while the mean Ai_{mod} is in line with the aforementioned group of samples (Table 2).

Assigned compound classes (Table 1 and Figure 2) allow a general overview about the composition of assigned molecular formulae and elemental composition in DOM of sample groups. For the independent factor site, differences were detected in aromatic compounds. Lake Nam Co has five times fewer aromatic oxygen (O)-rich molecular classes compared to the three catchments. For aromatic O-poor compounds, the lake has even 70 times fewer compound abundance, and for highly unsaturated O-rich molecules Lake Nam Co showed overall lowest values but still relative abundances of 40%. Within catchments, significant differences exist for the relative abundance of highly unsaturated O-rich formulae between Qugaqie and Zhagu and between Qugaqie and Niyaqu, with Qugaqie featuring the highest relative abundances. When assessing highly unsaturated O-rich compounds, the Lake Nam Co samples show higher values compared to DOM from the investigated catchments. This compound class was the most abundant one in lake DOM. Also, significantly more highly unsaturated O-rich molecules were encountered in Niyaqu compared to the samples drawn in the Qugaqie catchment. For the compound class of unsaturated O-poor molecules overall abundances were low, but Qugaqie had significantly higher abundances than Zhagu. The group of unsaturated O-rich molecules again was highest in Lake Nam Co DOM followed by Niyaqu > Qugaqie > Zhagu. The elemental group of unsaturated N containing molecules mostly shows higher abundances in the Nam Co Lake compared to the catchment samples. Saturated molecules are reported collectively for groups with an $O/C > 0.5$ and $O/C \leq 0.5$. Relative abundances were overall low, but higher in samples drawn from Niyaqu and Zhagu compared to Qugaqie and Lake Nam Co (Figure 2).

Table 2: Overview on mean and standard deviation of aromaticity and DBE proxies and mean and standard deviation of formulae counts for sites.

Variable	**Niyaqu**		**Qugaqie**		**Zhagu**		**Lake Nam Co**	
	Mean	***SD (±)***	***Mean***	***SD (±)***	***Mean***	***SD (±)***	***Mean***	***SD (±)***
Mean Ai_{mod}	0.33	0.05	0.32	0.02	0.34	0.04	0.24	0.01
Number of formulae Ai_{mod} >0.5	622	324	708	399	705	269	112	117
Number of formulae Ai_{mod} >0.67	163	103	174	124	183	85	14	8
Mean DBE	9.4	1.0	9.5	0.9	9.8	0.9	7.5	0.8
Number of formulae containing S	143	112	122	97	141	53	109	123
Number of formulae containing N	1412	941	1841	1103	1835	852	725	790
Number of formulae containing P	69	96	139	103	47	37	20	25
Total number of formulae	2867	1060	3393	1376	3218	857	1394	1115

Table 2 continued: Overview on mean and standard deviation of aromaticity and DBE proxies and mean and standard deviation of formulae counts for sample categories (including endmembers) ‡ indicates sample size n=1, no standard deviations are given then.

Variable	**Glacial effluent**		**Spring ‡**		**Wetland ‡**		**Stream water**		**Brackish water**		**Lake water**	
	Mean	***SD (±)***	***Value***		***Value***		***Mean***	***SD (±)***	***Mean***	***SD (±)***	***Mean***	***SD (±)***
Mean Ai_{mod}	0.31	0.04	0.25		0.33		0.33	0.05	0.35	0.02	0.25	0.02
Number of formulae Ai_{mod} >0.5	556	262	172		895		632	314	1135	458	172	133
Number of formulae Ai_{mod} >0.67	125	78	23		255		162	96	317	149	23	17
Mean DBE	9.1	1.0	7.8		9.7		9.4	0.9	10.4	0.7	7.8	0.7
Number of formulae containing S	103.	89	133		291		126	92	230	159	133	96
Number of formulae containing N	1548	1134	964		2549		1480	923	2586	1230	964	696
Number of formulae containing P	133	125	30		68		75	88	162	126	30	25
Total number of formulae	2965	1132	1738.		4091		2934	1062	4492	1638	1738	988

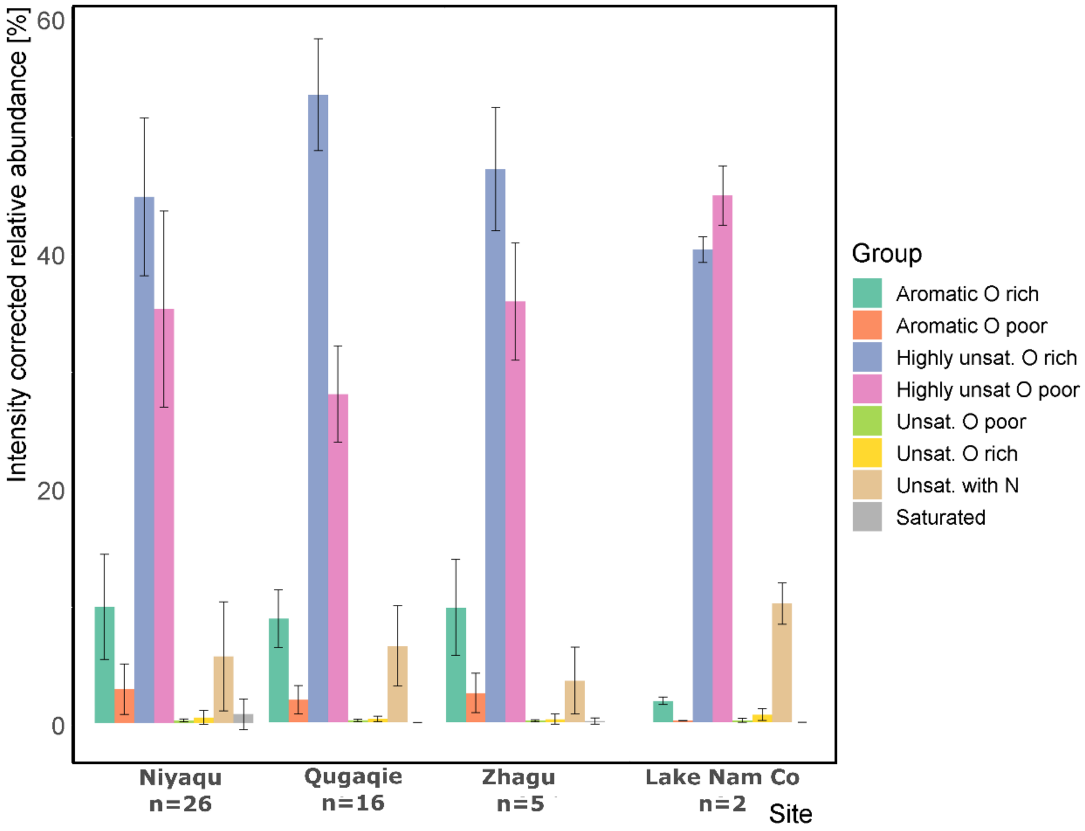

Figure 2: Mean relative intensity weighted counts of descriptive compound classes for stream waters of the three catchment and Lake Nam Co. Error bars indicate standard deviations.

For the sample categories including endmembers, assigned compound classes were as well analysed. Overall, glacial, wetland, spring and brackish waters show elevated values compared to lake DOM. Lake water samples contained one fourth of aromatic O-rich compounds compared to stream water samples. For aromatic O-rich compounds, large differences exist between lake water and water samples assigned to other sample categories, with samples from Nam Co having smallest abundances in this class. Highly unsaturated O-rich samples also differ between sample categories. Glacial effluents and spring waters show largest abundances in this class, while wetland waters and samples from Lake Nam Co show the smallest relative values. A different picture is visible for highly unsaturated O-poor molecular compounds. Water samples from the Nam Co Lake show highest relative abundances compared to all other groups, especially glacial effluents had 40 percent lower relative abundances in this class compared to lake water.

Unsaturated O-poor compounds were more abundant in stream, glacial and wetland waters compared to spring, brackish and lake waters, while unsaturated O-rich compounds were more frequent in lake waters and glacial effluents compared to the wetland and stream and brackish categories. Spring water shows the least compounds in unsaturated O-rich compounds, being almost

20 times lower compared to water from Lake Nam Co. Unsaturated molecular formulae containing N show a similar picture. Lake and glacial DOM were relatively rich in these molecules, followed by stream, brackish and wetland waters which show 50% less abundance in unsaturated N containing formulae compared to Lake DOM. Spring water again shows smallest intensity weighted formulae in this compound class, being 75% lower compared to Lake Nam Co (Figure 3). Saturated compounds were overall low, higher values were encountered in stream, spring and brackish waters.

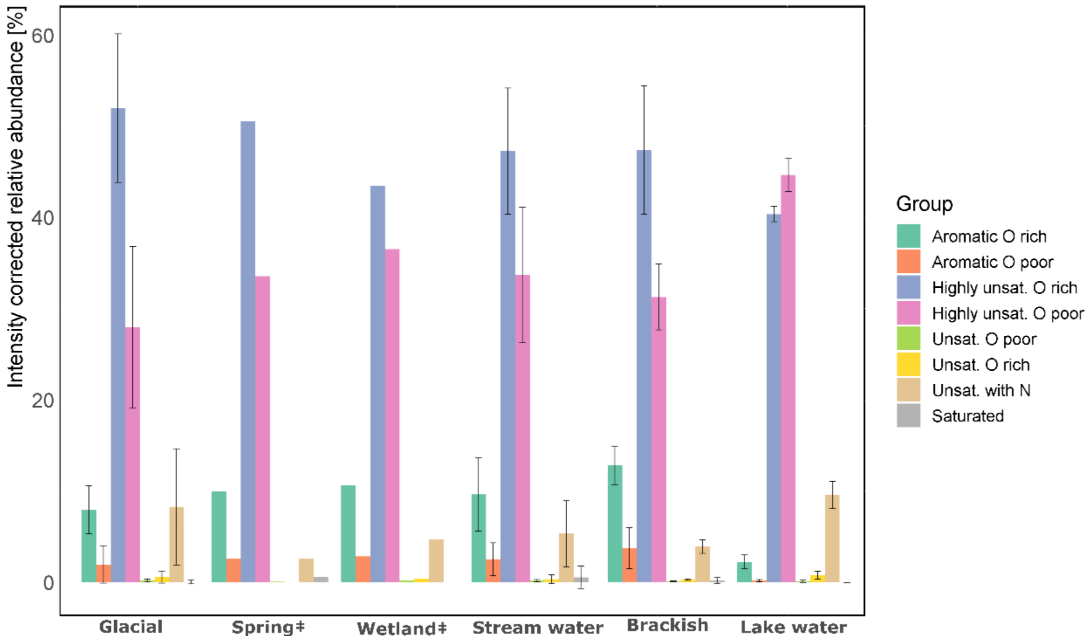

Figure 3: Mean relative intensity weighted counts of descriptive compound classes sorted for sample categories. Error bars indicate standard deviations. ‡ indicates sample size n=1, no standard deviations are given then.

NMDS Ordination

NMDS ordination was conducted for both independent factors (sites and sample categories including endmembers). Notably, ordination results remain unchanged by changing independent factors but these help to further resolve structures in data. The stress value of the NMDS is a goodness of fit measure for ordination. The stress value of 0.08 (Figure 4, 5) for this ordination is in line with accepted stress measures (< 0.2; Dexter et al., 2018) showing that data are positioned undistorted in the ordination plane.

The sites are depicted as colour information in the NMDS ordination (Figure 4). The ordination shows a clustering of the Lake Nam Co samples in the top-right of the plane. Water samples of the Zhagu catchment are mostly located on the left side, while water from Qugaqie locate in the lower centre and lower right side of the dimension plane. Water samples from the Niyaqu catchment are not uniformly positioned, but cluster in different areas of the ordination, forming three cores (top right, bottom right and the largest group in the middle to top left). Two dimensions are presented for NMDS ordination. In the positive direction of dimension 1, displayed on the horizontal axis, all unsaturated compound classes are resolved, while in the negative direction aromatic compound classes and Ai_{mod} and DBE indices are loading. The positive direction of dimension 2, displayed on the vertical axis, distinguishes samples with high abundances in highly unsaturated O-rich and saturated O-poor compounds, while the negative direction clusters samples with high amounts of heteroatoms and saturated O-rich and highly unsaturated O-poor compound classes.

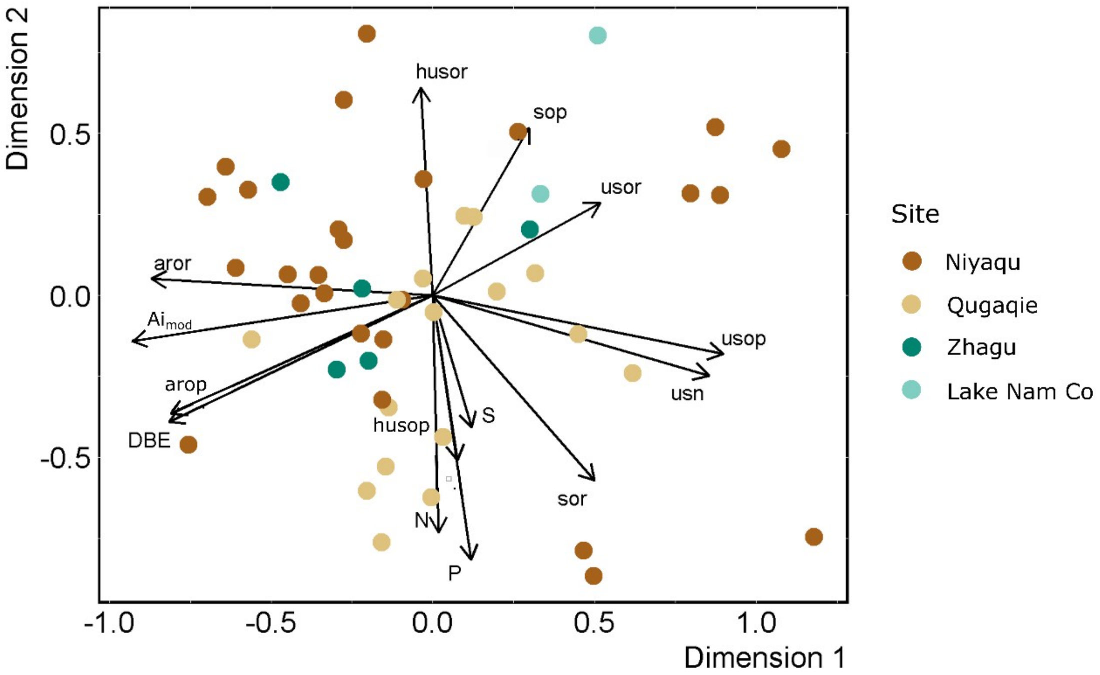

Figure 4: Non-metric multidimensional scaling of descriptive compound classes (abbreviations see Table 1), counts of heteroatoms (N, S, P) and aromaticity indices (Ai_{mod} & DBE), resolved for the independent factor site. Stress 0.08, k=3, dissimilarity index = Euclidean.

The independent variable for sampling categories allows a more refined understanding of clustering characteristics (Figure 5). Stream water samples were scattered over a wide plane of the ordination, while lake water samples and brackish samples are distinctively placed. The former are located in the top-right, while the latter are diametric in the bottom-left. The spring and wetland sample plot close to the centre of the ordination, where the mean of all samples are located, indicating compositional mixing. Glacial effluents furthermore are scattered over the ordination space from the top left to the bottom right, indicating a large molecular diversity in water samples of glacial origin, here the two sampled glaciers from Niyaqu and Qugaqie are distinct from each other.

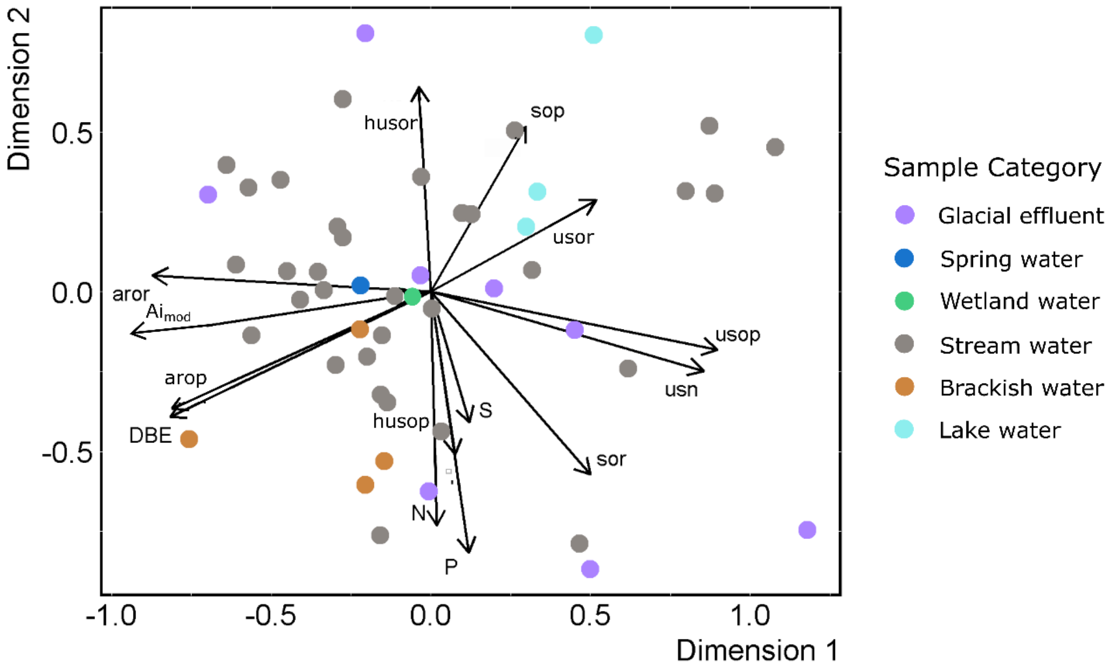

Figure 5: Non-metric multidimensional scaling of descriptive compound classes (abbreviations see Table 1), counts of heteroatoms (N, S, P) and aromaticity indices (Ai$_{mod}$ & DBE), resolved for the independent factor sampling category including endmembers. Stress 0.08, k=3, dissimilarity index = Euclidean.

Discussion

DOM Composition in Three Catchments of the Lake Nam Co Watershed

The three investigated catchments of the Nam Co Lake were analysed for undirected FT-ICR MS of solid phase extracted DOM. Between the catchments, differences were visible in the composition of DOM. Pronounced differences exist between the Qugaqie catchment compared to Niyaqu and Zhagu. The Qugaqie catchment shows the largest number of chemical formulae identified, together with an increase in formula containing P heteroatoms. Furthermore, unsaturated O-poor and highly unsaturated O-rich molecules were relatively enriched compared to the other catchments, while highly unsaturated O-poor formulae were depleted. Unsaturated O-rich compound classes indicate a larger share of molecular structures resembling readily bioavailable compounds. While complex polyphenolic biopolymers, like tannins (HUSOR) were enriched compared to highly unsaturated O-poor compounds more associated to lignin-like compounds. This shows, that higher complexity compounds originating from plants and soils dominate the DOM of Qugaqie and hints towards a larger microbial breakdown of less complex biopolymers, relatively leaving more complex highly unsaturated O-rich molecules behind and by this enriching them, as was shown in incubation studies (Mann et al., 2015). The Qugaqie catchment is mostly defined by its steep altitudinal gradient (>2000 m) and its glacial influence, with glacial meltwater as the dominating water source (Bolch et al., 2010; Buckel et al., 2020; Gao et al., 2015). As was pointed out by Spencer et al. (2014) and Hood et al. (2009) microbial-derived compositional signatures of DOM are strongly enriched in glacial ecosystems and their downstream rivers. Our data concordantly suggest, that glacial DOM is chemically different compared to primarily precipitation and groundwater fed catchments (Niyaqu, Zhagu), also visible by the higher number of assigned formulae (Table 2). The higher relative abundance of low-molecular compounds in our samples underlines a microbial DOM source in glacial DOM and suggest primary production by the microbial biome as well as decomposition of plant- and soil-borne materials (Anesio et al., 2009). The high proportion of P heteroatoms further suggests that DOM of glacial origin is highly biodegradable and rich in primary plant nutrients, corroborating the notion that glacial DOM is relatively labile (Hood et al., 2009).

The contribution of glacial meltwater is smaller in the Niyaqu catchment and absent in Zhagu. Here, the streams were less turbid and flowing slower (Maurischat et al., in prep.). The main water sources are precipitation and groundwater (Adnan et al., 2019a; Anslan et al., 2020; Tran et al., 2021). For both catchments, the NMDS indicates a relative enrichment of aromatic compound classes and high indices of aromaticity (Ai_{mod} and DBE). Molecular diversity was lower in samples from Niyaqu and Zhagu compared to Qugaqie, here resulting in a relative higher abundance of aromatic compounds. Alongside with higher Ai_{mod} also the abundance of highly unsaturated O-poor formulae was higher in

Niyaqu and Zhagu compared to water samples from the Qugaqie catchment. This class suggests a high contribution of compounds that originate primarily from plants such as lignin and other complex phenolic biopolymers (Roebuck et al., 2018; Seifert et al., 2016). Overall, the molecular composition of DOM suggests a DOM source originating from plants and soils close to the streams (Mann et al., 2015). The sites show differences with respect to the molecular composition of DOM. The Niyaqu and Zhagu catchments located in the east of the Nam Co catchment are clearly dominated by DOM originating from plants and soils in the catchment underlining the existence of a terrestrial – fluvial pathway, while the high-altitude Qugaqie catchment also comprises a microbial DOM source potentially increasing the biolability of this DOM and highlighting the glacial – fluvial pathway.

Heuristic Sample Categories shed Light on DOM Molecular Diversity

Heuristic sample categories were assigned to water samples and represent geographic units (i.e. three endmembers: glaciers, springs, wetlands and stream, brackish and lake water samples. The data show, that molecular diversity and DOM characteristics can be pinpointed by assigning abundances of chemical compounds to these landscape units. The NMDS ordination (Figure 5) suggest, that glacial effluents are diverse in chemical composition, but can differ between glacial ecosystems, as was also suggested by Spencer et al. (2014) in their study of two Tibetan glaciers. Glacial effluents in our study comprise two different signatures. 1) High abundances in unsaturated compounds with nitrogen as well as a relative enrichment in unsaturated chemical compounds. Alongside with 2) aromatic and highly unsaturated compound classes. Glaciers are understood to receive compounds with high molecular complexity from aeolian deposition. Either such as condensed aromatics from the burning of fossil fuels (Takeuchi, 2002) or compounds uncondensed but rich in phenols, usually associated with vascular plants and soils (Singer et al., 2012). The microbial DOM derived from autotrophic microbial activity in the glacial biome was reported for glacial ecosystems worldwide and is a feature of autotrophic activity on the ice shield (Hood et al., 2009; Telling et al., 2011; Anesio et al., 2009). The reported coexistence of autochthonous and allochthonous DOM sources on glaciers (Feng et al., 2016; Singer et al., 2012; Spencer et al., 2014) make them a diverse and complex biome largely influencing downstream river DOM in the Qugaqie catchment.

The spring water sample was taken in the upland of the Zhagu catchment and mainly comprises aromatic and highly unsaturated chemical compounds. Molecular diversity in form of the total number of assigned formula of spring water was low compared to other sources and especially only few formulae containing N and P heteroatoms were present. Spring water is generally expected to inherit aquifer and catchment characteristics in its DOM signature (Yoo et al., 2020). On the one hand, groundwater aquifers function as large filters and usually are characterised by a low productivity in terms of microbial growth, potentially retaining natural organic substances (Weinstein et al., 2011). Shallow groundwater on the other hand, is in contact with soil organic matter and can transport dissolved soil-borne OM (Connolly et al., 2020). Spring DOM was characterised by few molecular formula, a large share of them containing S heteroatoms. Current research shows, that sulphide oxidation plays a considerable role in geochemical processes of the eastern Nam Co catchment (Yu et al., 2019). The elevated number of S heteroatoms likely represents both, chemical weathering, and together with the relative increase of saturated O-poor formulae, the background of low molecular organic compounds derived from microbial production in this case by the soil microcosm (Griebler and Avramov, 2015) of the shallow aquifer, producing microbial DOM rich in sulphur bridge bonds. The overall reduced molecular diversity her also responds to an enrichment in aromatic and highly unsaturated formulae in spring water. These refractory organic matter compounds, probably originate from soils around the spring. Spring DOM therefore constitutes the background DOM signature of the catchment, especially of terrigenous origin.

Wetland DOM sampling was conducted in an extensive alpine wetland of the Niyaqu catchment. Compared to the Suwannee river standard (Green et al., 2015), wetland DOM consists of more saturated formulae. Wetland DOM shows a high molecular diversity (>4000 assigned formulae) was rich in N and S heteroatoms and relatively rich in unsaturated O-rich and unsaturated N containing compounds. In a multiproxy study from the same site, wetland DOM was enriched in mineralized nitrogen and dissolved organic carbon compared to the surrounding streams (Maurischat et al., in prep.). Bai et al. (2010) and Zhang et al. (2020) reported that alpine wetlands contain large amounts of nutrients in the organic biomass and that a degradation of wetlands can pose a nutrient threat to downstream ecosystems. This underlines the high productivity in alpine wetlands here mirrored in high molecular diversity and relative high abundance of readily bioavailable compounds. Alongside, as wetlands are usually located in depressions (Anslan et al., 2020), further lateral input of water and DOM from other sources increases molecular diversity.

Stream samples cluster relative widely around the ordination space in the upper and lower left site (Figure 5). Most samples show a predominance of aromatic compounds either associated with highly unsaturated O-poor or highly unsaturated O-rich formulae, suggesting mainly an input of plant-borne compounds, such as lignin and tannin (Mann et al., 2015), corroborating findings of further studies (Maurischat et al., in prep.). Along the streams the *K. pygmaea* biome spreads as an azonal pasture extension (Figure 1, Maurischat et al., in prep.). Roebuck et al. (2020) point out that agricultural sites (including pastures) usually feature terrestrial inputs, likewise Lu et al. (2015) found supremacy of aromatic and highly unsaturated compounds in watersheds dominated by pastoral activity. The *K. pygmaea* biome is a large alpine yak pasture with potential influence of faeces, known for their high biolability (Du et al., 2021) suggesting low molecular composition, the scattering of stream samples can therefore be explained by changing input compounds from the pasture biome. These inputs are associated with relative changes in N-containing unsaturated formulae and saturated formulae likely originating from faeces influence and products of their microbial utilisation (Vega et al., 2020). Still, the allochthonous input from soil and vascular plant materials clearly dominates stream samples.

DOM Processing and Composition of Lake Nam Co and its Brackish Estuaries

The brackish mixing zones of streams and Lake Nam Co were investigated, as well as water samples of the lake from the water column surface and from 30 m depth. Brackish water samples show the overall highest molecular diversity, expressed by the largest number of assigned molecular formulae, highest number of heteroatoms and as well highest numbers of aromatic and condensed aromatic compounds. Brackish water samples were mostly dominated by aromatic and highly unsaturated O-poor formulae and by this retained the terrestrial input signal of streams and spring samples. van Dongen et al. (2008) describe brackish waters as a zone of gradual mixing. Several studies found a connection of terrestrial-derived DOM signatures from streams to be exported to marine systems and showed gradual mixing in the brackish and shelf zone (Benner et al., 2004; Dittmar and Kattner, 2003; Pettersson et al., 1997; Ruediger, 2003). We suggest, that the high molecular diversity in brackish samples represents both the overruling terrestrial input from catchment streams mixing with the DOM signature of lake water.

Lake water differed in its DOM composition compared to all other samples. This water was relatively enriched in unsaturated and saturated compounds, such as lipids and carbohydrates, compared to other samples, but depleted in condensed aromatic, aromatic and highly unsaturated O-rich formulae. Correspondingly, mean values of Ai_{mod} and DBE indices, representing the overall aromaticity of DOM (Koch and Dittmar, 2006; Koch and Dittmar, 2016) decreased as compared to respective values in the streams feeding the lake. In contrast, S heteroatom containing formulae were relatively abundant, while N and P heteroatoms were just slightly enriched compared to stream water samples. Lake Nam Co is a large, deep, oligotrophic lake with a clear water column (Wang et al., 2020). Microbial decomposition and especially photooxidation of stream-derived allochthonous compounds can hence play a major role in explaining the composition of lake DOM and the processing of inflowing stream DOM. Spencer et al. (2009) reported photooxidation to remove lignin phenols from a large river system and Helms et al. (2014) investigated the loss of DOM optical properties after light exposition. This process can hence explain the depleted phenolic constituents corresponding to decreased Ai_{mod} and DBE indices. It is likely, that the plant- and soil-derived DOM signatures accounted in brackish water samples are underwent a transformation and disintegration when residing longer in the lake ecosystem. Miranda et al. (2020) found that highly unsaturated and aromatic compounds are not only degraded, but partly transformed to unsaturated N-rich compounds by UV radiation. This can further explain the increase in unsaturated and unsaturated N-containing compound classes in our data. Along with degradation and transformation of allochthonous DOM, autochthonous DOM production can play a large role for the natural organic matter characteristics of Lake Nam Co. Microbial autochthonous DOM sources have been suggested for Lake Nam Co (Spencer et al., 2014) and other large lakes of the Himalayas (Liu et al., 2020). Hu et al. (2016) report from a foodweb study utilising a coupled $\delta^{13}C/\Delta^{14}C$ isotope approach for Lake Nam Co, that mainly autochthonous organic carbon sources are utilised by lake zooplankton, further corroborating the suggested autochthonous DOM source. The high number in heteroatoms in combination with unsaturated compound classes in lake DOM further support a microbial-derived DOM source in lake water samples (Feng et al., 2016). The signature of water samples of Lake Nam Co resembles DOM characteristics of the open ocean, by being low in condensed aromatic and aromatic compounds and implicating an autochthonous DOM source derived from algal and microbial production (Zark and Dittmar, 2018; Seidel et al., 2015). DOM of this large saline, oligotrophic and endorheic Tibetan lake is evidently uninfluenced from DOM of inflowing streams.

Conclusion

Our aim was to elucidate the composition and processing of DOM along the flow path in the High-Asian endorheic Nam Co catchment. We investigated different landscape units of three catchments, including water samples of glaciers, springs, alpine wetlands, streams, the brackish mixing zone and the saline lake. Here, we showed that the water source influences DOM signatures in streams of the Lake Nam Co catchment. Molecular diversity was large in glacial influenced streams, which were also characterized by the largest proportion of low molecular weight compounds. In the glacial influenced Qugaqie catchment, we identified a unique dual source, on the one hand, a microbial-borne, low-molecular weight DOM fraction relatively rich in P heteroatoms and unsaturated compounds with and without nitrogen, such as peptides and aminosugars, suggesting high biolability and autotrophic production in the glacial ice shield and on the other hand condensed hydrocarbons with high aromaticity and highly unsaturated compounds, such as plant-borne lignin, hinting at a depositional source of aeolian transported black carbon or local dust, derived from soil. The Niyaqu and Zhagu catchment comprised a lower molecular diversity, and in comparison show a mainly allochthonous DOM source of highly unsaturated and aromatic structures, attributed to the input of surrounding plants and soils to the streams. The large influence of glacial meltwater in the Qugaqie catchment greatly modifies DOM signatures along the whole stream of the catchment and delivers biolabile compounds to the southern lake shoreline, underlining the existence of a glacial – lacustrine pathway.

Spring DOM showed a low molecular diversity and was relatively enriched in plant and soil-derived aromatic and highly unsaturated compounds alongside with an increase in S heteroatoms and saturated formulae. This suggests that spring DOM of the Zhagu upland constitutes background DOM signatures in the stream. Wetland DOM on the other hand showed a high molecular diversity and was especially rich in unsaturated, saturated and N-rich compounds, alongside with aromatic and highly unsaturated formulae. Wetland DOM likely represents an almost complete range of terrestrial DOM signatures of the catchment, driven by the high productivity, water-logging and the basin topography of the wetlands. The latter lead to a steady inflow of water and natural organic matter from surrounding slopes. If the reported degradation of alpine wetlands in High Asia drives a larger release of DOM, this can be considered a threat for oligotrophic terminal aquatic systems, such as Lake Nam Co.

Stream samples on the other hand were mostly associated with the changing input of allochthonous materials, originating from vascular plants and soils. These are attributed to the predominant *K. pygmaea* biome and pastoral practise stretching along the path of streams.

Brackish samples represent the mixing zone of stream and lake water, showing that the terrestrial DOM signal is being transported with stream waters into the lake. Lake DOM was chemically diverse compared to all other sources. Lake DOM suggests intense photooxidative processing and transformation of imported allochthonous stream DOM, alongside with an autochthonous DOM source from microbial and algal in-situ production in the large oligotrophic lake. These DOM signatures can be compared to terminal signatures encountered in the open ocean. Our study shows that water quality in the Nam Co catchment needs a thorough assessment, since it can be diverse between catchments and landscape units. In order to safeguard water resources and related ecosystem services, knowledge about the different sources and their later processing is indispensable. DOM properties have proven here as a selective proxy, suitable to be implemented as a monitoring agent in the Nam Co catchment, representative for processes on the southern TP.

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Study III

Zhang Z, Qin J, Sun H, Yang J, Liu Y. Spatiotemporal Dynamics of Dissolved Organic Carbon and Freshwater Browning in the Zoige Alpine Wetland, Northeastern Qinghai-Tibetan Plateau. Water 2020;12(9).

Study III – Supplementary materials

Investigation of dissolved organic matter properties of the large oligotrophic Tibetan Lake Nam Co and three catchments by non- targeted organic ultrahigh resolution Fourier transform ion cyclotron resonance mass spectrometry (FT-ICR MS)

Technical Report – Supplementary materials

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Study III – Supplementary materials

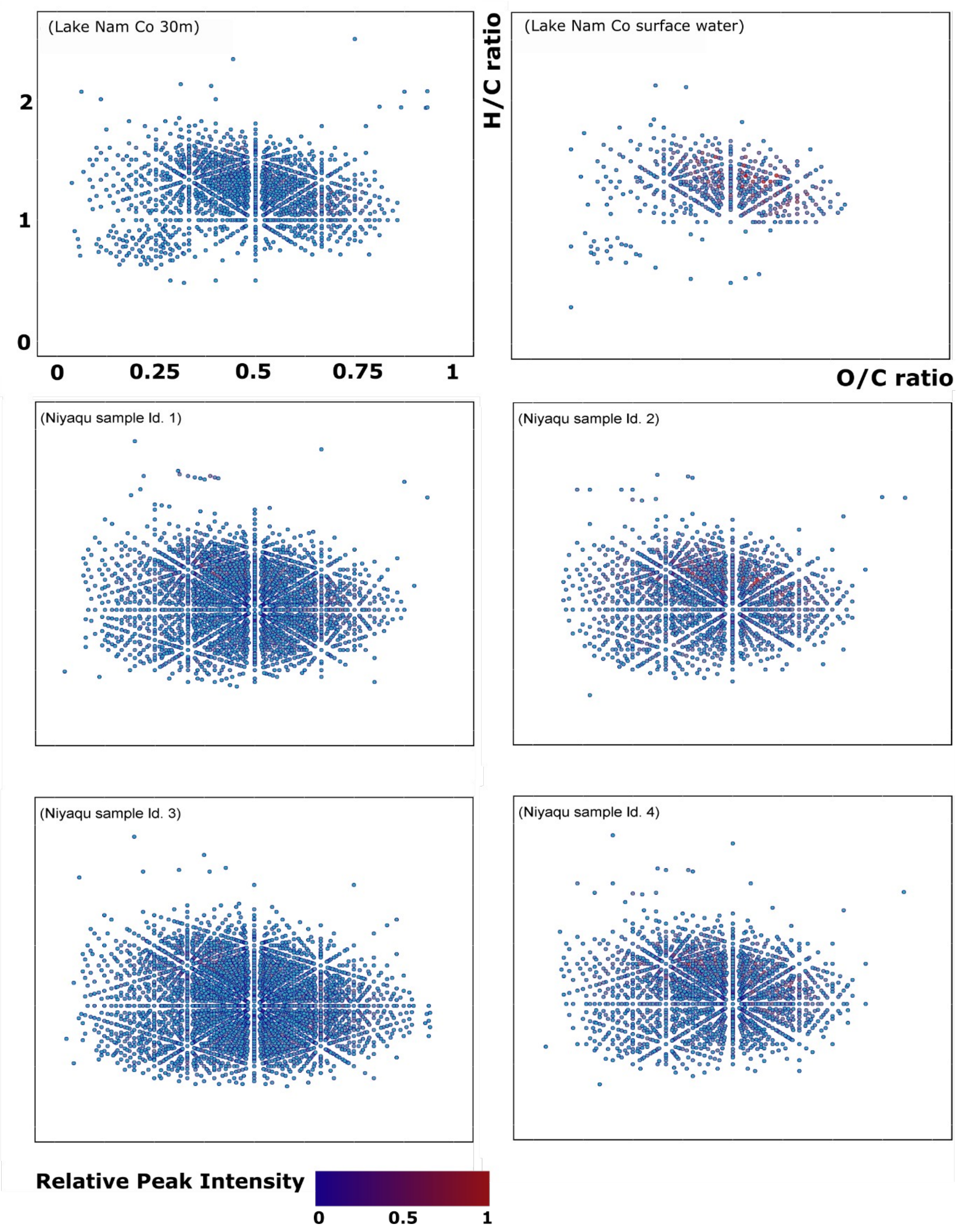
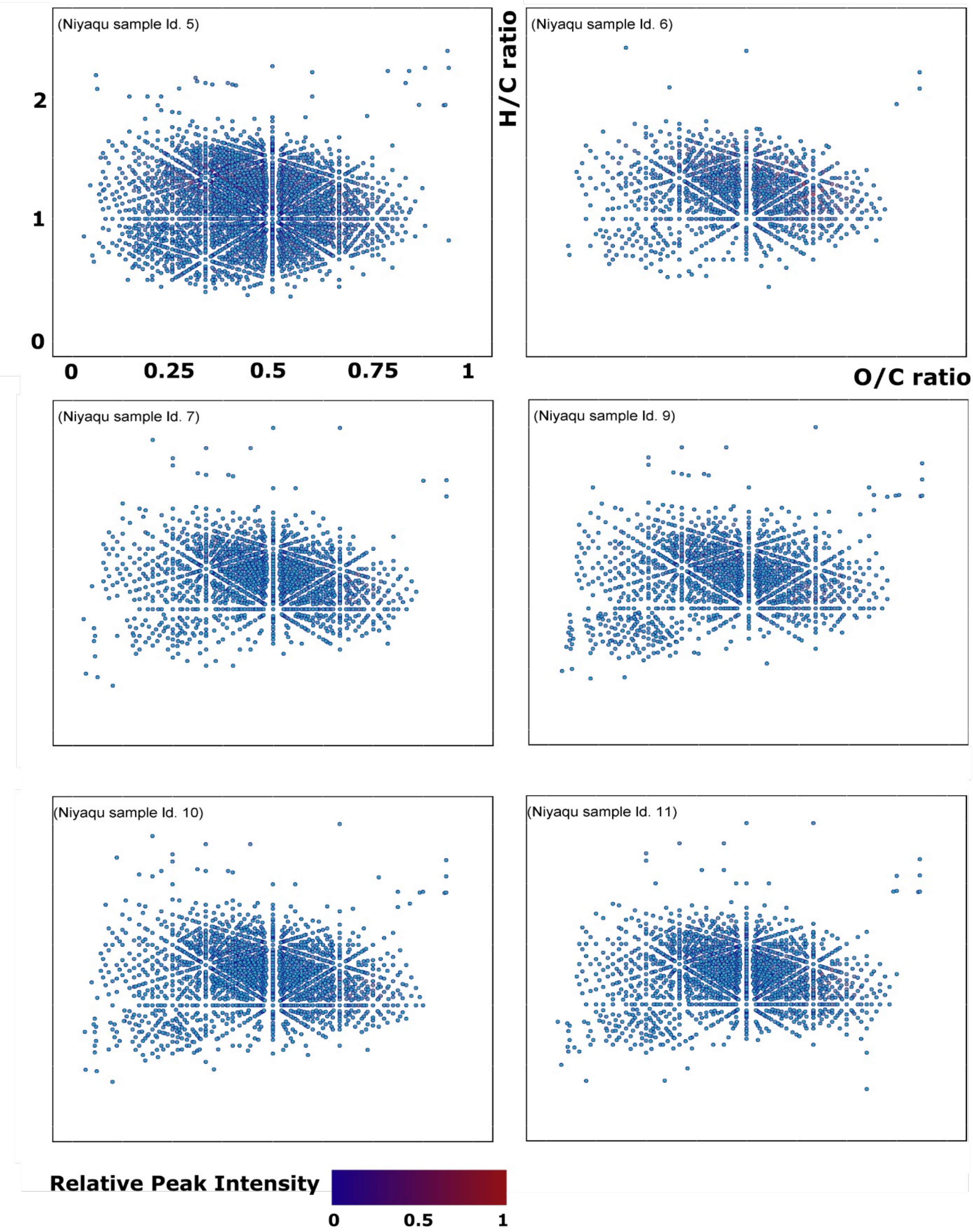


Figure S1: van Krevelen diagrams of FT-ICR MS sample dataset



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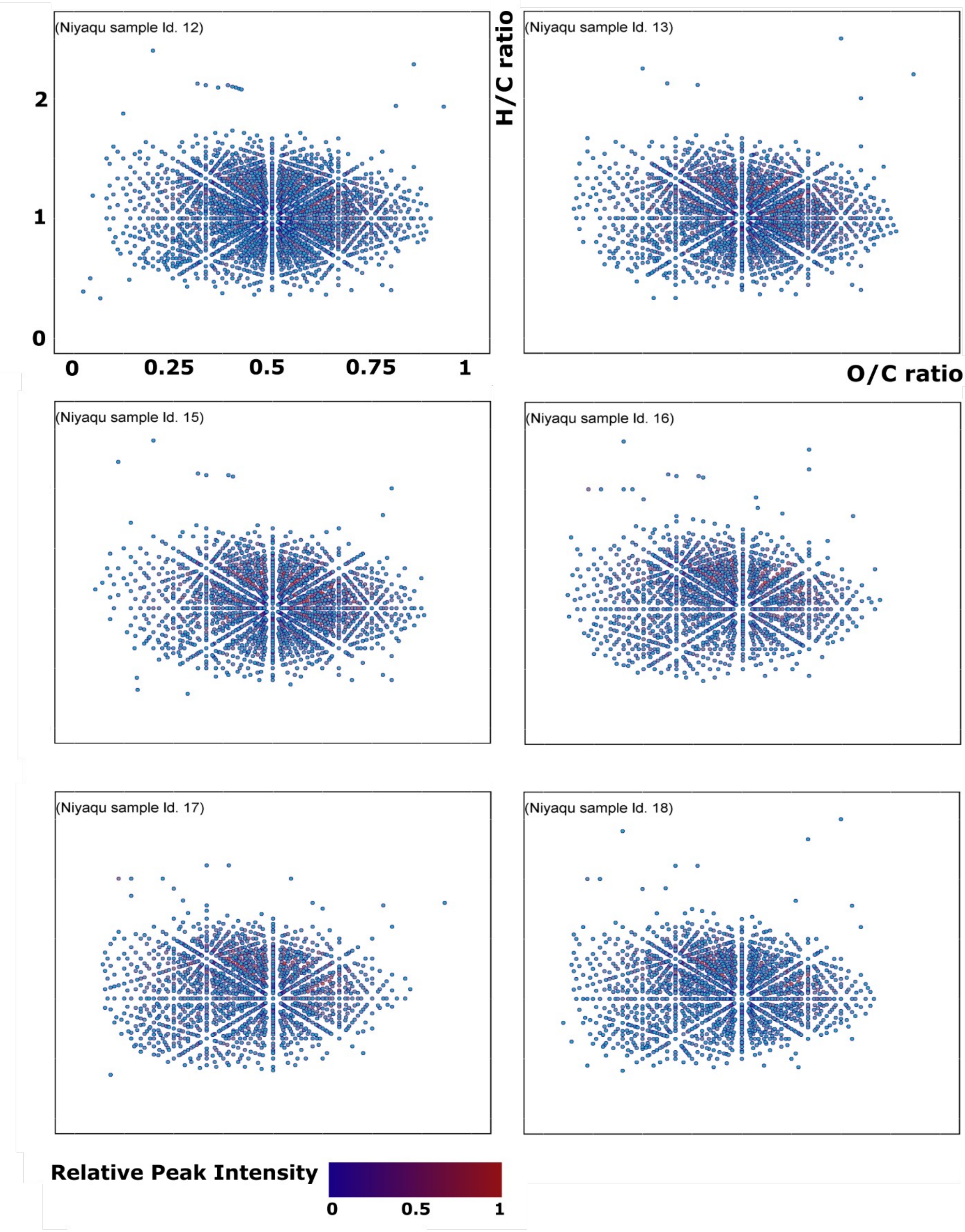

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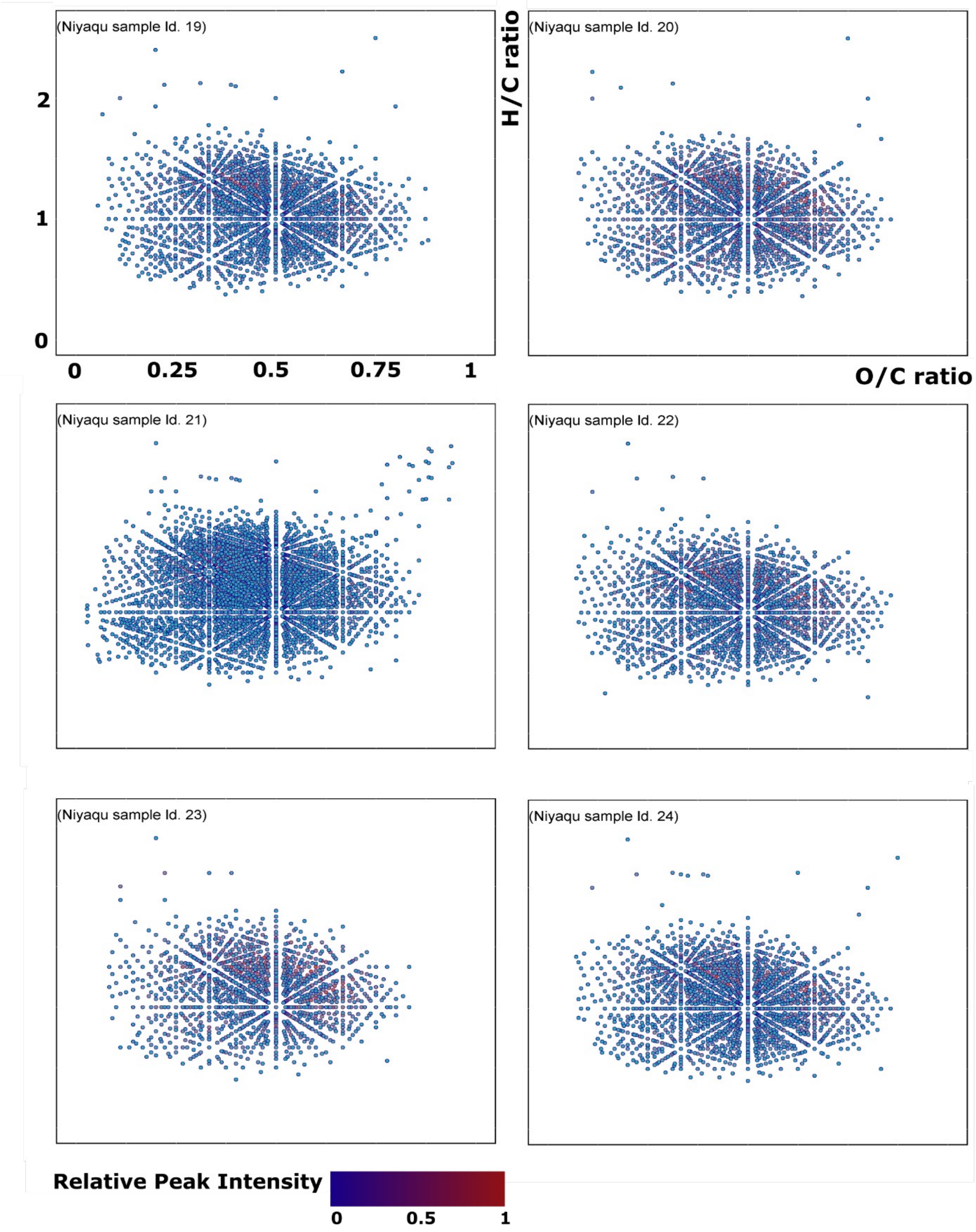

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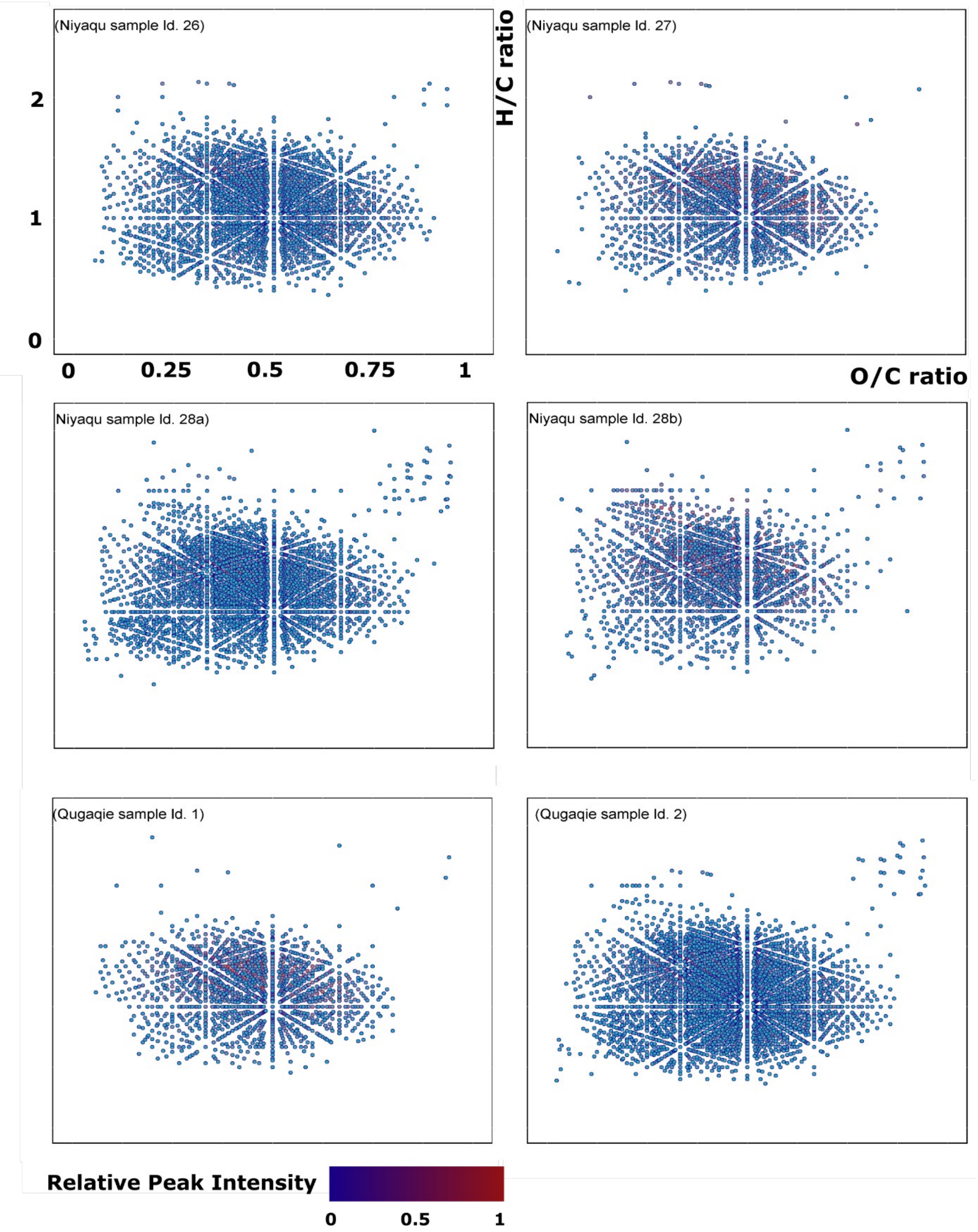

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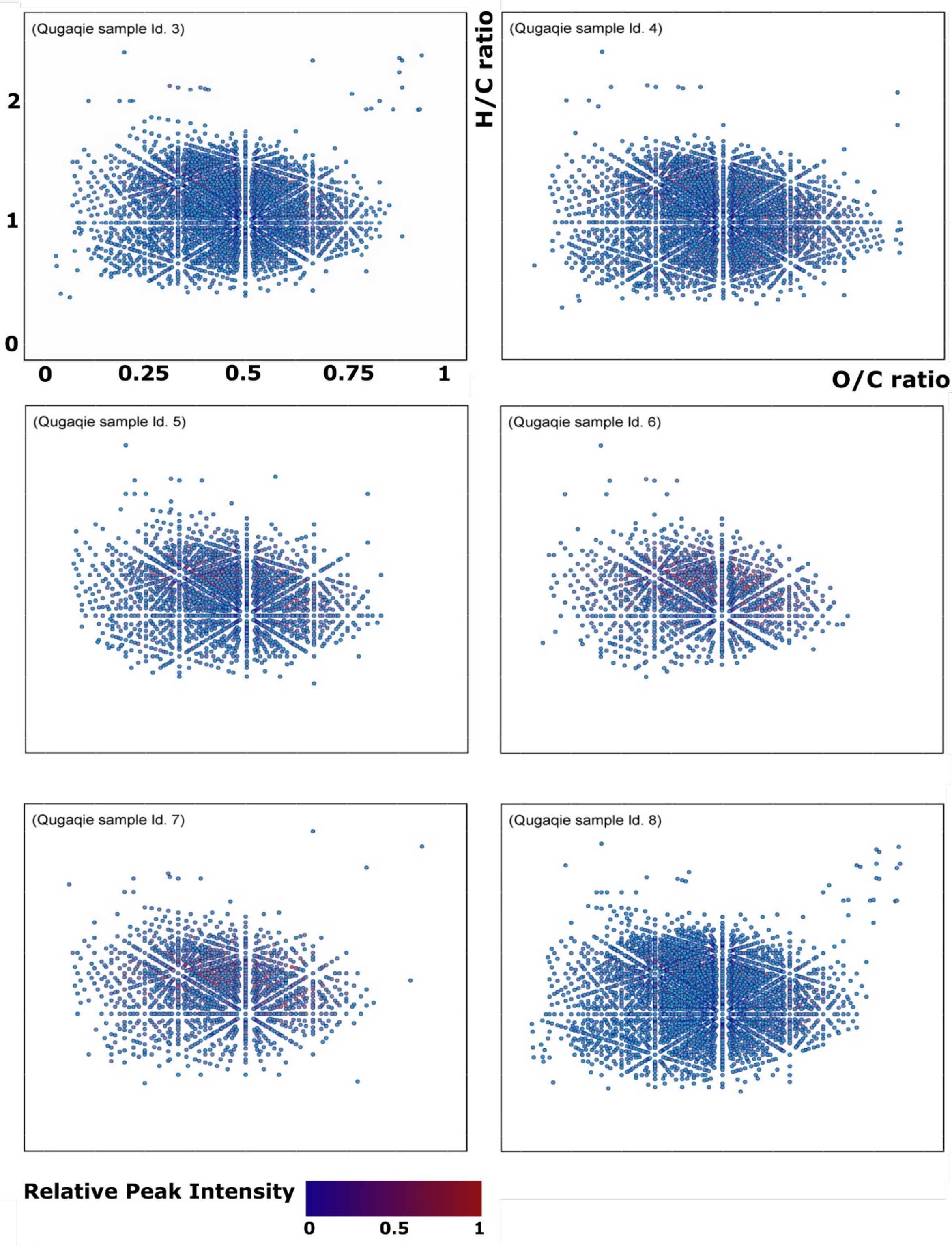

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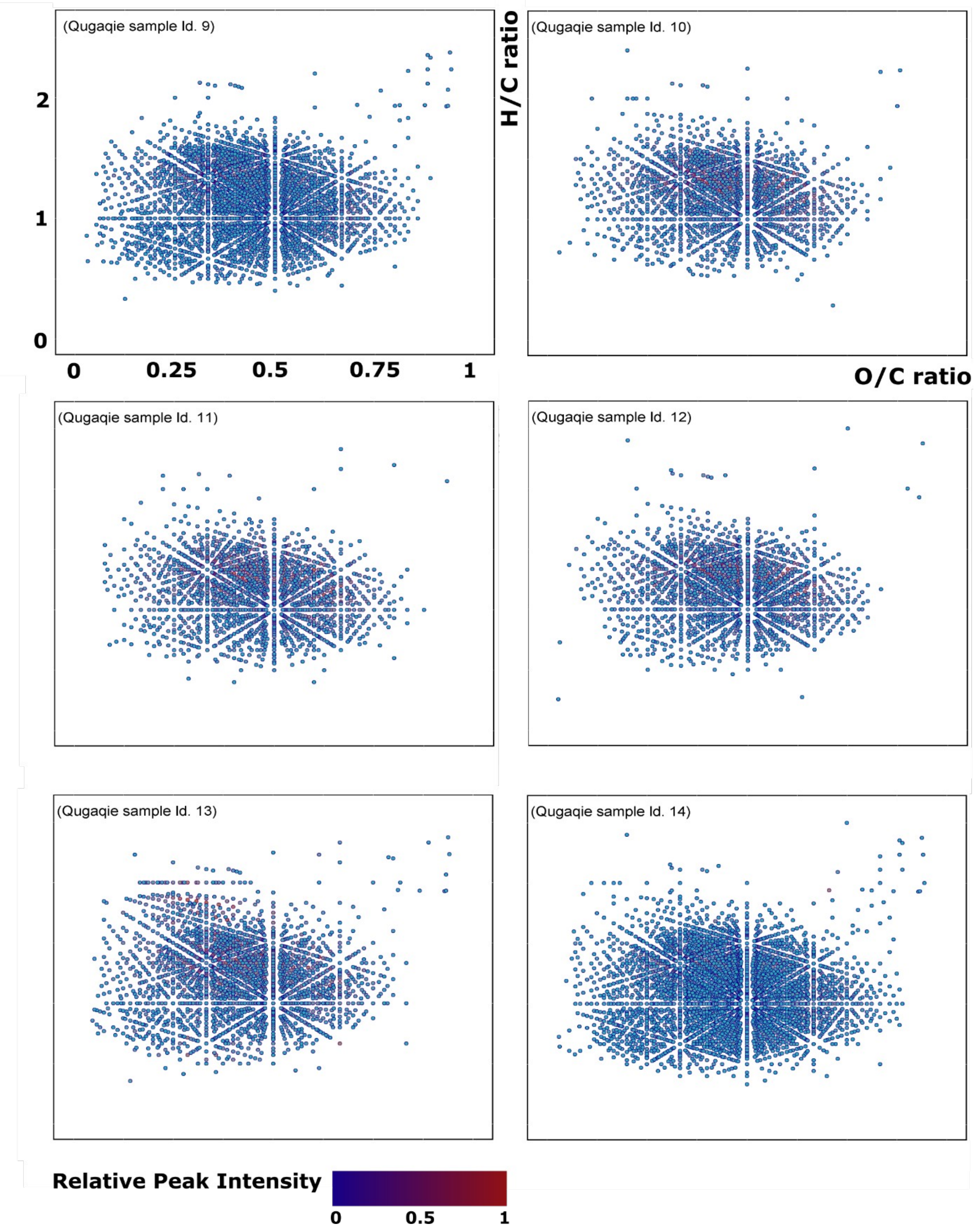

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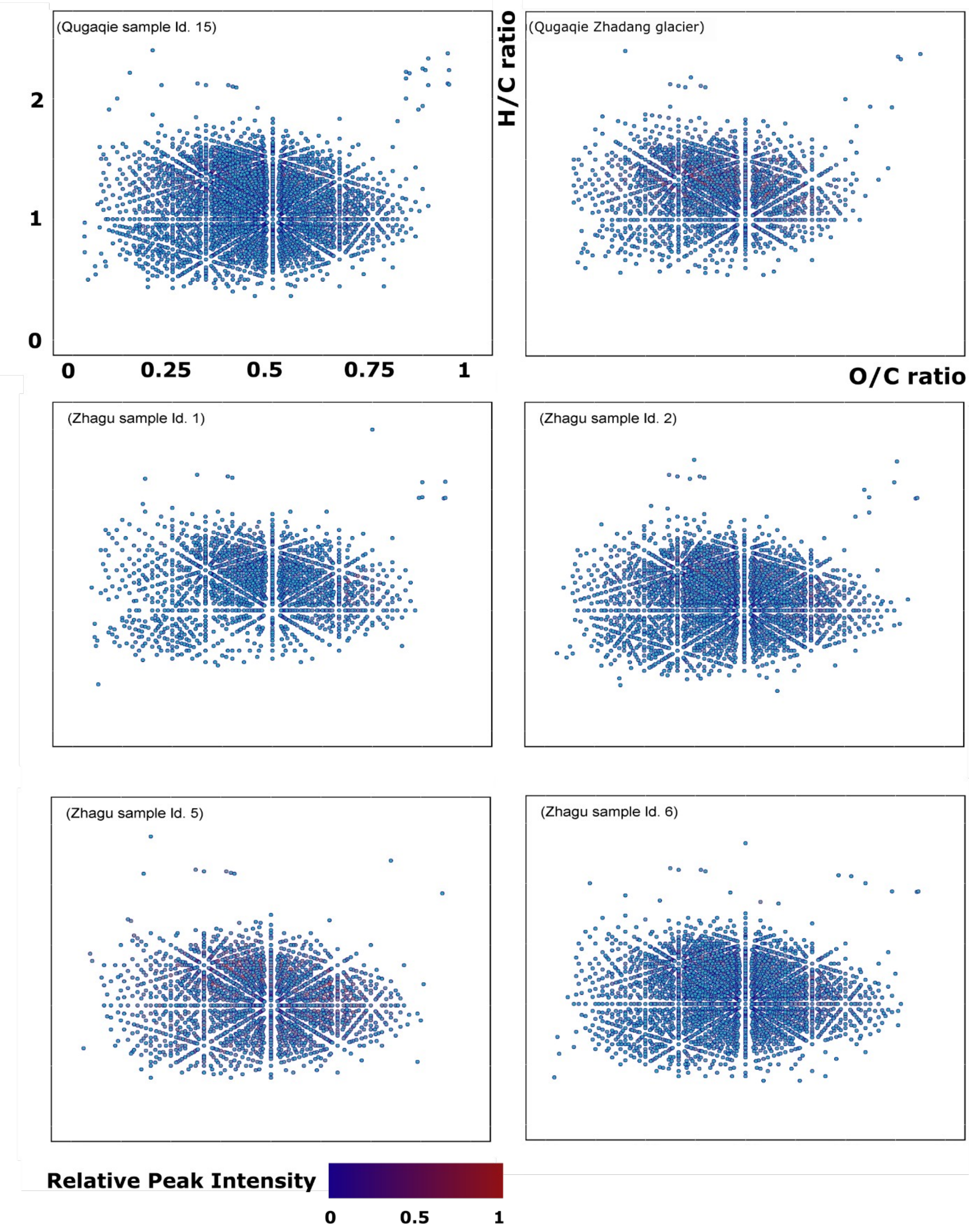

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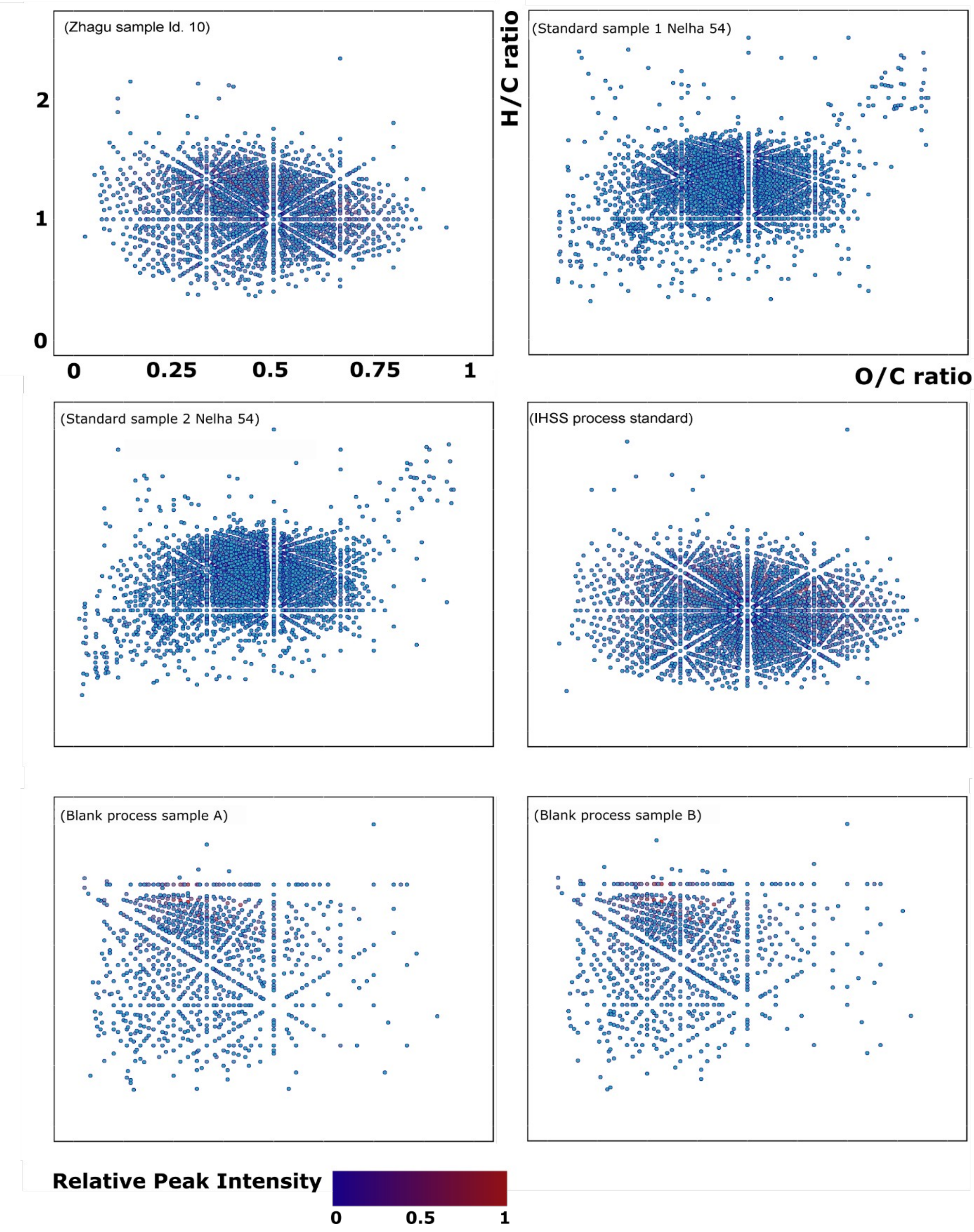

Figure S1 (continued): van Krevelen diagrams of FT-ICR MS sample dataset

Table S1: Multiple pairwise non-parametric Mann Whitney tests comparisons for the independent factor: Site. Tested was for differences in group ranks. “1” indicates that there are no rank differences between sites. Significance indication: * = α 0.05; ** = α 0.01; * = α 0.001**

Dependent variable:	**Mean amount of N heteroatoms**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	1		
Zhagu	1	1	

Dependent variable:	**Mean amount of S heteroatoms**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	1		
Zhagu	1	1	

Dependent variable:	**Mean amount of P heteroatoms**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	0.02*		
Zhagu	1	0.05*	

Dependent variable:	**Mean Ai_{mod}**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	1		
Zhagu	1	0.91	

Dependent variable:	**Mean count of formulas $Ai_{mod} \geq 0.5$**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	1		
Zhagu	1	1	

Dependent variable:	**Mean count of formulas $Ai_{mod} \geq 0.67$**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	1		
Zhagu	1	1	

Dependent variable:	**Mean DBE (double bond equivalents)**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	1		
Zhagu	1	1	

Dependent variable:	**Mean total count of formulas**		
Independent variable:	Niyaqu	Qugaqie	Zhagu

Site			
Niyaqu			
Qugaqie	1		
Zhagu	1	1	

Dependent variable:	**Mean intensity corrected formula abundance: Aromatic O-rich (AROR)**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	1		
Zhagu	1	1	

Dependent variable:	**Mean intensity corrected formula abundance: Aromatic O-poor (AROP)**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	1		
Zhagu	1	1	

Dependent variable:	**Mean intensity corrected formula abundance: Highly Unsaturated O-rich (HUSOR)**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	0.014*	1	
Zhagu	1	0.012*	

Dependent variable:	**Mean intensity corrected formula abundance: Highly Unsaturated O-poor (HUSOP)**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	0.0002***		
Zhagu	1	0.24	

Dependent variable:	**Mean intensity corrected formula abundance: Unsaturated O-rich (USOR)**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	0.052		
Zhagu	1	0.841	

Dependent variable:	**Mean intensity corrected formula abundance: Unsaturated O-poor (USOP)**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	0.850		
Zhagu	0.710	0.050*	

Dependent variable:	**Mean intensity corrected formula abundance: Unsaturated with N (USN)**

Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	1	1	
Zhagu	1	1	

Dependent variable:	**Mean intensity corrected formula abundance: Saturated (SAT) / O-rich and O-poor**		
Independent variable: Site	Niyaqu	Qugaqie	Zhagu
Niyaqu			
Qugaqie	1		
Zhagu	1	1	

Table S2: Multiple pairwise non-parametric Mann Whitney tests comparisons for the independent factor: Sample category. Tested was for differences in group ranks. “1” indicates that there are no rank differences between sites. Significance indication: * = α 0.05; ** = α 0.01; * = α 0.001.**

Dependent variable:	**Mean amount of N heteroatoms**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	1			
Lake water	1	1		
Stream water	0.62	1	1	

Dependent variable:	**Mean amount of S heteroatoms**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	1			
Lake water	1	1		
Stream water	1	1	1	

Dependent variable:	**Mean amount of P heteroatoms**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	1			
Lake water	1	1		
Stream water	1	0.69	1	

Dependent variable:	**Mean Ai$_{mod}$**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	0.655			
Lake water	0.343	1		
Stream water	1	1	1	1

Dependent variable:	**Mean count of formulas Ai$_{mod}$ ≥ 0.5**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	0.291			
Lake water	0.343	0.073		
Stream water	0.249	1	0.120	

Dependent variable:	**Mean count of formulas Ai$_{mod}$ ≥ 0.67**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	0.291			
Lake water	0.343	0.073		
Stream water	0.263	1	0.08	

Dependent variable:	**Mean DBE (double bond equivalents)**

Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	0.440			
Lake water	0.340	0.29		
Stream water	0.260	1	0.12	

Dependent variable:	**Mean total count of formulas**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	1			
Lake water	1	1		
Stream water	0.440	1	0.660	

Dependent variable:	**Mean intensity corrected formula abundance: Aromatic O-rich (AROR)**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	0.655			
Lake water	0.343	0.145		
Stream water	1	1	0.050*	

Dependent variable:	**Mean intensity corrected formula abundance: Aromatic O-poor (AROP)**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	0.170			
Lake water	0.343	0.073		
Stream water	0.852	0.611	0.075	

Dependent variable:	**Mean intensity corrected formula abundance: Highly Unsaturated O-rich (HUSOR)**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	1			
Lake water	0.34	0.29		
Stream water	1	0.36	0.27	

Dependent variable:	**Mean intensity corrected formula abundance: Highly Unsaturated O-poor (HUSOP)**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	1			
Lake water	1	0.51		
Stream water	1	0.61	0.94	

Dependent variable:	**Mean intensity corrected formula abundance: Unsaturated O-rich (USOR)**			
Independent variable:	Brackish water	Glacial effluent	Lake water	Stream water

Sample category				
Brackish water				
Glacial effluent	1			
Lake water	1	1		
Stream water	1	1	1	

Dependent variable:	**Mean intensity corrected formula abundance: Unsaturated O-poor (USOP)**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	1			
Lake water	0.34	1		
Stream water	1	1	0.37	

Dependent variable:	**Mean intensity corrected formula abundance: Unsaturated with N (USN)**			
Independent variable: Sample category	Brackish water	Glacial effluent	Lake water	Stream water
Brackish water				
Glacial effluent	1			
Lake water	1	1		
Stream water	0.34	1	1	

Table S3: NMDS ordination coordinates (loadings) of all samples from the study area for dimension 1 and 2; SCN = Niyaqu catchment, SCQ = Qugaqie catchment, SCZ =Zhagu catchment, L = Lake Nam Co.

Sample	NMDS 1 ordination coordinate	NMDS 2 ordination coordinate
SCN_1_1_mean	-0.34907631	-0.322503222
SCN_1_2_mean	-0.15261150	0.099628628
SCN_1_3_mean	-0.61332353	0.241336649
SCN_1_4_mean	-0.12900445	0.103503558
SCN_1_5_mean	-0.19720730	0.338868782
SCN_1_6_mean	0.14828826	-0.192145050
SCN_1_7_mean	0.46194704	-0.470204200
SCN_1_9_mean	0.66461866	-0.415008709
SCN_1_10_mean	0.59454904	-0.243046184
SCN_1_11_mean	0.53312668	-0.291190702
SCN_1_12_mean	-0.58174829	-0.098577977
SCN_1_13_mean	-0.58962850	-0.109459337
SCN_1_15_mean	-0.58043746	-0.101479527
SCN_1_16_mean	-0.22803615	-0.161935688
SCN_1_17_mean	-0.30989636	-0.022775226
SCN_1_18_mean	-0.21245367	-0.004803632
SCN_1_19_mean	-0.42842828	-0.070816490
SCN_1_20_mean	-0.52637291	-0.052112166
SCN_1_21_mean	0.54953693	0.537692089
SCN_1_22_mean	-0.47713164	-0.049933471
SCN_1_23_mean	-0.01040251	-0.354906688
SCN_1_24_mean	-0.34399457	-0.304586264
SCN_1_26_mean	-0.07889749	-0.026507438
SCN_1_27_mean	-0.25944421	-0.516174149
SCN_1_28a_mean	0.64422840	0.426812624
SCN_1_28b_mean	0.56541338	0.211680717
SCQ_2_1_mean	0.01854980	0.021197142
SCQ_2_2_mean	-0.14285698	0.559252194
SCQ_2_3_mean	-0.14184548	0.378724585
SCQ_2_4_mean	-0.55007567	0.140097562
SCQ_2_5_mean	0.08684673	0.030869175
SCQ_2_6_mean	0.17507420	-0.308177912
SCQ_2_7_mean	0.24990716	-0.168276701
SCQ_2_8_mean	0.09208665	0.482636135
SCQ_2_9_mean	0.08801431	0.440498255
SCQ_2_10_mean	0.10138810	-0.008289153
SCQ_2_11_mean	0.04760956	0.011825961
SCQ_2_12_mean	0.16872823	-0.350116566
SCQ_2_13_mean	0.41184496	-0.323617484
SCQ_2_14_mean	-0.23739353	0.469415344
SCQ_2_15_mean	-0.13342212	0.451970605
SCQ_Zhadang_Ice_mean	0.38391017	-0.110377558
SCZ_3_1_mean	0.19104626	-0.112995525
SCZ_3_2_mean	-0.20435145	0.240019469
SCZ_3_5_mean	-0.45969134	-0.364684021
SCZ_3_6_mean	-0.26903835	0.216893321

SCZ_3_10_mean	-0.16466657	0.007720929
L_Lake Surface_mean	0.12060423	-0.451053872
L_Lake 30 m_mean	0.18426949	-0.102036704

Table S4: NMDS ordination variable scores for all tested dependent variables of the FT-ICR MS dataset.

ID	Variable	NMDS Dimension 1	NMDS Dimension 2	r^2	Pr (test value)
1	N (number of heteroatoms)	0.02431	-0.99970	0.5355	0.001
2	S (number of heteroatoms)	0.28312	-0.95909	0.1810	0.010
3	P (number of heteroatoms)	0.14351	-0.98965	0.6802	0.001
4	Aimod	-0.98885	-0.14893	0.8831	0.001
5	DBE	-0.90154	-0.43271	0.8182	0.001
6	Aror	-0.99825	0.05911	0.7598	0.001
7	Arop	-0.91174	-0.41076	0.7885	0.001
8	Husor	-0.05864	0.99828	0.4134	0.001
9	Husop	0.14713	-0.98912	0.2646	0.001
10	Usor	0.87542	0.48336	0.3498	0.001
11	Usop	0.98024	-0.19781	0.8387	0.001
12	Usn	0.96050	-0.27828	0.7901	0.001
13	Sor	0.65729	-0.75364	0.5765	0.001
14	Sop	0.49928	0.86644	0.3564	0.001

Table S5: Extraction efficiency of SPE-DOM samples; SCN =Niyaqu, SCQ= Qugaqie, SCZ = Zhagu, SCL= Lake Nam Co.

ID	Sample Name	SPE-DOC extraction efficiency [%]
1	SCN_1_1	61.1
2	SCN _1_2	77.1
3	SCN_1_3	50.2
4	SCN_1_4	45.2
5	SCN_1_5	45.9
6	SCN_1_6	98.2
7	SCN_1_7	57.7
8	SCN_1_9	67.0
9	SCN_1_10	74.9
10	SCN_1_11	56.2
11	SCN_1_12	60.9
12	SCN_1_1	27.3
13	SCN_1_15	16.8
14	SCN_1_16	78.9
15	SCN_1_17	83.2
16	SCN_1_18	63.8
17	SCN_1_19	83.3
18	SCN_1_20	92.7
19	SCN_1_21	91.5
20	SCN_1_22	68.4
21	SCN_1_23	84.7
22	SCN_1_24	55.5
23	SCN_1_2	79.6
24	SCN_1_27	84.7
25	SCN_1_28a	>50%; initial DOC < 25 µM L^{-1}
26	SCN_1_28b	>50%; initial DOC < 25 µM L^{-1}
27	SCQ_2_1	46.8
28	SCQ_2_2	43.7
29	SCQ_2_3	52.8
30	SCQ_2_4	50.6
31	SCQ_2_5	45.5
32	SCQ_2_6	55.9
33	SCQ_2_7	37.7
34	SCQ_2_8	43.8
35	SCQ_2_9	52.5
36	SCQ_2_10	37.8
37	SCQ_2_11	54.7
38	SCQ_2_12	43.7
39	SCQ_2_13	84.1
40	SCQ_2_14	44.1
41	SCQ_2_15	67.5
42	SCQ_Zhadang_Ice	>50%; initial DOC < 25 µM L^{-1}
43	SCZ_3_1	34.1
44	SCZ_3_2	73.0
45	SCZ_3_5	50.1
46	SCZ_3_6	15.0
47	SCZ_3_10	91.3

48	SCL_Surface	77.4
49	SCL_30m	41.0
50	IHSS_Std	60.6
51	Blank_a	initial DOC < 25 µM L^{-1}
52	Blank_b	initial DOC < 25 µM L^{-1}

Synthesis

Aim of the thesis

The aim of the thesis was to provide information on characteristics, dynamics, processing and transformation of dissolved organic matter (DOM) for the endorheic Nam Co basin on the southern TP, to investigate the influence of site-specific catchment characteristics and seasonality on DOM and the fate of DOM leached from the terrestrial domain into the fluvial pathway. The first part of the synthesis, based on Study I, revises the current state of knowledge and defines persisting deficits. Study II addressed the characteristics, dynamics and processing of DOM sampled from streams, endmembers: glacial ecosystems, springs and an alpine wetland, as well as the Nam Co Lake. Three sampling campaigns were conducted to cover the seasonality aspect of DOM composition during freshet, the phase of the Indian summer monsoon (ISM) and the late year baseflow. Further, a watershed-wide plant cover estimate on 30m resolution, spanning 30 years was calibrated to investigate current trends in plant cover change and to form a basis to connect green land cover with DOM signatures in streams. Study III presents an ultra-high resolution mass spectrometry dataset (FT-ICR MS) for 48 DOM samples of streams, the named endmembers, the brackish mixing zone and the terminal lake. These samples were taken during baseflow conditions and allow an in-depth investigation of molecular composition of DOM, useful to be aligned with the proxy results produced in Study II.

The evolving topic of DOM characteristics in the Nam Co watershed

As outlined, DOM is an important component of natural element and nutrient cycles. Roulet and Moore (2006), describe DOM as the ‘agent browning the waters’, this referring to the high amount of phenols, such as aromatic compounds in DOM, originating mostly from terrestrial sources like soils and vascular plants (Kalbitz et al., 2006), which can make up a large share of exported compounds from terrestrial to aquatic ecosystems (Spencer et al., 2009) and can adversely affect lake ecosystems by oxygen depletion (Knoll et al., 2018). DOM can consist of a wide range of components (Kalbitz et al., 2000), being as diverse as the ecosystems it originates from. Through this, investigations of DOM allow a unique fingerprinting of ecosystem processes and connections (Jaffé et al., 2012). Since the process of DOM leaching and translocation, is largely dependent on water percolation and processes in the soil column (Fisher et al., 2018; Kaiser and Kalbitz, 2012), it can follow the seasonal and interannual variation in an ecosystem and also its site-specific influences (addressed as biogeodiversity in Study I). As is implicit from reviews (Lützow et al., 2006), studies on DOM tended to focus mainly on temperate regions with an ongoing expansion into more remote areas of the world, such as the Amazonas basin, and Artic ecosystems (Aufdenkampe et al., 2001; Prokushkin et al., 2011.; Kida et al., 2021; Lafrenière and Sharp, 2004). This also reflects the TP. A study on Chinese publication metrics shows that Tibet is the least productive region when counting total research output and research disciplines (Hu et al., 2017). Consequentially, an outcome of Study I was that only little is known about DOM origin, its connected characteristics, fluxes, degradation, processes and fate. Qu et al. (2017) investigated three large rivers on the Tibetan plateau: Yellow (Huang He), Yangtze and Yarlung Tsangpo identifying increases of DOC discharge during the ISM season, but overall small fluxes and Δ^{14}C aged DOC compared to large river systems in the tropics and arctic. Besides this examination of large rivers, up to now, three case studies from the Nam Co watershed are available. Kai et al. (2019) present a study in Chinese language, from the terminus of 21 rivers in the Nam Co watershed and two sampling points in the lake. They found a seasonality driven increase of DOC during the monsoon period, but inter-annual variability of the two studied years (2012 and 2013) was larger than the effect of seasonality. Another case study is available from Spencer et al. (2014), this study employs a multi-methodological dataset and is based on six samples, making statements about two lakes, including Nam Co, two glaciers and their proglacial streams. Here, small fluxes (0.2-0.3 mg C L^{-1}) of highly bioavailiable DOM from a southward oriented glacier of the Nyainqentanglha and its proglacial stream were reported. Spencer et al. (2014) further suggest that a fraction of DOM is aeolian deposition and pre-aged Δ^{14}C ages (~750-2350 ybp) and probably derives from fossil fuel residues. A higher DOC content was observed in Lake Nam Co (3.1 mg C L^{-1}) together with ancient Δ^{14}C ages (525 ybp) and a ^{13}C enriched signature of DOM (-22.9 ‰) suggesting autochthonous microbial production which findings from Study II (Figure 4e, Figure 5), are in line with this. The van Krevelen diagrams of DOM from Lake Nam Co and glacial DOM are fit to results of Study III (Supplementary materials, Figure S1 Lake Nam Co 30m, Lake Nam Co surface, Qugaqie Zhadang glacier) pointing at stability of DOM composition over time. The results of Spencer et al. (2014), suggest that lake DOM is unaffected from the impact of the catchments influences, these findings are corroborated by the multi-parameter approach of Study II and the ultra-high resolution dataset of Study III.

Further, a recent study by Li et al. (2021) in a subbasin of Nam Co reports water chemistry parameters, UV/VIS absorbance and DOC corrected specific ultraviolet absorbance (SUVA), as well as PARAFAC resolved fluorophore DOM (FDOM) composition for six sampling points along the flow path of the southern Niyaqu stream. The authors found aromatic DOM to be discharged from the glacial environment, which is in-line with results of Study II and Study III. The DOM of alpine wetlands was further described to have high humic –like FDOM peaks and to act as a water filter. Study II also found high contributions of humic-like FDOM, but also an increased total load of anions and cations and of mineralized nitrogen (Figure 8b).The latter likely results from the basin topography of the wetland together with slowly flowing water, probably corresponding to higher temperatures in wetlands and accelerated microbial productivity. However, the stream does not directly flow through the wetland (Study II Figure 2c), but is rather connected to the stream via groundwater flow. Hence, Study II alongside with other studies (Huo et al., 2013; Zhang et al., 2020) concludes, that wetlands, if they degrade, can pose a threat to freshwater resources by browning and eutrophication through potential release of particularly plant-derived, chromophoric DOM.

Li et al. (2021) suggest processing of DOM during the flowpath, especially by a loss of the humic glacial signal. Study II suggests similar results (Figure 8e) especially when comparing terminal lake signatures to those of stream water. But study II further found continuous inputs of plant-borne DOM from *K. pygmaea* pastures, masking the transformation of DOM during the stream path. This corroborates findings from Roebuck et al. (2020), where continuous input from agricultural practise overlayed DOM processing. Further, Study III (Table 2, Figure 3) shows that the brackish intermixing zone of streams at the estuary receives the terrestrial DOM signal transported with the streams, while this signal is absent in samples taken in the pelagial of the lake. This shows that only limited processing is probable during the rapid flow path, and that processing of DOM mostly takes place in the lake water column. Given the literature reviewed in Study I, it was concluded, that the characteristics and dynamics of DOM need further clarification by in-depth analysis of bio-and geodiversity drivers, especially through investigations that couple effects of seasonality with the influence of site-specific DOM signatures, this was then carried out in Study II and III. From a limnology perspective, it is of utmost importance to understand the processes that shape DOM composition in the watershed source area and to link it to the transformation of DOM in endorheic Lake Nam Co (Minor and Oyler, 2021; Tank et al., 2010).

Site-specific DOM characteristics

As stated, small watersheds with low Strahler numbers are known to inherit site-specific attributes of biogeochemistry in their DOM fraction, or in other words, they show a high biogeodiversity. This has been shown for several case studies around the globe (Coch et al., 2019; Jaffé et al., 2012; Kawahigashi et al., 2004; Prokushkin et al., 2011; Shogren et al., 2019) and has found its expression in the implementation of the mechanistic river continuum concept (Tank et al., 2010) to approach stream water DOM characteristics as influenced by terrestrial ecosystems (Vannote et al., 1980; Mosher et al., 2015; Roebuck et al., 2020). Consequentially Study II tested the applicability of the river continuum concept. The samples utilised for Study II and III are generated from the same sampling scheme. Samples were taken from three catchments within the Nam Co watershed, differing with respect, to altitudinal gradient, aspect and exposition. By this they represent the transition of the steep, moist southern branch in the watershed, compared to the arid and hilly, less dissected northward zone in the Nam Co basin. Furthermore, the transition in biome types from *K. pygmaea* dominated alpine pasture towards alpine steppe, comprising *Stipa purpurea* and further species of *Artemisia, Stipa, Poa, Festuca* and *Carex* (Miehe et al., 2011; Nieberding et al., 2021) was covered. The sampling for Study II consisted of three sampling campaigns, resulting in a total of 138 samples, of which 76 were taken in the largest catchment: Niyaqu, located west from Lake Nam Co, 31 samples were taken from the Qugaqie catchment situated in the southern margin of the catchment, in the Nyainqentanglha mountain ridge and 23 samples were taken from the smallest catchment, Zhagu, located in the north-eastern border of the Nam Co watershed (Study II, Figure 1). Further, eight samples were drawn from the pelagic zone of the lake (30 m depth and surface water). This setup allowed to test for site-specific influences on DOM characteristics over several gradients and transition zones. The application of a watershed-wide, satellite-based plant cover estimate presented in Study II further allowed an investigation of the degree of coverage. The data showed that catchments differed in the degree of plant cover, with Niyaqu comprising the highest plant cover (52 %) followed by Qugaqie (~50 %) and Zhagu (~45 %). Study II outlined, that this hints at degradation of *K. pygmaea* pastures in the northernmost Zhagu catchment. Study II further found, that alpine pasture stretches along the flow path of the stream making the input of plant- and soil-derived DOM from alpine pastures the most probable source along the stream, while input from alpine steppe is hampered (Study II, Figure 3a). The sampling setup and combined datasets of Study II and Study III further allow to test for site-specific DOM signatures.

The comparison of catchments in Study II and III yields consistent results for the three investigated catchments, and allows to compare results obtained with different techniques. When assessing water chemistry parameters from study II (Table 1), higher calcium concentrations are visible for the Niyaqu (9.4 mg L^{-1}) and for the Zhagu (8.1 mg L^{-1}) catchments compared to the Qugaqie catchment (3.9 mg L^{-1}), which is situated in the Nyainqentanglha. As reported, the three catchments are situated in different geologic zones (Keil et al., 2010). A previous study of weathering products in the Niyaqu and Qugaqie catchment (Yu et al., 2019) found calcium to be a discriminating parameter between weathering processes in the two catchments. Our data also suggest, that solute composition in stream water reflects the geologic background of the catchments by separating the Nyainqentanglha zone, characterised by igneous rock (Qugaqie catchment) from the hilly uplands (Niyaqu and Zhagu catchment) build up mostly from carbonaceous sedimentary rock.

Mean DOC concentrations in the three catchments were overall low (Study II: Niyaqu 2.2 mg C L^{-1}, Qugaqie 2.3 mg C L^{-1} and Zhagu 2.9 mg C L^{-1}) serving as an indication, that bulk DOC assays are not sufficiently informative to explain compositional differences. Concerning the composition of DOM, it is hypothesized in **H1** that differences exist between the three investigated catchments. To estimate the input, e.g. of soil- and plant-borne compounds on stream DOM, study II and III utilise different methodological approaches. Specific ultraviolet absorbance at 254 nm (Study II; $SUVA_{254}$ representing chromophoric DOM, CDOM) is employed as a proxy for phenolic compounds attenuating ultraviolet light and likely derived from plants (Kowalczuk et al., 2010; Li et al., 2021; Weishaar et al., 2003; Zhao et al., 2016). Mean $SUVA_{254}$ in the Niyaqu catchment was 4.5 L mg C^{-1} m^{-1}, 3.3 L mg C^{-1} m^{-1} in Qugaqie and 2.9 L mg C^{-1} m^{-1} in Zhagu, indicating higher relative input of plant derived compounds in Niyaqu and Qugaqie. Aromaticity indicators from Study III, such as the mean Ai_{mod} (Koch and Dittmar, 2006, 2016) calculated for all assigned molecular formulae, do not correspond to the described $SUVA_{254}$ values in Study II. Here, AI_{mod} for the three catchments were comparable with: Niyaqu 0.33, Qugaqie 0.32 and Zhagu 0.34 (Study III, Table 2) for samples taken during the baseflow season (September 2019), while $SUVA_{254}$ of stream water was: 4.7 L mg C^{-1} m^{-1} for Niyaqu, 5.0 L mg C^{-1} m^{-1} for Qugaqie and 2.6 L mg C^{-1} m^{-1} in Zhagu. This indicates the selectivity and limitation of UV/VIS spectroscopic methods (Mäntele and Deniz, 2017), making multi-parameter approaches and techniques with higher resolution necessary. Another proxy, the $\delta\ ^{13}C$ ratio of DOM *(*Study II), shows that the mean for catchments differ considerably between Niyaqu (-24.6 ‰) and Zhagu (-24.14 ‰) compared to Qugaqie (-20.0 ‰) (Study II Figure 5). Hence, $\delta^{13}C$ of DOM is a sensitive parameter to track the effect of input sources on DOM, with a more negative isotopic composition responding to the inputs from plants and soils (Study II, Figure 5). When comparing the baseflow phase, sampled in September 2019 for studies II and III. Mean $\delta\ ^{13}C$ of DOM was -25.6 ‰ in Niyaqu, -23.4 ‰ in Qugaqie and -26.2 ‰ in Zhagu (Study II Figure 5). These means are in line to the reported Ai_{mod} of catchments (Study III, table 2) and further fit with the ratio of molecular compounds with $AI_{mod} > 0.5$ divided by the total number of assigned formulae, which is 0.21 for Niyaqu, 0.20 for Qugaqie and 0.22 for Zhagu. While there was no statistical significant difference between relative contribution of aromatic and aromatic condensed compound classes (Study III, Figure 2), coupling the Ai_{mod} proxy with $\delta\ ^{13}C$ of DOM further elucidates differences in the composition of DOM. Coupling of $SUVA_{254}$ and $\delta\ ^{13}C$ of DOM (both Study II) on the other hand, bears the risk of drawing false conclusions. The correlation of $SUVA_{254}$ and $\delta\ ^{13}C$ of DOM is not linear (Figure 6; $R^2 = 0.5$), showing a large scattering of the $SUVA_{254}$ index for highly depleted signatures of $\delta\ ^{13}C$ in DOM. The comparison with the Ai_{mod} indicates that this effect is not linearly driven by aromaticity. It remains an open question, what drives the high $SUVA_{254}$ absorption.

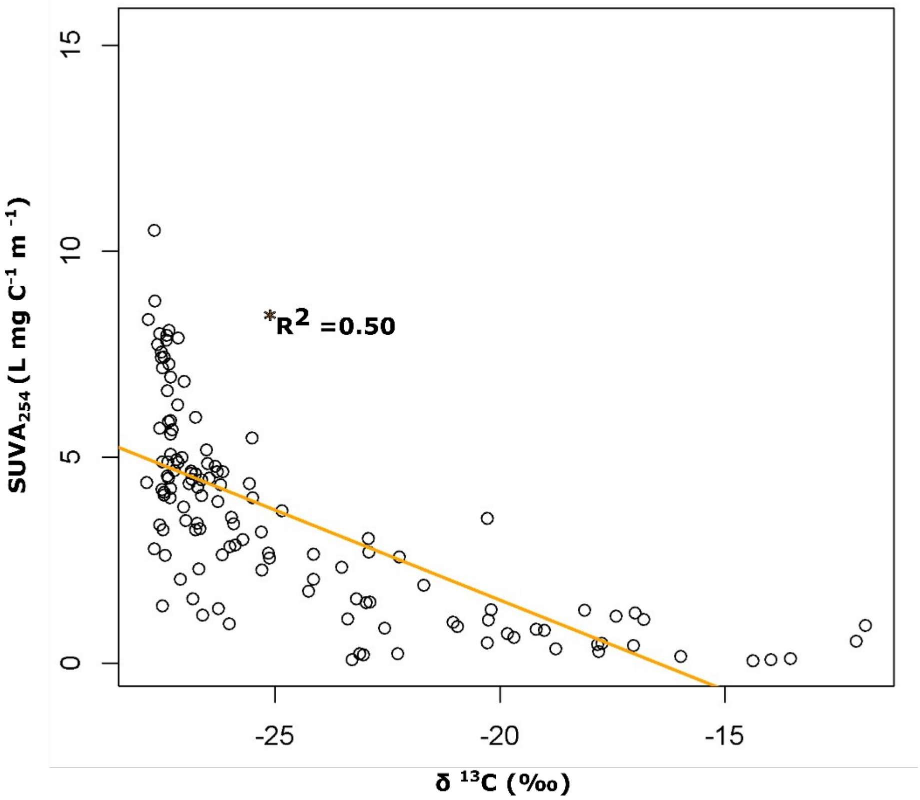

Figure 1: Scatterplot of the SUVA254 CDOM index and δ 1³C of DOM for all samples of Study II (n=138). Goodness of fit of the linear model is indicated by the linear regression line and expressed by explained variance (R^2) being 0.5, corresponding to 50% of total variance.

Data of Study III show that during baseflow conditions, significantly more highly unsaturated O-rich formulae were found in DOM from Qugaqie compared to Niyaqu and Zhagu (Study III, Figure 2). This indicates that relatively more higher molecular-weight compounds such as lignin and tannin- derived substances (Mann et al., 2015; Merder et al., 2020; Minor et al., 2014) were found in the DOM of Qugaqie compared to the other two catchments (Study III, Figure 2), while the DOM of Zhagu and Niyaqu was significantly richer in lower-molecular weight compounds (highly unsaturated O-poor). Here, DOM was derived from lower-molecular compounds of plants, such as alkene, or photooxidised lignin (Miranda et al., 2020; Mann et al., 2015; Merder et al., 2020; Minor et al., 2014) (Study III, Figure 2), which can be connected to the less turbulent flow of stream water in Niyaqu and Zhagu, leading to lower opacity of the water and making photooxidation possible.

Alongside with this, PARAFAC resolved fluorescence DOM (FDOM) was employed to separate DOM characteristics by assigning phenol-rich (humic-like) or microbial-like fluorophores (Baker and Inverarity, 2004; Chen et al., 2019; Hernes et al., 2009; Stubbins et al., 2014). In Study II, two FDOM models were produced, splitting the dataset in two groups, one with relatively high fluorophore response and a second group with low response. The high response group shows no significant differences for catchments in FDOM sums (Figure 7a), but the NMDS ordination of Study II (Figure 8a) shows that water samples of the Zhagu catchment had highest abundances for fluorophore component 3, defined as a microbial-like component. The water samples of Zhagu grouped in the low response group likewise cluster with two microbial-like FDOM components (FC1, FC2; Study II, Figure 8d). Additionally, Zhagu shows higher relative contributions of saturated compounds in its DOM, compared to Qugaqie (Study III, Figure 2). This suggests that DOM is richer in molecular structures resembling amino sugars and carbohydrates (Mann et al., 2015), together with nitrite and significantly highest nitrate concentrations (mean: 3.75 mg L^{-1}) (Study II, Table 1) at Zhagu. For DOM samples from Zhagu, depleted $\delta\ ^{13}C$ of DOM (-24.14 ‰) is connected to high Ai_{mod} indices (0.34). This is associated to microbial-like FDOM. This combination was interpreted as the influence of pasture degradation, in combination with the high grazing pressure, visible by trampling (Study II, supplementary materials, Figure S7, Figure S8). Microbial FDOM and N_{min} can be deduced to a contribution of yak faeces in the few remaining productive pastures. This also connects to the formation of nitrite close to artesian springs in the lowland of the catchment (Study II). Similar observations were reported for nomad settlement sites that had higher grazing pressure and were connected to loss of topsoil and higher nitrogen input (Miehe et al., 2008).

The Qugaqie catchment showed less depleted $\delta\ ^{13}C$ of DOM (-20.0 ‰) associated with lower Ai_{mod} (0.32), generally indicating a higher contribution of microbial-derived DOM. Further, more P containing formulae and higher input of unsaturated O–poor molecular formulae were found in DOM of Qugaqie compared to Niyaqu and Zhagu, respectively (Study III, Table 2; Figure 2). This points at high bioavailability of this DOM, as was suggested by previous work by Spencer et al. (2014). Meanwhile, a phenolic fraction of DOM was found, also showing a considerable input of plant- and soil-derived DOM considered more recalcitrant. DOM characteristics of the Qugaqie catchment showed indications for a larger input of biolabile, microbial-derived DOM, while another fraction of DOM was particularly rich in phenols. This was interpreted to represent three DOM sources, a microbial derived DOM source originating from the glacial microbiome. Alongside plant- and soil-derived inputs from *K. pygmaea* pastures were found. As well as phenolic compounds released with glacial effluents originating from aeolian deposition of either locally derived dust sources or black carbon from the burning of fossil fuels.

The Niyaqu catchment showed strong indications of a considerable input of plant- and soil-derived DOM by indicators from all studies, further Niyaqu samples in the low response group strongly corresponded to a phenol-rich (or *so-called* humic-like) FDOM component (FC 4; Study II, Figure 8d) in NDMS ordination, while the high response group responded to $SUVA_{254}$ as an aromaticity indicator. The significantly highest plant cover in the Niyaqu catchment (Study II, Supplementary materials,

Table S1), mostly associated to *K. pygmaea*, underlines that here DOM characteristics are strongly connected to the plant and soil related input from alpine pastures, stressing the importance of the terrestrial-fluvial pathway of DOM. The presented differences from study II and III between stream DOM characteristics of catchments indicate, that the described geologic predispositions, through i.a. orientation and steepness are influential on DOM characteristics. Further, the changing degree to which catchments are superimposed by hydrological and climatic forcing, vegetation, land degradation and human-induced effects of pastoralism have considerable effects on DOM composition as shown in studies II and III. These findings are in line with hypothesis **H1**.

Besides differences resolved on catchment-scale, study II and III aimed to investigate site-specific effects of bio-geodiversity as drivers of DOM biogeochemistry. In study II and III, endmembers, defined here as potential sources of river water, were sampled (Study II, Supplementary materials, Figure S1; Study III, Figure 1). For the endmember of glacial effluents, samples from Zhadang glacier in the Qugaqie catchment and a smaller glacier in the higher realms of the Niyaqu catchment were sampled. Spring water was sampled from the north-eastern Zhagu catchment, study II employs three spring samples, two from the degraded lowlands, with high-grazing pressure (Study II, Supplementary materials, Figure S7, Figure S8) and one from the hilly upland, featuring intact alpine pastures. Study III employs only the latter sample. Further, the water of an open wetland in the Niyaqu catchment was sampled for both studies.

Glacial effluents

Similar results were yielded in describing glacial effluents by study II and III. In study II, DOM drawn from the glacial mouth had high $SUVA_{254}$ (4.9 L mg C^{-1} m^{-1}) corresponding to relatively high aromaticity indices (Ai_{mod} =0.31, DBE = 9.15) in Study III. These results were seemingly exceptional, because the glacial foreland was free of vegetation cover. Along with other work (Spencer et al., 2014; Takeuchi, 2002) an aeolian deposition of plant- and soil-borne or black carbon-rich, fossil fuel derived particles was suggested in studies II and III to explain the terrestrial-like DOM signatures of the glacial environment. Besides, glacial DOM showed less depleted $\delta^{13}C$ values (-24.21 ‰) in study II, as compared to DOM sampled further downstream. This was accompanied by relatively high concentrations ($2.31 * 10^4$ relative intensity) of a microbial-like FDOM component (FC4 low fluorescence response dataset), hinting at a microbial derived DOM signature. Correspondingly, in study III, glacial effluent DOM had a high number of molecular formulae, hinting at a diverse composition, which was rich in P heteroatoms (133 formulae of total 2965) compared to other sample categories (Study III, Table 2). Alongside, glacial DOM was rich in unsaturated N containing molecular formulae (8%). These parameters suggest a second DOM source, showing microbial-like characteristics, pointing at formation of DOM in the glacial environment, e.g. by autotrophic organisms. Microbial activity in glaciers, so-called 'ice-cold hot spots' (Edwards et al., 2013) connected to DOM signatures, were reported by several authors (Anesio et al., 2009; Cook et al., 2016; Hodson et al., 2008; Hood et al., 2009). Summing up, both studies yield similar results, hinting at a duality of sources for glacial effluents, showing signs of an allochthonous terrestrial- or fossil fuel-derived DOM source, probably deposited by wind, together with an autochthonous source making DOM formation subsequent to biological activity in the glacial biome likely. The reported duality of sources, make glacial DOM diverse and complex.

Spring water

While study II employed water samples from three springs of the Zhagu catchment, two lowland springs in a degraded environment and one upland spring in intact *K. pygmaea* pasture surrounding, only the upland spring was investigated in study III. The latter work showed a relatively low molecular diversity of spring water (2819 formulae in total; Table 2) with 96% of molecular formulae suggesting plant- or soil-borne input such as from lignin, tannins and other phenol-associated structures. Correspondingly, Ai_{mod} was high (0.35) also suggesting this source. Alongside, spring DOM in the Zhagu upland was low in N and P heteroatoms, hinting to the filter function of the aquifer (Conley et al., 1995), hampering the formation of dissolved nitrogen and dissolved phosphorous. In total, upland spring DOM was characterised to contain mostly refractory, plant-influenced DOM probably originating directly from the source area. The larger sample set of study II showed a different picture. Spring DOM showed high concentration in microbial-like FDOM (FC1, FC2, FC3 –low subset; Study II, Table 2) and high values for nitrate (3.37 mg L^{-1}) and occurrences of nitrite (0.01 mg L^{-1}) while $SUVA_{254}$ as an indicator of aromaticity, was only medium high (3.45 L mg C^{-1} m^{-1}). Correspondingly, $\delta^{13}C$ of DOM for spring samples was less depleted (-24.22 ‰) (Study II, Table 2) compared to the potential plant (-26.8 ‰) and groundwater sources (-25.8 ‰) (Study II, Figure 5). This light fractionation can be caused by microbial decomposition processes and by the input of yak faeces: Especially the input of yak faeces is likely, because animals flock at the springs in the otherwise dry and unproductive catchment. Studies under laboratory conditions (Amelung et al., 1999) showed that enrichment of ^{13}C in yak dung takes place by digestion of ungulates, explaining the light fractionation in spring DOM. Yak dung was reported to be high in nitrogen (Du et al., 2021; Ma et al., 2013) matching with the higher content of N_{min} in water samples from lowland springs. Connected findings of studies II and III allow to trace the influence of intense pastoral activity on DOM characteristics. The upland spring sample, investigated with the ultra-high resolution approach in study III showed low molecular diversity attributed to the filtering of the shallow aquifer (Tran et al., 2021) and DOM primarily derived from the influence of plants, while the two lowland springs showed pronounced influence of animal husbandry and the influence of microbial reworked DOM associated with the input of nitrogen likely originating from yak faeces (see Study II, Supplementary materials, Figure S7 / S8 for supporting documentation).

Alpine wetland water

DOM from the standing water column of an alpine wetland of Niyaqu was investigated for both studies (Study II, supplementary materials Figure S1; Study II Figure 1). The non-metric multidimensional scaling (NMDS) of study II delivered results, plotting the wetland samples with a large ordination difference compared to other samples (Study II, Figure 8b) showing that they have a distinct composition, while NMDS results of study III placed these samples in the middle of the cluster core (Study III, Figure 5). This hints to the fact, that parameters of study II are better suited to describe specific properties of wetland DOM compared to the molecular assay of study III. Study II showed that wetland DOM had a high concentration of DOC and a $\delta^{13}C$ signature of DOM that was close to the C3 plant signal, as well as high abundances for three phenolic-rich FDOM components (FC1, FC2, FC4-high fluorescence response dataset) (Study II, Table 2 / Figure 8b). These parameters hint at an influence of plant-borne compounds, as was also suggested by the study of Li et al. (2021). This notion was only partly corroborated by study III, where the proportion of aromatic compounds in wetland DOM with ~14 % was overall within the range of samples from other sources (Study III, Figure 3), along with this $SUVA_{254}$ of wetland DOM in study II is lower compared to glacial and spring DOM, while also the microbial-like FDOM component (FC 3) of the high fluorescence response dataset showed highest values in wetland DOM (Study II, Table 2). Correspondingly, the molecular composition of wetland DOM was most complex for all endmembers in study III, with 4091 molecular formulae assigned, which is almost 30 % more than was found in stream DOM (Study III, Table 2). Along with this, study III revealed a high number of N and S containing molecular formulae, wetland DOM was relatively rich in saturated, unsaturated O-rich and unsaturated N containing formulae, reflecting the contribution of carbohydrates, amino sugars and proteins (Mann et al., 2015; Minor et al., 2014), while P was in limitation, as usual for productive terrestrial systems (Vitousek et al., 2010). Further, NH_4 was significantly increased compared to other sample categories (0.17 mg L^{-1}; Study II, Table 2) correlated to high N contributions in natural organic matter as reported by further studies (Thomas et al., 1995). Summing up, studies II and III found alpine wetland DOM to be influenced by *in-situ* production of plant biomass, together with input from microbial decomposition activities. Alpine wetlands show a large variety of chemical compounds, probably also related to the basin topography, where large amounts of DOM are received from seepage. Alpine wetlands hence are a hotspot of natural organic matter diversity and can greatly influence oligotrophic environments such as the lake, when wetland DOM is released to streams in greater quantities.

Finally, endmember groups show very different DOM characteristics, resolved by the two studies and by utilising a diverse combined dataset. These findings underline the postulated influence of site-specific properties and are thus corroborating hypotheses **H1**.

Influence of seasonality on DOM characteristics

The influence of seasonality was tracked by the three sampling campaigns conducted in paper II. The study covered the freshet phase of 2019 (sampled in May), the phase of the Indian summer monsoon (ISM), sampled in June/July 2018 and the phase of baseflow after the monsoon (sampled in September 2019). During freshet, higher concentrations of N_{min} (ammonium, nitrite and nitrate) as well as higher oxalate concentrations were shown, while DOC concentrations were lower (Study II, Table 1). Markers of plant-borne DOM were changed during freshet in study II: $SUVA_{254}$ in water samples was significantly increased compared to the other two seasons, while the signature of $\delta^{13}C$ of DOM was more depleted compared to the ISM season, indicating a larger influence of plant-derived, phenolic compounds in freshet DOM. This notion was further corroborated by an decrease of microbial-like FDOM components (FC1, FC2 of the low fluorescence response dataset) and a concomitant increase of phenol-rich FDOM (FC2, FC4 of the high fluorescence response dataset), also hinting towards greater influence of plant-borne compounds in DOM. These parameters indicate a flush of plant-borne compounds to be associated with snow melt at freshet, a notion that was also reported for boreal and tundra biomes (Lafrenière and Sharp, 2004; Shatilla and Carey, 2019; Prokushkin et al., 2011). Plant-derived freshet DOM was also connected to higher nitrogen and oxalate concentrations, indicating, that especially young and nutrient-rich plant material and low-molecular plant exudates are entering the streams, this was also stated by Amon et al. (2012) for larger rivers of the Arctic. As is in line with **H2**, freshet effects were observed to change water chemistry and characteristics of DOM.

During the ISM season, DOC concentrations were found about 30% increased compared to freshet and baseflow. This was accompanied by a change in the composition of DOM towards a shift of higher input of microbial-altered DOM by several parameters. $\delta^{13}C$ of DOM was strongly enriched (up to +7 ‰) (Study II, Figure 5) as compared to both other seasons. This fractionation is strong compared to other studies from the TP (Song et al., 2020). Concentration of PARAFAC resolved FDOM components responded to this, as higher contributions of microbial-like FDOM were reported (FC1, FC2 low fluorescence response dataset) during the ISM season. Along with the higher input of microbial-like DOM, proxies for plant-derived DOM decreased, $SUVA_{254}$ showed a depletion by a three- and two-fold, compared to freshet and baseflow, respectively (Study II, Table 1). In total this was interpreted as an ISM induced flush of natural organic matter, visible by higher DOC concentration, together with a change of DOM sources by leaching of microbial-derived compounds from the soil column and washing away of biofilms from water bearing structures, such as small ponds or wadis. As was postulated in **H2**, during the ISM, a profound shift in DOM characteristics was observed in Study II.

Baseflow conditions are connected to the periods outside of freshet and ISM. Study II investigated baseflow conditions compared to the two other seasons and found decreased DOC concentration in water samples (Study II, Table 1) along with a more depleted ^{13}C signature (-26.5 ‰; Study II, Figure 5) compared to the ISM season (-20.4 ‰). Concurrently with this, $SUVA_{254}$ increased by 2.5 fold and highest phenol-rich FDOM concentrations (FC1, FC2 and FC4 high fluorescence response dataset) were found in Study II (Table 1, Figure 7b, Figure 8c). This indicates, that after the substantial leaching of microbial-borne DOM and high DOC flushes during the ISM, the system changes again towards smaller releases of plant-borne natural organic matter, as probably influenced the *K. pygmaea* pastures. In summary, results of Study II suggested unique changes of DOM composition and processing with respect to seasonality. Freshet played a smaller role, as compared to boreal and tundra biomes, where snow-melt is an important factor governing changes in DOM composition. The ISM in contrast had large effects on DOM composition for small watersheds of this Tibetan study area, with a high contribution of microbial-derived DOM, while baseflow conditions after the ISM represent the influence from the *K. pygmaea* biome by input of small amounts of plant-borne DOM. These finding are in line with hypothesis **H2**, stress the importance of seasons for DOM investigations and highlight the importance of the ISM for DOM dynamics in the Nam Co watershed. Changes in the onset of the monsoon and in monsoon intensity, as are reported (Liu et al., 2019; Yao et al., 2019) can have tremendous consequences for DOM dynamics and by this also influence the lake ecosystem.

Transformation and processing of DOM

DOM characteristics are subject to changes in all ecosystems, such as during percolation in the soil column (Kaiser and Kalbitz, 2012) or during fluvial transport in rivers (Aufdenkampe et al., 2001; Perdrial et al., 2010), estuaries (Seidel et al., 2015) and the open ocean (Kowalczuk et al., 2010). The chromophoric DOM fraction is especially suited to track the transformation and processing of DOM through UV/VIS proxies, especially if photooxidation takes place (Hansen et al., 2016). While changes in the isotopic composition of organic carbon allow the investigation of a wide range of fractionation processes, such as the utilisation of compounds by microbes (Balesdent et al., 1987; Pettersson et al., 1997; Wang et al., 2019). Fluorescence spectroscopy can track the response of fluorophore composition to bio- and photodegradation (Chen et al., 2019; Hernes et al., 2009), while ultra-high resolution mass spectrometry allows to track changes in the molecular composition of DOM as caused by processing and transformation and hence is a promising methodological approach

(Miranda et al., 2020; Seidel et al., 2015). The degree of chemical alteration, that processing causes on DOM is an important parameter to assess the potential fate and impact of DOM when entering ecosystems, such as here from terrestrial ecosystems to the oligotrophic Lake Nam Co, leading to the formulation of two hypothesis (**H3** and **H4**) tested by study II and study III.

DOM processing during transport in streams

Study II found signs of processing of DOM in the Qugaqie catchment. Here, the signal of the glacial endmember, superimposing DOM characteristics in the headwaters of the catchment, gradually vanishes along the flow path. Correspondingly, $SUVA_{254}$ was reduced by 30 % from the glacial endmember compared to the stream, as well as the phenol-rich FDOM component (FC 4 low fluorescence response dataset) which decreased by 25%. The initially high ammonium concentrations vanished (Study II, Figure 8e, Table 1, Table 2). These changes were attributed to microbial utilisation of N_{min} and to photooxidation of DOM, leading to the reduced indication of phenolic compounds by $SUVA_{254}$ and FDOM. Further, a more depleted signature in ^{13}C of DOM (-2.9 ‰ by comparing the glacial effluent mean: -24.2 ‰ to the mean of baseflow conditions: -27.1 ‰) during baseflow suggests the concomitant input of plant-borne compounds along the stream path, probably from *K. pygmaea* pastures. Findings of study II suggest a compositional change and processing of glacial DOM during the flowpath. However these changes were less pronounced than indicated for larger river systems of boreal and tundra biomes (Perminova et al., 2019; Riedel et al., 2016; Shatilla and Carey, 2019) and also driven by the additional input of alpine pasture signals. Consistently, study III allowed the discrimination of brackish samples in the intermixing zone of streams and the lake (Study III, Figure 1, Figure 5). These samples still showed a strong influence of terrestrial signals not to be statistically discriminated from stream DOM (Study III, Supplementary materials, Table S2). This was interpreted as a clear indication of limited processing of terrestrial-borne DOM in the streams, as DOM signatures from this origin intermix with lake water in the brackish zone. Further, no processing of DOM was evident in catchments other than Qugaqie in study II, which was explained by the permanent input of *K. pygmaea* derived compounds. The problem of gradual inputs is especially pronounced in smaller watersheds, where stream water has a short residence time, hampering processing as was pointed out by other authors (Roebuck et al., 2020). Concluding, **H3** could neither be fully corroborated nor completely rejected for our investigation, because of the ongoing site-specific input of DOM compounds. Our investigations did reveal however, that glacial-borne DOM is altered during fluvial transport and that especially labile compounds in water, such as N_{min} are utilised and by this unlikely to enter the fragile oligotrophic lake ecosystem.

DOM processing in the large endorheic Lake Nam Co

Lake water was obtained from the pelagial at 30 m depth and from the lake surface (Study II, Supplementary materials, Figure S1; Study III, Figure 1). Water chemistry parameters and DOM of lake water were found to be distinctive from all other investigated catchments and sources in both studies, as revealed by plotting distance in NMDS ordination (Study II, Figure 8d; Study III, Figure 4). In study II, water samples from the Nam Co lake showed higher concentrations in anions, cations, and DIC compared to other samples (Table 1), this was related to the evapoconcentration effect for salts as well as to the geologic formation in the Nam Co basin, reported to be rich in carbonaceous matter (Wang et al., 2010) with respect to the lime carbonic acid equilibrium. Along with this, calcium was found to be a geodiversity marker for the eastern Nam Co watershed (Yu et al., 2021), water in Lake Nam Co was comparably poor in calcium (Study II, Table 1), showing that the lake is unaffected by the water chemistry of inflowing streams. DOM composition of inflowing streams entered the lake in the brackish intermixing zone, as was displayed in study III (Figure 3). These water samples still bore signs of terrestrial influence, as was visible by the high relative abundance of aromatic and condensed aromatic compounds, likely originating from plant input in the catchments. When compared to DOM of lake water, a significant decrease of aromatic O-rich compounds was evaluated in study III (Supplementary materials, Table S2), alongside to a decrease in molecular compound classes which represent a high contribution of phenols: condensed aromatic, aromatic and highly unsaturated O-rich formulae (Mann et al., 2015; Minor et al., 2014). These classes accounted only to 42 % of the total molecular composition in lake DOM, compared to 60 % in stream samples (Study III, Figure 2, Figure 3). Correspondent, decreases of aromaticity indices such as Ai_{mod} (by 30 %) and DBE (by 20 %) compared to values in the stream samples were observed (Study III, Table 2). Study II corroborated this, by 90 % loss in the $SUVA_{254}$ index, when comparing lake and stream DOM. Accordingly the phenol-rich FDOM component FC4 (low fluorescence response dataset) decreased by 65 % (Study II, Table 1). In both studies, the immense depletion of phenolic compounds and aromaticity indices was interpreted as the effect of photooxidation of DOM, especially of chromophoric DOM, in the clear and deep lake water column. This was demonstrated in both, laboratory experiments (Helms et al., 2014; Weishaar et al., 2003) and field studies (Kowalczuk et al., 2010; Spencer et al., 2009; Tfaily et al., 2015). The combined findings of study II and III support hypothesis **H4a**, where preferentially depletion of chromophoric DOM was postulated as a processing indicator.

Further, data of study II and III do not only suggest processing of terrestrial-borne DOM in the lake, but also a distinct DOM source for lake water. Study II found that lake water had a higher DOC concentration by almost 50 % and DOM was enriched with up to +5 ‰ compared to stream DOM during freshet and baseflow conditions (Study II, Table 1), this was interpreted to suggest a change in the DOM source towards autochtonous algae or microbial-derived DOM. Study III underlined this interpretation, as lake DOM was investigated to be relatively enriched in unsaturated molecular compounds, 11 % in lake DOM, compared to 6 % in stream DOM, resembling a higher share of lipids and carbohydrates (Mann et al., 2015; Minor et al., 2014)in the molecular composition of lake DOM. Further, heteroatoms (S, N, P) indicating labile DOM (Chen et al., 2016) , such as enriched in natural organic matter by microbes (Feng et al., 2016), were relatively abundant in lake DOM compared to stream DOM (Study III, Table 2). Hypothesis **H4b** was corroborated by both studies (II and III), as less depleted signatures in ^{13}C of DOM conjointly with a lower-molecular weight composition of DOM and relative increase of heteroatoms point towards a microbial derived, and by this, independent DOM source.

Conclusion and outlook

In this thesis, DOM characteristics, processing and the transformation of DOM were investigated in the High Asian endorheic lake watershed of Nam Co, Tibet. Following an extensive review of current literature in this field, it was found that DOM was understudied on the TP despite its important function in linking ecosystems, such as the glacial, terrestrial and aquatic sphere. Subsequent, the monitor function of DOM was investigated, to draw conclusions about processes in the source areas, such as the influence of seasonality and site-specific characteristics, for example, the degree of glaciation, plant cover and pasture degradation as well as impacts of alpine wetlands. For this, water samples from three diverse catchments and the lake were taken. A multiparameter dataset including: FT-ICR MS, PARAFAC-resolved FDOM, $\delta^{13}C$ of DOM, $SUVA_{254}$, DOC, DIC, EC, pH and analysis of cations and anions was employed to investigate the impact of landscape units such as glaciers, springs, alpine wetlands, the stream course, the brackish mixing zone and the oligosaline lake on DOM composition. The aim was at showing that bio-geodiversity in the form of site-specific catchment properties and seasonality modifies DOM characteristics at catchment scale, while DOM is processed until the terminal aquatic basin. Three sampling campaigns were conducted, in order to examine seasonal climatic controls of DOM, influenced by freshet, the phase of the Indian summer monsoon (ISM) and the late year baseflow conditions. Further, a watershed-wide plant cover estimate was calibrated on 30m resolution, allowing to track significant changes in plant cover over a 30 year period and to assess the approximate distribution of biome types in the study area. The results underline a greening trend for the TP, reported for the latest decades. Yet, greening was prevalent mainly in the alpine steppe biome, whereas the alpine pastures remained mostly unchanged. Driven by this, significant differences in total plant cover were evaluated for the catchments under investigation, underlining the aridity gradient reinforcing towards the northern, inward plateau.

Results showed, that site-specific characteristics in endmembers of the catchments were strongly influencing DOM characteristics at a local level. Correspondingly, unique DOM signatures were found at glaciers, alpine wetlands and springs: Glacial effluents were characterised by a unique dual source, showing high index values and abundance of phenolic compounds, pointing at a depositional source of local dust or aeolian transported black carbon derived from burning of fossil fuels. While, glacial DOM was rich in low-molecular weight compounds, containing P and N, this suggests contribution of peptides and amino sugars as well as high bioavailability of microbial derived DOM of autotrophic activity in the glacier.

Conclusions and outlook

DOM of spring water in the uplands showed low molecular diversity, mostly dominated by aromatic compounds and highly unsaturated plant or soil derived compounds, while springs in the more arid lowlands suggested inputs from pastoral activity, such as yak faeces. This shows that human activities can profoundly impact DOM composition in this remote High Asian watershed.

DOM composition in alpine wetlands was different compared to other sample categories, showing high molecular diversity and suggesting inputs from vascular plants, which was modified in soil, as well as microbial activity in the standing water column of the wetland. The basin topography of the wetland led to a broad range of terrestrial DOM signatures, in addition to a high load of anions and cations, probably received by input from surrounding soils. Wetlands are a diversity hotspot of DOM, with indications of eutrophic conditions and hence can pose a danger to the sensitive, oligotrophic lake ecosystem.

Correspondingly to endmembers, the molecular diversity of DOM was largest in proglacial streams of the glaciated Qugaqie catchment, while DOM signatures in the Niyaqu and Zhagu catchment had lower molecular diversity. The latter two catchments were greatly influenced by the input of highly unsaturated as well as aromatic structures, connected to the input of soils or plants, especially from the *K. pygmaea* pasture biome, which stretches along the streams. Besides site induced influences on DOM characteristics, the results further showed that seasonality had a decisive influence on the molecular composition of DOM. Seasonal variations of DOM characteristics suggest a strong climatic control, especially by the ISM, as the most important hydrological driver of the TP. During ISM, a flush of dissolved organic carbon (DOC) and a shift towards more microbial-derived DOM was observed. Effects of freshet were less pronounced compared to those of ISM. During freshet, a small input of young plant derived DOM was indicated alongside with elevated oxalate concentration. The limited influence of snowmelt was assigned to the dry and cold winters, resulting in an absence of snow and hampering DOM formation. Baseflow conditions of DOM were again characterised by plant- and soil-borne inputs from the alpine pasture biome and low concentrations of DOC.

The results suggested processing and transformation of DOM in two different stages: First, in proglacial streams of Qugaqie, the loss of labile DOM compounds and N_{min} was observed during stream path, while also a gradual input from alpine pastures overruled the glacial signal of DOM. Second, the investigation of the brackish mixing zone showed that the DOM signal of catchments is transported into the lake, while the pelagial of the lake showed a chemical distinction between DOM signatures of stream and lake water. The loss of catchment characteristics in lake DOM was interpreted as the influence of decomposition of phenolic DOM compounds in the lake, mostly by photooxidation. Processing of DOM predominantly takes place in the terminal aquatic basin and further, strong processing of DOM also renders the seasonal variability of DOM irrelevant to the lake.

Both was attributed to processing of terrestrial DOM in the well-intermixed lake. Additionally, DOM characteristics implied an autochthonous DOM source, derived from *in-situ* production by microorganisms and algae in Lake Nam Co.

The study showed that DOM is well suited to indicate catchment-specific influences on the composition of natural organic matter. Further the fate of DOM in the catchment was elucidated for this High Asian case study. DOM analysis allows a direct screening of catchment processes, such as the influence of seasonality and chemical composition in endmembers. The three catchments are diverse and influences of abiotic and biotic factors greatly modify the characteristics of DOM. The application of indices and proxy methods together with ultra-high resolution mass spectrometry was especially suited to identify compositional changes in DOM. Multi-parameter approaches are useful to discern and forecast the effects that climate change and anthropogenic land-use have on the sensitive alpine ecosystems of the TP. The results indicate that water quality in the Nam Co watershed is potentially threatened by changing influences from the Indian summer monsoon or the degradation of alpine wetlands. Safeguarding of freshwater resources and the sensitive oligotrophic Lake Nam Co environment can be facilitated by monitoring DOM. In the future, DOM investigations should be continued on a long-term basis, by correlating the results of this multi-parameter application to a simplified *in-situ* monitoring toolbox. A high-frequency assessment of DOM characteristics with high temporal and spatial resolution will allow an improved understanding of fluxes and is a promising approach to transfer the findings presented here to larger parts of the sensitive southern TP.

References synthesis, conclusion and outlook

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