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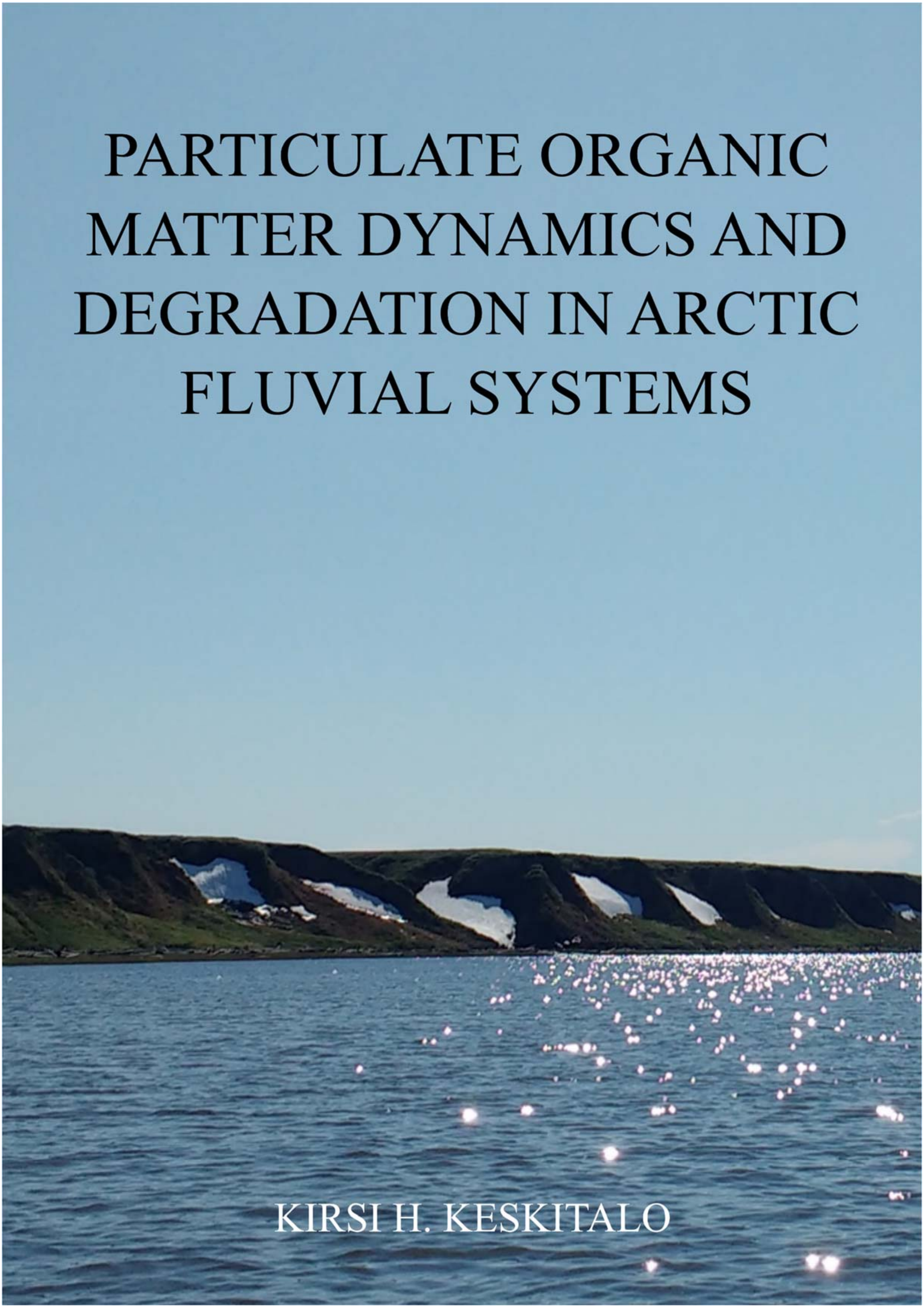

PARTICULATE ORGANIC MATTER DYNAMICS AND DEGRADATION IN ARCTIC FLUVIAL SYSTEMS

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Particulate organic matter dynamics and degradation in Arctic fluvial systems

PhD Thesis, Department of Earth Sciences, Faculty of Science, Vrije Universiteit Amsterdam, The Netherlands

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Cover: Kolyma River by Kirsi H. Keskitalo

VRIJE UNIVERSITEIT

PARTICULATE ORGANIC MATTER DYNAMICS AND DEGRADATION IN ARCTIC FLUVIAL SYSTEMS

ACADEMISCH PROEFSCHRIFT

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door

Kirsi Helena Keskitalo

geboren te Oulu, Finland

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Publication I:

Keskitalo K H, Bröder L, Shakil S, Zolkos S, Tank S E, van Dongen B E, Tesi T, Haghipour N, Eglinton T I, Kokelj S V and Vonk J E. 2021. ***Downstream evolution of particulate organic matter composition from permafrost thaw slumps.*** *Frontiers in Earth Science, 9, 1-21. https://doi.org/10.3389/feart.2021.642675*

I participated to the study design together with my supervisor and collected the samples on a fieldwork campaign together with co-authors. I executed all the laboratory work from lipid extractions to pyrolysis (except grain size analysis), and prepared samples for OC, $\delta^{13}C$ and $\Delta^{14}C$ analyses with support from co-authors. I was the lead author of the study with valuable contribution to the writing from all the co-authors.

Publication II:

Keskitalo K H, Bröder L, Jong D, Zimov N, Davydova A, Davydov S, Tesi T, Mann P J, Haghipour N, Eglinton T I and Vonk, J E. 2022. ***Seasonal variability in particulate organic carbon degradation in the Kolyma River, Siberia.*** *Environ. Res. Lett., 17, 034007 doi:https://doi.org/10.1088/1748-9326/ac4f8dt*

I executed all the experimental work (i.e., incubations) with support from a co-author and developed and fine-tuned the method with my supervisor and a co-author. I collected the samples on two field campaigns over two consecutive years with support from co-authors. I executed all the laboratory work, source apportionment modelling, data analysis and statistical analysis. All the co-authors contributed to the writing.

Publication III (submitted):

Keskitalo K H, Bröder L, Tesi T, Mann P J, Jong D, Bulte Garcia S, Davydova A, Davydov S, Zimov N, Haghipour N, Eglinton T I and Vonk J E. 2022. ***Seasonal carbon dynamics of the Kolyma River tributaries, Siberia.***

I collected the samples during two fieldwork campaigns with support from co-authors. I executed all the laboratory work, source apportionment modelling, spatial analysis with support from a co-author, statistical analysis, data analysis and was the lead author of the study. All the co-authors contributed to the writing.

Summary

The Arctic is warming two to four times the rate of global average. The increase in air temperatures causes permafrost (i.e., perennially frozen ground) to thaw and release previously frozen organic carbon (OC) to the contemporary carbon cycle. Permafrost stores large amounts of organic carbon (~1300 ± 200 Pg), which equals up to half of the belowground OC globally. Re-mineralization of the released permafrost OC can add greenhouse gases (CO_2, CH_4) to the atmosphere enhancing climate warming.

Gradual permafrost thaw happens when the active layer (i.e., the topmost layer of permafrost that thaws during summer months) deepens due to climate warming releasing largely dissolved organic carbon (DOC). On the contrary, in permafrost regions with high ground ice-content, permafrost thaw happens abruptly (i.e., thermokarst) as landscapes subside or collapse due to melting of ice. Abrupt permafrost thaw releases dominantly particulate organic carbon (POC). While degradation of DOC has been extensively studied in Arctic fluvial systems, degradation of POC is still poorly characterized.

In this study, we investigate POC composition and degradation in two different areas: i) in the thaw streams draining abrupt permafrost thaw features, retrogressive thaw slumps (RTS), on the Canadian Peel Plateau, and ii) in the Kolyma River, which is one of the major Arctic rivers draining to the Arctic Ocean. We also study carbon dynamics and water chemistry parameters in lower order streams within the Kolyma watershed in two hydrologically distinct seasons: spring freshet and summer. We use macro(molecular) methods, pyrolysis – gas chromatography mass spectrometry and lipid biomarkers (*n*-alkanes, *n*-alkanoic acids), to analyse POC composition and degradation status. For further compositional analysis, we use carbon isotopes ($\delta^{13}C$, $\Delta^{14}C$). Additionally, we employ spatial data analysis and statistical modelling to characterize the watersheds and POC sources.

Our results indicate that POC composition is seasonally dependent, and it defines biodegradability of POC. On the Peel Plateau, POC consists largely of aromatic moieties and includes petrogenic carbon that are not easily degradable. By contrast, Kolyma River POC degrades relatively fast during summer, when it is mostly of autochthonous sources. However, freshet POC, dominated by allochthonous POC, is not readily degradable. During freshet, DOC is susceptible to adsorption to particles and/or flocculation, potentially attenuating its climate impact. The lower order streams within the Kolyma River watershed react fast to increase in air temperatures during spring freshet with increased surface water temperatures and depletion in $\delta^{13}C$-POC, suggesting early onset of primary production. Changes in water temperature and $\delta^{13}C$-POC were not as pronounced in the Kolyma River. These results suggest that lower order

streams may start primary production and POC degradation earlier in the season than the larger ones and thus, start emitting greenhouse gases earlier. The degraded POC is mostly autochthonous, and more studies are needed to investigate whether degradation of autochthonous POC may stimulate degradation of allochthonous or permafrost POC. These results highlight the heterogeneity of the Arctic fluvial networks and the differences in their response to climate warming.

Tiivistelmä

Arktiset alueet lämpenevät kahdesta neljään kertaa globaalia keskiarvoa nopeammin. Ilmaston nopea lämpeneminen aiheuttaa pohjoisten ikirouta-alueiden (ts. alueiden joiden maa pysyy jäässä vähintään kahtena peräkkäisenä vuotena) sulamista. Ikirouta-alueiden varastoiman orgaanisen hiilen määrä (~1300 ± 200 Pg) kattaa yli puolet maailmanlaajuisista maanalaisista hiilivarastoista. Kun ikiroutaan sitoutunut orgaaninen hiili vapautuu sulamisen seurauksena takaisin aktiiviseen hiilen kiertokulkuun, se voi aiheuttaa kasvihuonekaasupäästöjä (CO_2, CH_4) ilmakehään mikrobien hajottaessa sen sisältämää orgaanista ainesta. Ikiroudan sulamisen seurauksena syntyvät päästöt voivat vahvistaa ilmaston lämpenemistä.

Ikirouta sulaa ylhäältä alaspäin, kun sen 'aktiivinen kerros' eli vuosittain sulava, lähinnä maanpintaa oleva kerros, syvenee ilmaston lämpenemisen seurauksena. Ikiroudan aktiivisen kerroksen syveneminen vapauttaa orgaanista hiiltä pääasiassa liuenneessa muodossa (dissolved organic carbon; DOC). Asteittaista sulamista tapahtuu kaikilla pohjoisen ikirouta-alueilla. Sitä vastoin, ikiroudan äkillistä sulamista tapahtuu alueilla, joiden maaperän jääpitoisuus on korkea. Ikiroudan jään äkillinen sulaminen johtaa valtavien kraattereiden (ts. sulakuoppien) syntymiseen maaperän nopean luhistumisen seurauksena, minkä vaikutuksesta vapautuu orgaanista hiiltä pääasiassa partikkeli-muodossa (particulate organic carbon; POC). Ikiroudasta peräisin oleva DOC hajoaa nopeasti, mutta POC:n hajoamista ja sen ilmastovaikutuksia on tutkittu vasta vähän.

Tässä tutkimuksessa perehdytään POC:n hajoamiseen kahdella eri alueella: ikiroudan sulamisesta syntyneiden kraattereiden sulapuroissa Pohjois-Kanadan Peel Plateau -alueella sekä Kolyma-joessa, joka on yksi Siperian suurimmista Jäämereen laskevista joista. Lisäksi tutkimme orgaanisen hiilen kiertoon liittyviä prosesseja ja hiilen koostumusta Kolyma-joen sivujoissa, kahtena hydrologisesti toisistaan poikkeavana vuodenaikana, kevät-tulvien aikaan ja kesällä. Tutkimuksessa käytettiin useita tutkimusmenetelmiä POC:n koostumuksen ja sen hajoamiseen vaikuttavien tekijöiden ymmärtämiseksi. POC:n makromolekylaarisen koostumuksen tarkastelussa käytettiin pyrolyysi-kaasukromatografi-massaspektrometriaa sekä lipidi (ts. rasvamolekyyli) biomarkkereita (alkeenit ja karboksyylihapot). Lisäksi tutkimuksessa käytettiin hiilen isotooppeja ($\delta^{13}C$, $\Delta^{14}C$) hiilen iän ja alkuperän selvittämisessä, sekä spatiaalisia analyysimenetelmiä ja tilastotieteellistä mallinnusta.

Tutkimustuloksien perusteella POC:n koostumus vaihtelee vuodenajasta mukaan ja vaikuttaa partikkeleiden hajoamiseen. Kanadan Peel Plateau -alueella POC koostuu lähinnä aromaattisista yhdisteistä sekä sisältää petrogeenistä hiiltä, minkä vuoksi POC ei ole helposti

hajoavaa. Kolyma-joessa POC hajoaa nopeasti kesäisin, jolloin hiili on lähinnä vesistöissä yhteyttämisen seurauksena syntynyttä orgaanista ainesta. Kevät-tulvien aikaan POC on puolestaan peräisin maalta eikä hajoa helposti vesistöissä. Keväällä DOC sitoutuu herkästi maalta peräisin oleviin mineraalipartikkeleihin tai flokkuloituu (ts. DOC muodostaa partikkeleita sitoutuessaan toisiin liuenneisiin hiiliin). Liuenneen hiilen siirtyminen partikkelimuotoon voi mahdollisesti heikentää sen ilmastovaikutuksia. Kolyman sivujoissa δ^{13}C-POC arvot ovat matalia jo kevät-tulvien aikaan, mikä viittaa yhteyttämisen alkamiseen vedessä. Sitä vastoin Kolyma-joessa δ^{13}C-POC arvot eivät olleet erityisen matalia. Yhteyttäminen ja POC:n hajoaminen alkaa pienemmissä joissa todennäköisesti suuria jokia aikaisemmin, mikä mahdollisesti johtaa kasvihuonekaasupäästöjen aikaisempaan vapautumiseen. Suurin osa päästöistä lienee peräisin yhteytetyn hiilen hajoamisesta, tosin lisää tutkimuksia tarvitaan, jotta voidaan selvittää stimuloiko yhteytetyn hiilen hajoaminen myös maalta ja ikiroudasta peräisin olevan hiilen mikrobista hajoamista. Nämä tutkimustulokset korostavat arktisten virtavesien heterogeenisyyttä sekä eroavaisuuksia niiden vasteissa ilmastonmuutokseen.

List of abbreviations in alphabetical order

ANOVA – analysis of variance
C:N ratio – molar carbon to nitrogen ratio
CPI – carbon preference index
DEM – digital elevation model
DIC - dissolved inorganic carbon
DO – dissolved oxygen
DOC – dissolved organic carbon
EA-AMS - elemental analyzer - accelerator mass spectrometry
EC – electrical conductivity
EI – electron ionization
ETH – Swiss Federal Institute of Technology (Eidgenössische Technische Hochschule)
GC – gas chromatography
GF/F – glass fiber filter
HMW – high molecular weight
IAEA – International Atomic Energy Agency
IPCC – Intergovernmental Panel on Climate Change
IRMS – isotope ratio mass spectrometer
LIS – Laurentide Ice Sheet
LMW – low molecular weight
MCMC – Monte Carlo Markov Chain (statistical model)
OC – organic carbon
PES - polyethersulfone membrane filters
POC – particulate organic carbon
py-GCMS – pyrolysis - gas chromatography - mass spectrometry
RCP - Representative Concentration Pathway
RTS – retrogressive thaw slump
SA – mineral-specific surface area
SOCC – soil organic carbon content
SPM – suspended particulate matter
TN – total nitrogen
TOC – total organic carbon
TPN – total particulate nitrogen
TSS – total suspended solids
VPDB - Vienna PeeDee Belemnite

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Chapter 1

Introduction

1.1 Warming of the Arctic

Over the past decades the Arctic has warmed two to four times the rate of the global average (Meredith et al., 2019; Rantanen et al., 2021). The rapid warming of the Arctic, called Arctic amplification, started in the mid-20th century - prior to that the Arctic was still cooling (England et al., 2021). The increase in air temperatures imposes cascading effects across these northern landscapes from increase in precipitation (Bintanja et al., 2020) to changes in snow cover and albedo (Connolly et al., 2019), increase in the number of wild fires (Chen et al., 2021), changes in lake distribution (Smith et al., 2005) and lengthening of growing season (Collins et al., 2021). Some of these changes are induced by permafrost (i.e., perennially frozen ground) thaw, while others enforce thawing of permafrost. Since the last glacial period, permafrost has stored organic carbon (OC) underground, locked away from the contemporary carbon cycle. Warming of the climate, has driven temperatures in permafrost to rise across the Arctic over the past decades accelerating its thaw (Biskaborn et al., 2019). Permafrost thaw re-mobilizes previously frozen carbon deposits back to the contemporary carbon cycle. Mineralization of the newly thawed permafrost OC adds greenhouse gases (CO_2, CH_4) to the atmosphere and further enhances climate warming (e.g., Meredith et al., 2019; Schuur et al., 2008; Vonk and Gustafsson, 2013).

1.2 Permafrost distribution and different types of thaw

Permafrost covers ~24 % of the land surface of the northern hemisphere (Zhang et al., 2003) and holds ~1300 ± 200 Pg of OC, which is about half of the belowground organic carbon globally (Hugelius et al., 2014; Tarnocai et al., 2009). At the higher latitudes, permafrost cover is continuous (>91 %), while towards the lower latitudes the coverage drops to discontinuous (51-90%), sporadic (10-50 %) and isolated (<10 %) (Fig. 1; Obu et al., 2019; Tarnocai et al., 2009).

Permafrost thaw occurs either top-down as active layer (i.e., the top layer of permafrost that thaws seasonally) deepening or as abrupt thaw (i.e., thermokarst) causing rapid collapse of the landscape such as river bank erosion or retrogressive thaw slumps (RTS) (Schuur et al., 2015). Active layer deepening happens across permafrost landscapes, and it releases mainly dissolved organic carbon (DOC; Fig. 2A). Abrupt permafrost thaw is confined to areas with high ground ice content and it releases mainly particulate organic carbon (POC; Fig. 2B) (Bonnaventure and Lamoureux, 2013; Kokelj and Jorgenson, 2013; Shakil et al., 2020). In Siberia and Alaska, these ice-rich permafrost deposits are often referred to as Yedoma (Strauss et al., 2017). While active layer deepening is a gradual process, abrupt thaw is non-linear and more difficult to predict as just single weather events can trigger rapid thaw (Turetsky et al., 2019).

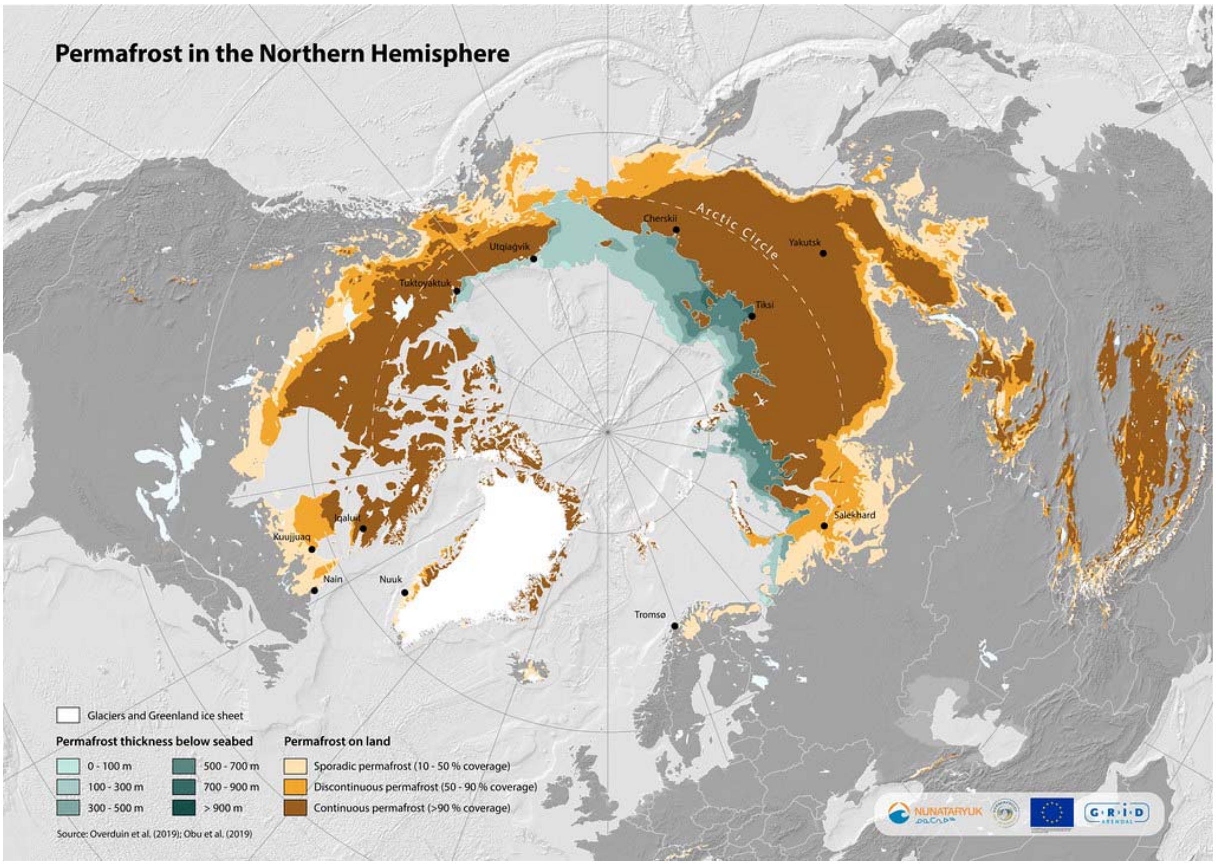

Figure 1. Permafrost distribution (sporadic, discontinuous, and continuous) across the Arctic. Also, permafrost thickness below seabed is shown. Map by GRID-Arendal/Nunataryuk, 2020.

Beside ice content, permafrost characteristics and vulnerability to thaw are partially defined by the landscape history (Tank et al., 2020). A large distinction in landscape history traces back to the last glacial period when part of the Arctic was covered with ice sheets while other parts remained ice-free (Lindgren et al., 2016). These differences in ice-cover still define the composition of permafrost today. The Canadian Arctic was largely enclosed by the Laurentide Ice Sheet (LIS) and therefore, the landscape is characterized by glacifluvial and glacio-lacustrine deposits and ice-rich tills (Kokelj et al., 2017a; Lacelle et al., 2013). Periglacial processes have shaped the landscape further, and particularly fluvial incisions add to the landscape's susceptibility to thaw slumping, due to the topographic relief enhancing erosional processes (Kokelj et al., 2017a). Especially glacial margins, such as the Peel Plateau, are highly prone to initiation of RTS features (Fig. 2C; Kokelj et al., 2017b). By contrast, the mammoth-steppe tundra of eastern Siberia remained ice-free during the last glacial period (Lindgren et al., 2016). There, permafrost is characterized by alluvial and aeolian loess deposits, mostly of poorly sorted silts (Strauss et al., 2012; Zimov et al., 2006). Without the ice cover, permafrost aggradation incorporated OC from vegetation, and thus the Siberian (and Alaskan) permafrost is very rich in OC compared to the glaciated landscapes in Canada (Ewing et al., 2015).

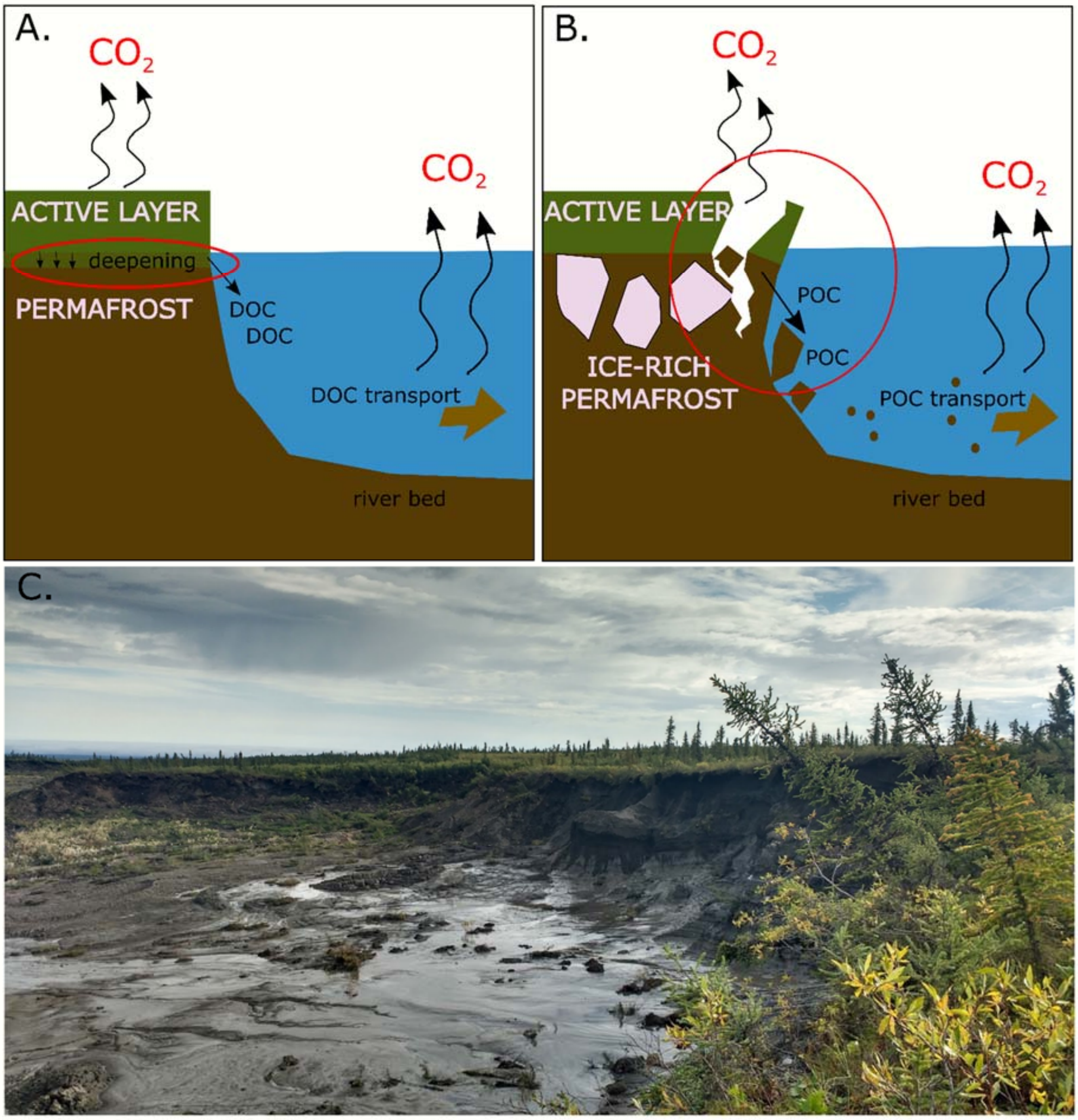

Figure. 2. A. Conceptual figure of gradual permafrost thaw as active layer deepens, and releases mainly dissolved organic carbon (DOC). B. Abrupt permafrost thaw that occurs in ice-rich permafrost areas causing rapid collapse of landscape such as riverbank erosion and thaw slumping. Abrupt thaw releases dominantly particulate organic carbon (POC). C. Retrogressive thaw slump on the Peel Plateau, Canada, a type of abrupt permafrost thaw. Photo credit of the author.

1.3 Estimates on greenhouse gas emissions from permafrost thaw

Permafrost thaw has been named as one of the planetary tipping points that may trigger large scale global changes (Lenton et al., 2019). Passing these critical thresholds exposes the whole Earth system to changes, which will impact ecological systems and societies (Lenton et al., 2008). Large-scale climate models are used to predict future climate under different warming scenarios ranging from low greenhouse gas concentrations (RCP2.6, i.e., Representative Concentration Pathway) to very high concentrations (RCP8.5) (Meredith et al., 2019). Near-surface permafrost area has been predicted to decrease by 2-66 % under the low warming scenario RCP2.6 or by 30-99 % under the higher warming scenario of RCP8.5 by the end of 2100 (Meredith et al., 2019). This level of permafrost thaw, is estimated to add 120 ± 85 Pg of greenhouse gases to the atmosphere by 2100, subsequently warming the planet by an additional 0.29 ± 0.21 °C (Schaefer et al., 2014). Some estimates are even higher, projecting up to 240 Pg of added greenhouse gases to the atmosphere (Meredith et al., 2019).

One of the largest uncertainties in climate models is that they exclude emissions from abrupt permafrost thaw and focus only on gradual permafrost thaw (Natali et al., 2021; Treharne et al., 2022). Given that about half of the permafrost carbon is stored in the areas that are susceptible to abrupt thaw, while covering only 20 % of the Arctic (Olefeldt et al., 2016), their inclusion to models is crucial for accurate predictions. Early estimates suggest that emissions from abrupt thaw may double the current permafrost emission estimates (Turetsky et al., 2019).

1.4 Fluvial systems integrate landscape signals and transport organic carbon

Arctic rivers are major conduits of fresh water to the Arctic Ocean, and they deliver 5800 Gg of POC and 34 000 Gg DOC annually to the Arctic Ocean (Holmes et al., 2012; McClelland et al., 2016). Arctic rivers and streams are characterized by their distinct seasonality. The rivers remain frozen during winter months with only baseflow, while the highest discharge peak occurs during the freshet period when ice breaks up at the end of the spring, and lowers again towards the summer (Shiklomanov et al., 2021). On land, permafrost largely controls the hydrological pathways, and most of the surface flow occurs within the active layer (Frey and McClelland, 2009). Delivery of allochthonous (i.e., terrestrial) OC from land to the fluvial system plays a crucial role in fuelling primary production in northern rivers and further in the Arctic Ocean (Terhaar et al., 2021; Wild et al., 2019).

Rivers act as integrators of biogeochemical signals across their watersheds. These fluvial channels are dynamic and modify river constituents during downstream transport (Cole et al., 2007; Drake et al., 2018a; Tank et al., 2020). Previous studies have shown that permafrost derived DOC degrades rapidly in the fluvial system (e.g., Mann et al., 2015; Textor et al., 2019; Vonk et al., 2013b). This degradation may be so rapid that it largely occurs in headwater streams, making the permafrost DOC signal hardly detectable in the major Arctic rivers, thus their emissions consist mostly of modern carbon (Dean et al., 2020; Wild et al., 2019). By contrast, the permafrost signal is traceable in the POC fraction of the Arctic rivers (Wild et al., 2019). Yet, degradation of POC has been poorly characterized, especially in the Arctic. Understanding potential degradation of POC is important, as thermokarst processes, releasing dominantly POC to fluvial systems, are accelerating due to climate warming. Thus, focal part of this thesis is to focus on POC composition, degradation, and dynamics in Arctic fluvial systems.

Arctic fluvial systems are changing in the warming climate. Discharge has been increasing over the past decades and is predicted to increase further (Peterson et al., 2002). Similarly, bioreactivity of OC is predicted to increase with increasing discharge (Mann et al., 2022). Moreover, spring freshet is predicted to occur earlier (Stadnyk et al., 2021), water temperatures to increase (Blaen et al., 2013) and hydrological flow paths to change due to permafrost thaw (Ala-Aho et al., 2018; Tank et al., 2020). All these processes may change fluvial carbon dynamics, thus investigating response of fluvial systems to changing conditions is important to be able to better comprehend their future state.

1.5 Research objectives

This thesis focuses on carbon dynamics in Arctic fluvial systems with a particular focus on POC, including its composition, degradation, and seasonality. The aim is to understand how composition of POC might affect its lability and analyse how seasonality affects fluvial carbon dynamics. Observational data generated for this thesis, improves our understanding of abrupt permafrost thaw, OC mineralization in fluvial systems and potential climate impact of POC. These data may help us to comprehend composition of POC in Arctic fluvial systems, its susceptibility to degradation and furthermore, improve global climate models by providing degradation rates for POC in the Kolyma River, one of the major Arctic rivers. Additionally, this thesis provides biogeochemical data and sheds light on fluvial carbon dynamics in lower order streams within the Kolyma watershed – a region which remains understudied (Virkkala et al., 2019). The research objectives for each chapter were as follows:

- Establish the potential impact of RTS features to climate warming by characterizing composition of the thawed particulate organic matter on a downstream transect from RTS features on the Peel Plateau and determine degradation status of this material. (chapter 2)
- Quantify seasonal degradation rates for POC in the Kolyma River and investigate POC-DOC interactions. (chapter 3)

- Investigate seasonal carbon dynamics in lower order streams within the Kolyma River watershed and characterize their carbon composition and sources with focus on POC. (chapter 4)

1.6 Thesis outline

This thesis consists of five chapters. This first chapter is an introduction to the thesis topics and the fifth chapter is a concluding chapter, which includes an outlook for the future.

Chapter 2: This chapter focuses on abrupt permafrost thaw from RTS features on the Peel Plateau, Canada. The increase in precipitation and air temperatures have increased the number of RTS features across the Canadian Arctic in the past decades. These features release dominantly POC to the fluvial system, however, degradation of POC during downstream transport has been poorly characterized. In this study, we use carbon isotopes (δ^{13}C, Δ^{14}C), lipid biomarkers (*n*-alkanes, *n*-alkanoic acids) and pyrolysis - gas chromatography - mass spectrometry to study composition and degradation status of POC across a 12 km downstream transect of two RTS features on the Peel Plateau.

Chapter 3: In this chapter, we use incubation experiments to study microbial degradation of POC in the Kolyma River, Siberia. We establish degradation rates for both POC and DOC based on carbon losses during incubations. To capture seasonal dynamics of degradation, we conducted the incubation during two distinct hydrological seasons: spring freshet and summer. Further, we investigate interactions between dissolved and particulate OC phases, and how they might affect OC degradation patterns. We use source apportionment modelling and carbon isotopes (δ^{13}C, Δ^{14}C) to characterize composition of degraded POC.

Chapter 4: This chapter links to the previous chapter and focuses on carbon and water chemistry dynamics of lower order streams within the Kolyma River watershed with a focus on POC. With this study, we wanted to shed light on seasonal carbon dynamics, drivers of carbon composition, spatial characteristics of smaller tributaries and compare these to the Kolyma River mainstem. We use spatial analysis to characterize landscape of these tributary watersheds, and additionally, water chemistry, carbon concentrations and carbon isotopes (δ^{13}C, Δ^{14}C) to describe these diverse fluvial systems.