



Organic matter chemistry controls greenhouse gas emissions from permafrost peatlands



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ABSTRACT

Large tracts of arctic and subarctic peatlands are underlain by permafrost. These peatlands store large quantities of carbon (C), and are currently under severe threat from climate change. The aim of this study was to determine the size and organic chemistry of the easily degradable C pool in permafrost peatlands and link the functional organic chemistry to temperature and moisture controls of greenhouse gas emissions. First, we used a combination of field measurements and laboratory experiments to assess the influence of increased temperature and flooding on CO₂ and CH₄ emissions from sixteen permafrost peatlands in subarctic Sweden and Canada. Second, we determined the variation in organic matter chemistry and the associated microbial community composition of the peat active layer, with depth using quantitative ¹³C solid-state NMR and molecular biomarkers respectively. We demonstrate that the peat organic chemistry strongly controls CO₂ release from peat and that ca. 35 and 26% of the peat organic matter, at the Swedish and Canadian peatlands sites, respectively, is easily degradable by heterotrophic microorganisms. In contrast to CO₂, CH₄ emissions were decoupled from peat functional organic chemistry. We show a strong relationship between the microbial community structure and the peat organic chemistry suggesting that substrate type and abundance is an important driver of microbial composition in sub-arctic peatlands. Despite considerable variation in peat chemistry and microbial community composition with depth the temperature sensitivity was comparable throughout the active layer. Our study shows that functional organic chemistry controls both soil respiration rates and the composition of the microbial community. Furthermore, if these peatlands collapse and flood on thawing, they are unlikely to become large emitters of CH₄ without additional input of labile substrates.

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1. Introduction

Subarctic peatlands rich in carbon (C) account for ca. 20% of permafrost area across the arctic and store ca. 94.3 Gt-C (Tarnocai et al., 2009; Schuur and Abbott, 2011). With current estimates of anthropogenic fossil fuel emissions at 11.8 Gt-C yr⁻¹ (Friedlingstein et al., 2014), this represents a substantial C reservoir at risk, with severe implication for future global climate (Schneider von Deimling et al., 2012). The arctic is predicted to undergo mean annual temperature increases of over 5 °C (IPCC, 2014) leading to estimated C losses of 232–380 GtC by 2111 from permafrost soils (Schuur and

Abbott, 2011). These high C loss rates is supported by incubation and modelling studies suggesting that within 50 years ca. 40% of the soil organic material (ca. 60 Gt-C) currently held in organic permafrost soils could be mineralised and released to the atmosphere (Schädel et al., 2014).

While it is well established that the extensive C stores in permafrost peatlands are especially susceptible to losses through a combination of expected climate warming (Dorrepaal et al., 2009; Wang et al., 2010; Harden et al., 2012) and high concentrations of labile constituents (i.e. easily degraded by microorganisms) (Dorrepaal et al., 2009; Schuur et al., 2009; Schuur and Abbott, 2011; Schädel et al., 2014), uncertainties remain about the functional composition of the permafrost peatland C pool (e.g. the proportion of alkyls, O-alkyls, aromatics in the peat matrix) and how this may control C losses in a warming arctic. Furthermore,

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permafrost thaw in this region will result in deeper active layers which may subside, flood and result in thermokarst formation as the ice rich core is lost (Osterkamp, 2007; Åkerman and Johansson, 2008). This increased water logging, with associated anoxic conditions, may increase CH₄ emissions and potentially lower heterotrophic CO₂ losses (Christensen et al., 2004; Schuur et al., 2008; Treat et al., 2014).

The susceptibility of peat to decomposition by microbes is linked to its organic chemistry. Peat chemistry has been shown to influence potential CO₂ and CH₄ emissions from subarctic, temperate and tropical peats in short term incubations with higher CO₂ and CH₄ production resulting from peat high in carbohydrates (O-alkyls) and proteins (White et al., 2002; Andresen and White, 2006; Reiche et al., 2010; Wright et al., 2011; Treat et al., 2014). However, anaerobic CH₄ production is both less efficient and more strongly limited by substrate quality (Ström et al., 2012) than aerobic CO₂ production.

One of the factors determining if a given exothermic reaction will occur is the activation energy (E_a) of the reaction (Atkin, 1994). For organic materials, such as peat and litter, the relationship between E_a and the structure of molecules are described by kinetic theory (e.g. Lloyd and Taylor, 1994; Craine et al., 2010): Kinetic theory postulates that decomposition of recalcitrant, structurally complex, organic compounds that have greater activation energies, puts higher energy demands on microorganisms. Recalcitrant organic compounds therefore have greater temperature sensitivity than more labile compounds with lower E_a (Fierer et al., 2005). In the context of permafrost peatlands, understanding the temperature response and E_a of peat decomposition in different peat layers provides information which can be used to assess peat lability and vulnerability to decomposition at higher temperatures. However, there exists a severe lack of data quantitatively linking functional organic chemistry of peat to its temperature sensitivity (but see Treat et al., 2014 who used a semi-quantitative pyrolysis-GCMS biomarker approach to link peat chemistry to temperature sensitivity). In the context of permafrost soils, the temperature sensitivity of the constituent organic material may regulate how climate warming affects C release to the atmosphere.

The microbial community, through use of carbon for respiration and growth, ultimately controls the release of stored carbon from organic soils, and its activity is dependent on environmental conditions such as temperature, hydrology and pH (Bergman et al., 1999; Yu et al., 2007) as well as the quality and quantity of resources as influenced by organic chemistry and the nutrient status of the soils (Myers et al., 2012; Basiliko et al., 2006). Greater fungal abundance in peat has been associated with more efficient microbial communities i.e. communities with low respiration rates relative to the microbial biomass and lower respiration quotients (qCO_2 , i.e. the respiration rate per unit biomass) (Basiliko et al., 2006), although others have found less clear cut depth effects (Myers et al., 2012). Fungi are limited to aerobic environments and lower O₂ levels in deeper peat layers are likely to inhibit fungal growth, with implications for degradation of more complex organic molecules (Freeman et al., 2004). For example, lignin degradation by lignolytic microorganisms (mainly fungi) require O₂ to efficiently depolymerize and solubilize lignin (Zeikus, 1981) and is thus likely to be hampered in deep and/or waterlogged peat layers.

To further our understanding of the fate of permafrost peatland carbon and greenhouse gas feedbacks under future climate change conditions, this study explored the overarching hypothesis that organic matter chemistry is the primary driver of decomposition in permafrost peatlands, through its influence on greenhouse gas production, the temperature sensitivity of decomposition processes, and microbial community composition in sub-arctic peatlands. The objectives of this study were therefore to determine the

peat functional chemistry, microbial community composition, and CO₂ and CH₄ release from permafrost peatlands under different moisture and temperature treatments. The study focused on the seasonally thawed active layer which stores ca. 500 Pg of C (mineral and peat soils combined) across the Pan arctic (Hugelius et al., 2014).

To achieve our objectives we tested the following specific hypothesis relating to the vulnerability of the peatland carbon store to environmental change:

1. *Ex situ* experimental flooding of permafrost plateau peat will result in a shift from net CH₄ uptake under mesic conditions to CH₄ efflux throughout the active layer.
2. Peat organic chemistry, as determined by quantitative ¹³C MAS NMR, are linked to CO₂ and CH₄ emissions rates from plateau peat with higher CO₂ and CH₄ emissions from peat with a greater proportion of labile peat.
3. Deeper and more degraded peat contains more recalcitrant organic matter (e.g. alkyls), have higher E_a and Q_{10} values, and its decomposition is hence more sensitive to increases in temperature than surface peat, provided that other limiting factors are controlled (e.g. optimal pH, moisture, and nutrient conditions).
4. The composition of the microbial decomposer community is driven, at least in part, by peat organic chemistry.

2. Materials and methods

2.1. Site description

Two study areas were investigated, the Torneträsk area, northern Sweden (68°12'N, 19°03'E, 351 m asl) and the Churchill area, north eastern Canada (58°44'N, 93°49'W, 25 m asl). These areas were chosen as they are currently undergoing permafrost thaw (Lawrence et al., 2008; Sannel and Kuhry, 2011; Åkerman and Johansson, 2008). The mean annual temperature (MAT) in the Torneträsk area ranged between 0.8 and 1.0 °C and the mean annual precipitation (MAP) ranged from 304 mm in the west to 424 mm in the east, in Churchill, MAT was −7 °C and MAP was 414 mm. Both areas support peatlands with permafrost cores, so called palsas. The initiation of peat formation in the Torneträsk area is ca. 800–900 (Kokfelt et al., 2009) and ca. 3500–5200 years BP in the Churchill area (Hugelius et al., 2010). Total peat depths, including the permanently frozen layer, range from 90 to 160 cm with a maximum active (i.e. seasonally thawed) layer depth of 95 cm (Kuhry, 2008; Åkerman and Johansson, 2008). The depth to the permafrost varies between wetter and drier areas with a shallower active layer in drier areas. The sites were characterised by areas of raised peat plateaus, supported by an ice-rich core, with relatively dry surface conditions (mesic), dominated by bryophytes, lichens and evergreen dwarf shrub (Supplementary information 1). At the Torneträsk sites *Sphagnum fuscum* was the dominant moss species while *Dicranum elongatum* contributed to ground cover to a large extent at the Churchill sites. The main evergreen shrubs species at both areas were *Empetrum nigrum* and *Ledum* sp. while *Betula nana* was the dominant deciduous shrub. Lichens were more abundant at the Churchill peatlands than in the Torneträsk area. The most common herbaceous species for both areas was *Rubus chamaemorus*. The Torneträsk sites showed signs of small scale permafrost thaw and areas of peat collapse, with collapsed areas ranging between tens to hundreds of metres across). In Churchill, thermokarst areas were actively forming adjacent to plateau areas. Marginal collapsed areas tended to be vegetated by graminoids mainly *Carex* and *Eriophorum* species. See Supplementary

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