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Permafrost thaw and implications for the fate and transport of tritium in the Canadian north

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ABSTRACT

Layers of permafrost developed during the 1950s and 1960s incorporated tritium from the atmosphere that originated from global nuclear weapons testing. In regions underlain by substantial permafrost, this tritium has been effectively trapped in ice since it was deposited and subject to radioactive decay alone, which has substantially lengthened its environmental half-life compared to areas with little or no permafrost where the weapons-test era precipitation has been subject to both decay and hydrodynamic dispersion. The Arctic is warming three times faster than other parts of the world, with northern regions incurring some of the most pronounced effects of climate change, resulting in permafrost degradation. A series of 23 waterbodies across the Canadian sub-Arctic spanning the continuous, discontinuous and isolated patches permafrost zones in northern Manitoba, Northwest Territories and Labrador were sampled. Surface water and groundwater seepage samples were collected from each lake and analyzed for tritium, stable isotopes ($\delta^{18}O$ and $\delta^{2}H$) and general water chemistry characteristics. Measured tritium was significantly higher in surface waters (SW) and groundwater seepage (GW) in water bodies located in the sporadic discontinuous (64 ± 15 TU. in SW and 52 ± 9 TU. in GW) and extensive discontinuous (53 ± 7 TU. in SW and 61 ± 7 TU. in GW) permafrost regions of the Northwest Territories than in regions underlain by continuous permafrost in northern Manitoba (< 12 TU. in both SW and GW) or those within isolated patches of permafrost in Labrador (16 ± 2 TU. in SW and 21 ± 4 TU. in GW). The greatest tritium enrichment (up to 128 TU.) was observed in lakes near Jean Marie River in the Mackenzie River valley, a region known to be experiencing extensive permafrost degradation. These results demonstrate significant permafrost degradation in the central Mackenzie River basin and show that tritium is becoming increasingly mobile in the sub-Arctic environment—at concentrations higher than expected—as a result of a warming climate. A better understanding of the cycling of tritium in the environment will improve our understanding of Arctic radioecology under changing environmental conditions.

1. Introduction

1.1. Permafrost degradation in response to climate change

Permafrost occupies about 50% of Canada's land area (Smith and Burgess, 2004) and plays an important role in many hydrological, geochemical and ecological processes in these regions. Permafrost, however, is particularly sensitive to climate change and a number of recent studies have shown that permafrost is warming and degrading, resulting in a deepening of the active layer (Åkerman and Johansson, 2008; Smith et al., 2009; Jorgenson et al., 2012; Quinton and Baltzer, 2013). Recent studies have identified a general increasing trend in the active layer thickness in many regions, including Russia (Anisimov and Reneva, 2006), Tibet (Wu et al., 2010), northern Europe (Åkerman and Johansson, 2008), Alaska (Jafarov et al., 2013) and the Canadian Arctic (Smith et al., 2009; Quinton and Baltzer, 2013). Widespread warming in permafrost regions will result in substantial changes in terrestrial hydrology (Woo et al., 2008), vegetation composition (Danby and Hik, 2007), landscape topography (Rowland et al., 2010), methane fluxes (Christensen et al., 2004), ecosystem function (Grosse et al., 2016) and fate and transport of contaminants (Klaminder et al., 2008, 2010; Rydberg et al., 2010; Schuster et al., 2018), including radionuclides (AMAP, 2011).

In the northern portion of the permafrost region, permafrost is spatially continuous and may be several hundred metres thick, typically persisting at ground temperatures less than −5 °C (Heginbottom et al., 1995). Farther south, permafrost conditions become discontinuous and patchy and the permafrost layer is only several metres thick, persisting

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at ground temperatures around −2 °C to 0 °C (Smith, 2011; Quinton and Baltzer, 2013). Such conditions make permafrost in these discontinuous zones sensitive to climate warming and many regions are experiencing extensive permafrost degradation (Payette et al., 2004; Camill, 2005; Smith, 2011). Camill (2005) found that permafrost thaw rates have accelerated significantly across the Canadian sporadic and discontinuous permafrost zones over the past 50 years, with thaw rates increasing by 200–300% since the 1940s. Overall, the area of near-surface permafrost in the northern hemisphere is projected to decline by 20% relative to today's area by 2040, and could retract by as much as 60% by 2080 under a scenario of high greenhouse gas emissions (AMAP, 2017).

1.2. Permafrost degradation and mobilization of weapons-era tritium

The extent to which thawing permafrost will change the fate and transport of contaminants in the Canadian north remains a significant gap in our knowledge of how ecosystems will respond to climate warming (Vonk et al., 2015). The long-range transport of contaminants has brought elevated concentrations of pesticides, metals and radionuclides to the Canadian north, which are then deposited into various compartments of the environment by precipitation and atmospheric deposition (Barrie et al., 1992). Permafrost can accumulate atmospherically-derived contaminants as they are washed out of the atmosphere during precipitation events and act as a long-term storage unit for contaminants and other solutes (Schindler and Smol, 2006; Grannas et al., 2013; Vonk et al., 2015). However, climate warming will likely disrupt this sequestration of contaminants in permafrost soils, and permafrost thaw may result in increased mobility of contaminants and other solutes from catchment soils to surface waters due to accelerated soil/peat erosion, increased hydrological connectivity and increased runoff leading to exposure of soluble contaminants (Dyke, 2001; Fortier et al., 2007; Klaminder et al., 2008, 2010; Rydberg et al., 2010; Gordon et al., 2016; Bouchard et al., 2017).

Most of the studies on the interactions between contaminants and permafrost thaw have focussed on petroleum products, persistent organic pollutants and metals such as mercury and lead (Vonk et al., 2015; Schuster et al., 2018). However, relatively little attention has been paid to the effects of permafrost thaw on the fate and transport of radionuclides such as tritium (^{3}H) in the northern environment. Tritium is a naturally occurring radioisotope of hydrogen with a half-life ($t_{1/2}$) of 12.32 years. Tritium is directly incorporated into the water molecule, making it highly mobile in the environment and a useful marker for hydrological studies (Clark and Fritz, 1997). Atmospheric nuclear weapons testing, which occurred from about 1945 to 1980 and peaked in the early 1960s, led to elevated tritium emissions in the atmosphere around the globe (Clark and Fritz, 1997). The northern hemisphere received the bulk of these tritium emissions, with the sub-Arctic and Arctic receiving the highest loads (Aarkrog, 1994, Fig. 1a). For example, tritium concentrations in precipitation peaked at ~10,000 tritium units (T.U.; ~1000 times background) in Whitehorse, Yukon, and reached 6960 T U. (~696 times background) in Fort Smith, Northwest Territories, both occurring in 1963 (IAEA and WMO, 2017). Fig. 1 shows a snapshot of June 1963 tritium fallout around the northern hemisphere and the distribution of different permafrost zones across the Canadian north. Since the 1960s, tritium concentrations in precipitation and most surface waters have declined exponentially and are now close to natural levels (Clark and Fritz, 1997; IAEA and WMO, 2017). This exponential decline in tritium concentrations in surface waters is far greater than that accounted for by radiological decay alone, owing to hydrodynamic dispersion and mixing with waters not enriched by weapons-era tritium. In many permafrost regions, however, it is anticipated that the influence of hydrodynamic dispersion on tritium concentration would be minimal because (*i*) precipitation is incorporated into permafrost relatively quickly (Woo, 2012) and (*ii*) in flat, peat– and bog– dominated permafrost landscapes, hydraulic

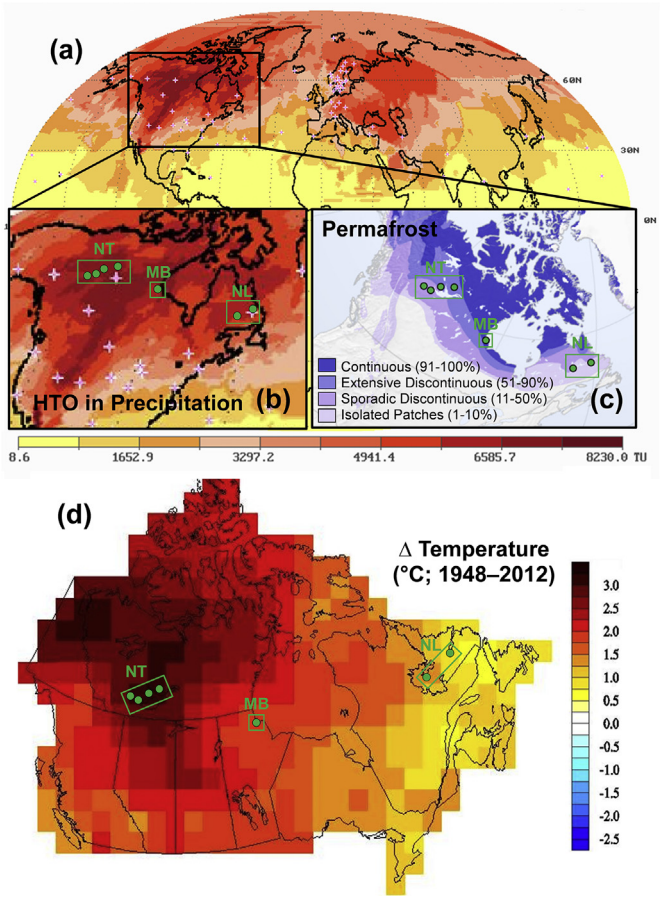

Fig. 1. (a) Tritium concentrations in precipitation in June 1963 at the global scale (from Aggarwal (2016) using the GNIP database (IAEA and WMO, 2017)); (b) tritium concentrations in North America in June 1963 (monthly mean); (c) permafrost classification across Canada (from NASA Earth Observatory Program (2016)); and (d) the change in mean annual air temperature in Canada from 1948 to 2012 (from the Government of Canada (2015)). Sample locations included in this study are shown as green points and HTO values are expressed as Tritium Units (T.U.). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

conductivity is low and waters typically have a long residence time within a watershed. In a region of discontinuous permafrost and peat plateaus in northern Alberta, Gibson et al. (2015) found that unfrozen wetlands contained tritium-enriched waters from 1960s precipitation, which were attributed to long water residence times in a low-hydraulic conductivity peat landscape.

Positioned between the active layer and the permafrost is an ice-rich and solute-rich layer called the transition zone (Shur et al., 2005; Kokelj and Burn, 2005). Using a stable isotope approach, Wang et al. (2018) found that active layer water sourced from precipitation was the dominant source of this ground ice near the permafrost table. The transition zone experiences freeze-thaw transitions on a sub-decadal to multi-centennial scale (Shur et al., 2005). The top of the transition zone is most susceptible to thaw when active layer thaw is deep enough to penetrate the zone (Shur et al., 2005). With deeper active layer thaw occurring in many permafrost regions due to rising mean annual air and ground temperatures, there is potential for these solutes, including tritium, to be released into surface waters. In regions with long water residence times, it could take many decades for this tritium-enriched water to leave the watershed and be dispersed (Gibson et al., 2015).

As described by Gibson et al. (2015), relatively old (i.e., pre-nuclear weapons test era) permafrost is expected to contain little or no tritium (i.e., < 10 T U.), whereas modern permafrost that formed from post-