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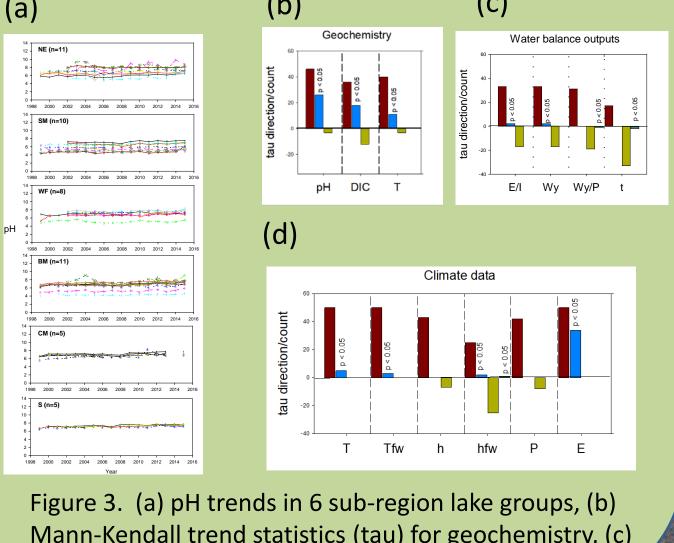
# Permafrost Thaw as a Mechanism for Widespread pH Increase in Boreal Lakes: Isotopic and **Geochemical Evidence**

# I.J.Gibson<sup>1,2</sup>

Geography, University of Victoria, Victoria BC, Canada 2 InnoTech Alberta, Victoria BC, Canada

**Introduction**: Stable isotopes and geochemistry were measured in 50 boreal lakes over the last two decades to study and monitor acid sensitivity in Alberta's Oil Sands region. The region is a wetland-dominated, boreal forest region with discontinuous, thawing permafrost (Panel 1). Unexpectedly, widespread pH increase has occurred in many of the study lakes (Panel 2). Using an isotope mass balance approach we applied watershed, climatic and isotopic data (oxygen-18, deuterium, carbon-13, tritium, radon-222) to investigate drivers of water balance and water chemistry in the lakes and their respective watersheds. Sitespecific differences in runoff to lakes was found to be driven by gradients in climate, lake/watershed configuration, substrate, as well as wetland type, and permafrost extent (Panel 3). Our hypothesis for pH increase is that DIC export has accompanied bog collapse in the watersheds leading to basification of the lakes (Panel 4). We propose a new permafrost thaw classification for identifying the stage of permafrost thaw at specific sites (Panel 5) and use it to provide initial evidence of linkage between pH trends, permafrost thaw stage, and thawdriven water yield (Panel 6).

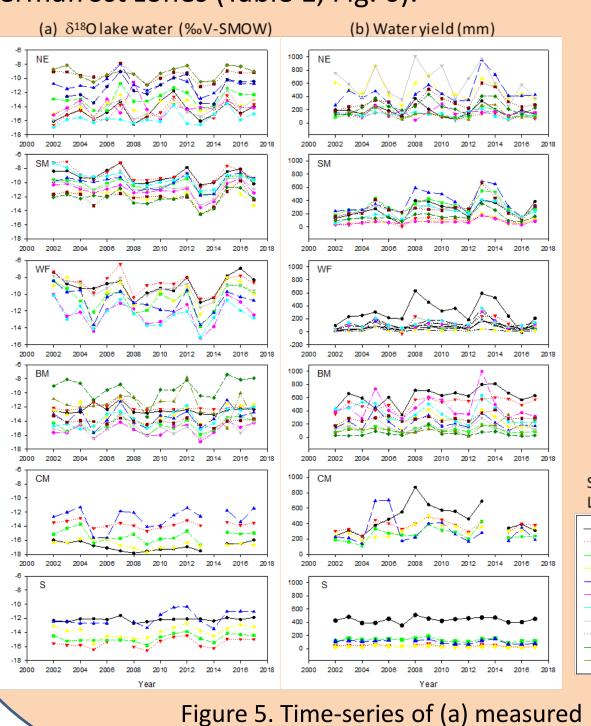
Landscape and hydrology are changing in the boreal forest of northeastern Alberta due to development and climate change impacts. We noted increasing trends in pH of 46 of 50 lakes monitored in the region over the past 2 decades (Fig. 3a&3b), as well as similar trends in dissolved inorganic carbon (DIC) and water temperature (T) (Fig. 3b). While climate drivers were generally found to be trending upward over time (Fig. 3d), water balance parameters were found to be trending non-monotonically (Fig. 3c). Further investigation of landcover and permafrost characteristics provided a hypothesis for why.



Mann-Kendall trend statistics (tau) for geochemistry, (c) water balance parameters and (d) climate drivers.

## **Panel 3**: Isotope Mass Balance (IMB)

Method: IMB was applied to monitoring data for 50 lakes including time series <sup>18</sup>O and <sup>2</sup>H and less frequent sampling for  ${}^{13}C_{DIC}$  and radon-222. Systematic enrichment in <sup>18</sup>O and <sup>2</sup>H is noted in lakes compared to input sources (Fig 4a & 4b) which is attributed to varying proportions of water loss by evaporation. Incorporating a Craig-Gordon model approach to characterize boundary layer processes, and using NARR reanalysis climate data, site-specific estimates of evaporation/inflow, residence time and water yield to the lakes are determined. Time-series <sup>18</sup>O and derived water yield illustrate non-monotonic interannual trends (Fig. 5). A sub-region summary is also provided highlighting different responses in various permafrost zones (Table 1, Fig. 6).



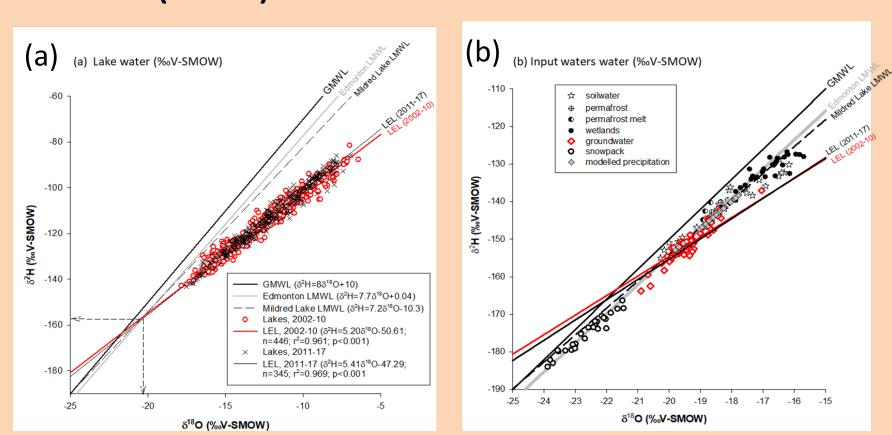


Figure 4. Crossplots showing <sup>18</sup>O and <sup>2</sup>H composition of (a) lakes in comparison with (b) input sources. Lakes are enriched along a local evaporation line (LEL) whereas input sources such as precipitation and groundwater fall close to two local meteoric water lines (LMWL) as defined by sampling at nearby climate stations, and the Global Meteoric Water Line (GMWL).

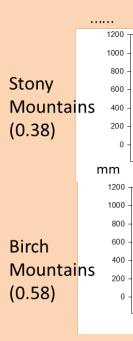
### Table 1. Quantitative IMB results summarized by lake sub-region, 2002-2017.

	11	(%/100)	(mm/yr)
Northeast Fort McMurray (NE)	11	<b>0.18</b> (0.04 to 0.46)	<b>270</b> (108 to 611)
Stony Mountains (SM)	10	<b>0.42</b> (0.24 to 0.64)	<b>215</b> (77 to 369)
West Fort McMurray (WF)	8	<b>0.50</b> (0.36 to 0.66)	112 (22 to 302)
Birch Mountains (BM)	11	<b>0.23</b> (0.10 to 0.48)	<b>287</b> (58 to 618)
Caribou Mountains (CM)	5	<b>0.18</b> (0.11 to 0.29)	343 (258 to 468)
Shield Lakes (S)	5	<b>0.19</b> (0.09 to 0.26)	<b>149</b> (32 to 435)
Low Permafrost (<2%)	31	<b>0.30</b> (0.08 to 0.54)	<b>166</b> (22 to 435)
Fen-dominated Permafrost (17-62%)	5	<b>0.24</b> (0.12 to 0.49)	159 (58 to 250)
Bog-dominated Permafrost* (2-88%)	14	<b>0.21</b> (0.08 to 0.43)	405 (137 to 618)

n= No. lakes; Values in brackets indicate range of annual modelled values for lakes. \*Thaw lakes of Gibson et al. (2015)

Lake No

Figure 6 (Right). Selected outputs highlighting the contrast between water yield and precipitation depth over the watershed for two contrasting subregions, one with minor permafrost (Stony Mountains) and one with active permafrost thaw (Birch Mountains).



<sup>18</sup>O in lakes, and (b) derived water yield.

### Panel 2: What's happening and why?

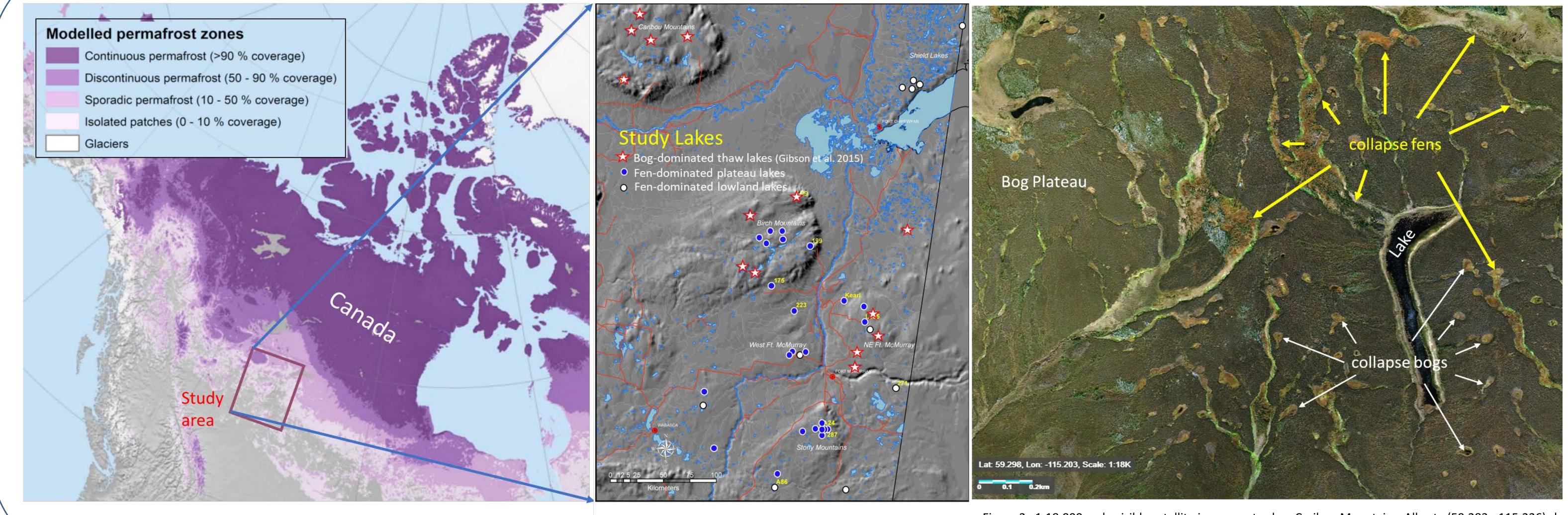


Figure 1. Maps showing (i) modelled permafrost zones (modified from Obu et al. 2019), and (ii) location of the study lakes classified according to three main wetland settings.

**Residence time 0.49** (0.20 to 1.10) **0.84** (0.31 to 1.67) **0.38** (0.13 to 0.65) **1.14** (0.49 to 1.96 **0.20** (0.04 to 0.55) **1.06** (0.61 to 1.77) **0.58** (0.11 to 1.20) **2.06** (0.22 to 10.81) **0.74** (0.54 to 0.92) **2.41** (1.12 to 3.74) **0.36** (0.08 to 1.05) **3.02** (1.17 to 7.80) **0.31** (0.04 to 1.05) **1.36** (0.31 to 7.80) **0.33** (0.11 to 0.53) **0.56** (0.37 to 0.78) **0.80** (0.25 to 1.20) **2.44** (0.22 to 10.81)

<sup>È</sup>≜╤╶╴<sup>╤</sup>╋╄╶╤<sub>┻</sub>┥┥┥╸ 2002 2004 2006 2008 2010 2012 2014 2016 2018 **Year** 

**<u>Results</u>**: Hydrology of lakes is found to be governed mainly by land cover characteristics and to a lesser extent by climate, as shown in a PCA biplot (Fig.7). Bog-dominated plateaus with collapse scars account for the majority of permafrost occurrences. On average, lakes in these settings are found to have 300+ mm/year higher runoff due to contributions from permafrost thaw (see bogdominated permafrost, Table 1). Water yield in bogdominated systems is found to respond systematically along a thaw trajectory (Fig. 8).

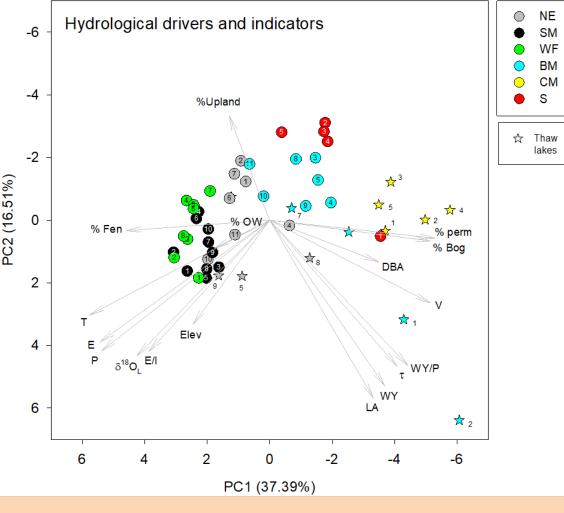


Figure 7. PCA showing hydrologic controls by sub-region.

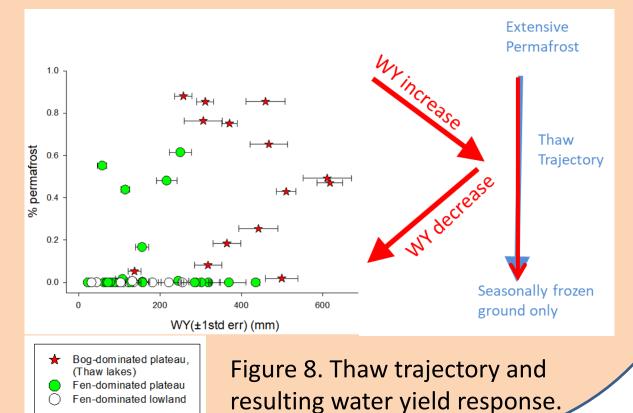
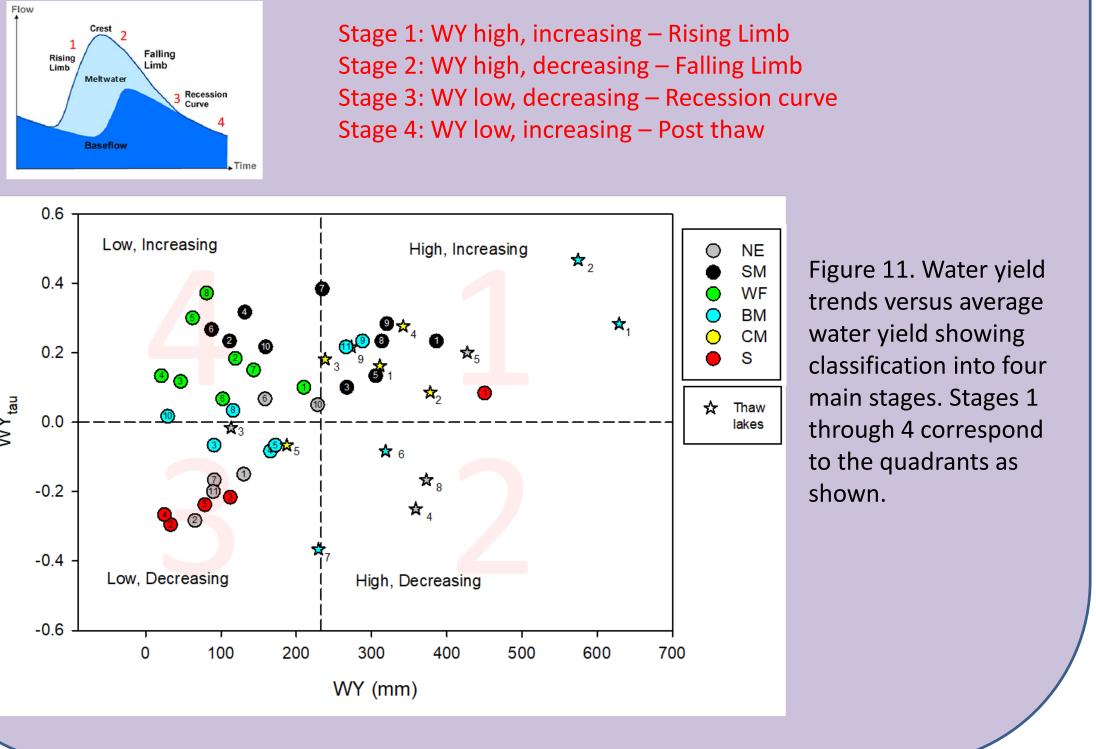


Figure 9. Photos of typical wetlands: (a) open fen – treed bog - lake, (b) drunken forest adjacent to collapse scar, and (c) frozen peat core, Birch Mountains, (f) schematic showing water fluxes in ombrotrophic bogs compared to miner trophic fens, (g) collapse bogs versus collapse fens (modified from Hayashi et al. 2004), (h) wetland moisture and geochemical gradients showing expected changes along a permafrost thaw trajectory (modified from Vitt et al. 1996)

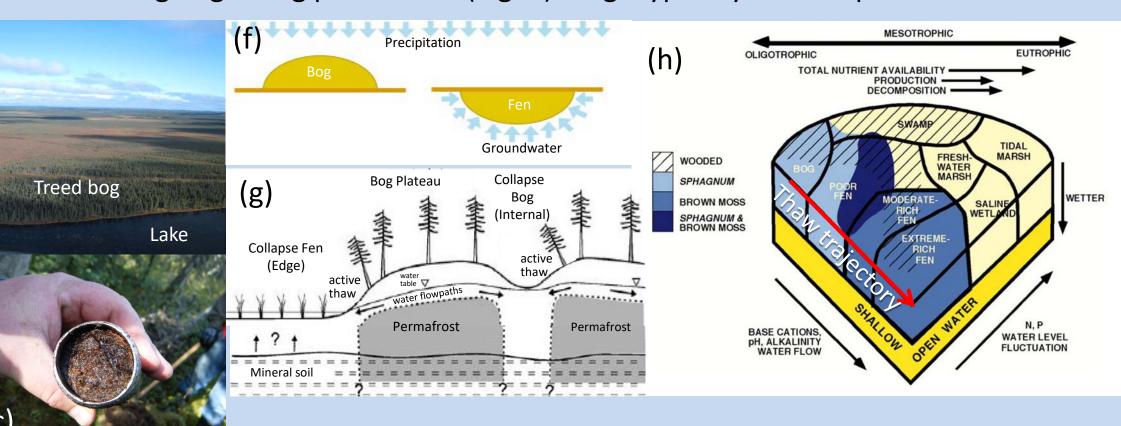
We propose a new site-specific classification that allows for identification of the stage of melt and offers a framework for predicting future hydrologic changes. Four stages of melt can be distinguished, assessed based on water yield results and Mann-Kendall tau statistics (Fig. 11). Conceptually, runoff changes can be viewed as a response to a permafrost thaw pulse or event hydrograph.



**Panel 1**: Study region and evidence of landscape changes due to permafrost thaw

# **Panel 4**: Proposed Mechanism for pH Increase

Peat-forming wetlands in the region have accumulated to depths of up to 10 m, forming a veneer blanketing drift deposits and bedrock. Precipitation-fed (ombrotrophic) bogs and groundwater-fed (minerotrophic) fens, account for greater than 50% of land cover in most watersheds, with several common variations as well as collapse bogs and fen types indicating degrading permafrost (Fig. 9). Bogs typically contain permafrost whereas fens do not.



Bogs and fens are known to sequester carbon and other elements, creating steep depth-wise gradients in chemical constituents (Fig. 10). They are also an important link in the biogeochemical cycle of lake-|<u>∔</u> ━ <sup>≠</sup> watershed systems. We postulate that basification of lakes has occurred due to hydrologic and geochemical changes accompanying collapse of bog plateaus in lake drainage areas, causing bogs to transform hydrologi 🛖 🜩 🌩 ically and geochemically into fens, thus altering delivery of dissolved solids. Fundamental changes expected along thaw trajectories include increased water table relative to ground surface, increased groundwater interaction, and enhanced runoff. Geochemical changes include mobilization of total dissolved solids (TDS), base cations (e.g. Ca2+), and DIC WTSMDWTSMDSM which are conveyed to lakes via recently thawed flowpaths. Note also Figure 10. Depth-wise shifts in selected parameters for water table (WT) and that increases in redox potential (eH) are also expected to lead to shallow to deep piezometers (S,M,D) oxidation and mobilization of metals and nutrients. at typical sites.

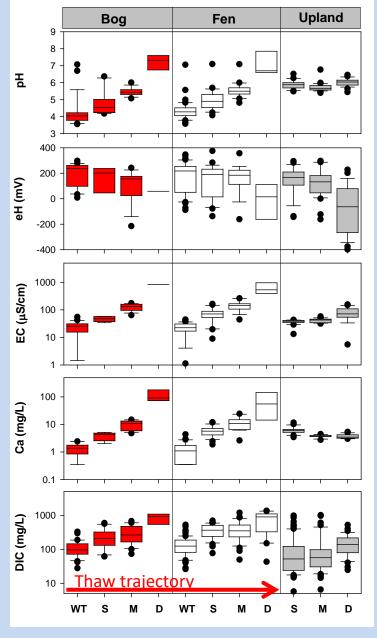
### **Panel 5**: Permafrost Thaw Classification

permafrost thaw stage (Fig. 12). 🖢 Bog-dominated plateau, Thaw lakes) Fen-dominated plateau Fen-dominated lowland 0.2 -0.4 --0.2 0.0 -0.4 ★ Bog-dominated plateau, 600 -Fen-dominated plateau 500 Fen-dominated lowland **3**00 -0.0

Figure 12. (a) DIC vs. pH tau , (b) water yield vs. pH tau. Lakes are labelled by watershed type and thaw stage (1 to 4).

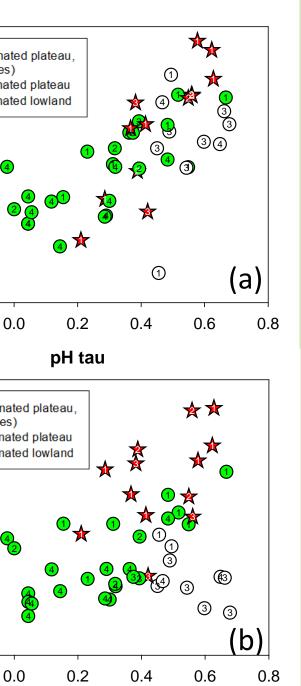


Figure 2. 1:18,000 scale visible satellite image centred on Caribou Mountains, Alberta (59.283, -115.226) showing characteristic examples of permafrost degradation features (collapse bogs and fens) situated near typical lakes.



### **Panel 6**: pH Response to Permafrost Thaw

Trends in DIC and pH are found to be mutually related (Fig 12a), as are trends in water yield and pH (Fig. 12b), although the latter shows greater variation in response depending on watershed type and



Conclusions: While parallel shifts in DIC and pH provide clear evidence of an underlying carbonate equilibria mechanism, linkage of pH changes to catchment water yield also establishes probable cause that permafrost thaw has affected this process through mobilization of DIC from collapse fens to lakes. Different responses are noted for sites on the plateaus compared to lowlands. While both bog-dominated and fen-dominated plateau lakes show increasing pH trends with increasing water yield, lowlands appear to show the opposite effect. As most lowlands are in the receding or post-thaw stages of permafrost degradation (i.e. stages 3 and 4), we postulate that the proposed basification mechanism may continue after the meltwater pulse is complete due to increase in fen/bog ratios in the watershed and associated establishment of greater subsurface connectivity and minerotrophic properties of post thaw drainage areas. **Future studies**: While water sampling was paused during 2018 and 2019 during a government program review, sampl-ing is expected to resume in 2020. Recommended studies include field-based assessment of permafrost conditions and more detailed characterization of isotopic and geochemical evolution of water from precipitation to discharge. References: Gibson et al. 2015. Hydrol. Process. 29, 3848–3861. Hayashi, M. et al., 2004. J. Hydrol. 296, 81-97.

Obu et al. 2019. The Cryosphere Discuss., doi: 10.5194/tc-2019-148. Vitt et al. 2016. Peatland inventory of Alberta, Phase 1. University of Alberta, Edmonton.