Environmental Science & Technology LETTERS
pubs.acs.org/journal/estlcu
Letter

Glacial Erosion Drives High Summer Mercury Exports from the Yukon River, Canada

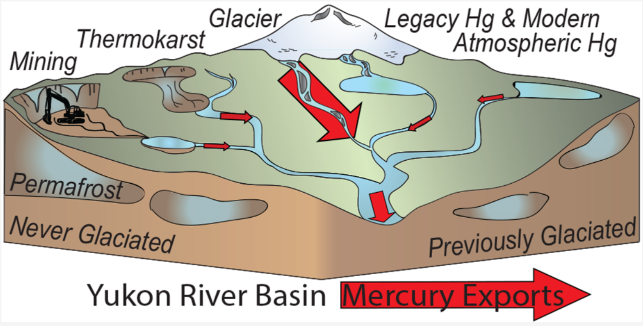
Published as part of the Environmental Science & Technology Letters virtual special issue "Accelerating Environmental Research to Achieve Sustainable Development Goals".

Kasia J. Staniszewska,* Alberto V. Reyes, and Colin A. Cooke

Cite This: *Environ. Sci. Technol. Lett.* 2023, 10, 1117−1124

Read Online

ACCESS | Metrics & More | Article Recommendations | Supporting Information



ABSTRACT: Mercury concentrations and yields in the Yukon River are the highest of the world's six largest panarctic drainages. Permafrost thaw has been implicated as the main driver of these high values. Alternative sources include mercury released from glacial melt and erosion, atmospheric mercury pollution, or surface mining. To determine the summer source and speciation of mercury across the Yukon River basin within Canada, we sampled water from 12 tributaries and the mainstem during July 2021. The total (unfiltered) mercury concentration in the glacier-fed White River was 57 ng/L, >10 times higher than all other sampled tributaries. The White River's high total mercury concentrations were driven by suspended sediment and persisted ~300 km downstream of glacierized headwaters. Total mercury concentrations were lowest (typically <2 ng/L) in tributaries downstream of still-water landscape features (e.g., lakes and settling ponds), suggesting these features are effective sinks for sediment-bound mercury. Low total mercury concentrations (~2 ng/L) were also observed in five tributaries across diverse thawing permafrost landscapes. These results suggest that glacial erosion and meltwater transport, not permafrost, drive enhanced exports of mercury with suspended sediment. Mercury exports may decline as glacial watersheds pass peak water. Other factors, including mercury released from permafrost thaw, are minor components at present.

KEYWORDS: *sediment, permafrost, glacier, methylmercury, climate change, geomorphology, physiography, hydrochemistry*

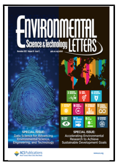
INTRODUCTION

Mercury (Hg) is a global contaminant that can convert to the neurotoxin methylmercury (MeHg) in aquatic settings and biomagnify through food webs, posing a risk to communities that rely on country foods.[1,2] There is particular concern about mercury contamination in the arctic and subarctic, where snow and glacier melt,[3−8] glacier erosion,[7,9] and permafrost thaw[4,10−14] have been implicated as important sources of mercury to rivers.

The Yukon River has the highest annual mercury yield of all large Arctic rivers.[13,15] Uniquely, more than half of its annual Hg export occurs during the summer (July−October) instead of the spring melt (May−June).[13,15] In the summer, Yukon River mercury yields are ~2.4 g/km^2, while the other five large Arctic rivers yield <1.2 g/km^2.[13,15] Thaw of organic-rich permafrost with high mercury stocks has been implicated as the main source of Yukon River mercury,[10,13] and in other Arctic fluvial systems.[11] Modeling results indicate that Yukon River fish may exceed consumption guidelines by 2050 as permafrost thaw increases mercury and organic matter exports.[14] Other sources for the high Yukon River mercury have been proposed, including placer gold mines that may be contaminated with mercury,[16] the release of mercury from glaciers that archive legacy mercury pollution,[5,8,13,16,17] atmospheric mercury pollution derived from coal combustion in East Asia,[2,5,13] glacial erosion and suspended sediment transport in currently glaciated tributaries,[8,9,13,18] or mixed anthropogenic contributions.[19]

Here we show that glacial erosion—not permafrost thaw—is the single most important source of mercury to the Yukon River during the summer. These results challenge the current understanding of how the Yukon River's mercury export may respond to climate change.[14] The results imply a low potential for mercury methylation and biotic uptake and limited delivery of mercury by the Yukon River to Arctic Ocean food webs and country foods.[20,21]

Received: June 23, 2023
Revised: September 29, 2023
Accepted: October 2, 2023
Published: October 9, 2023



ACS Publications

https://doi.org/10.1021/acs.estlett.3c00427

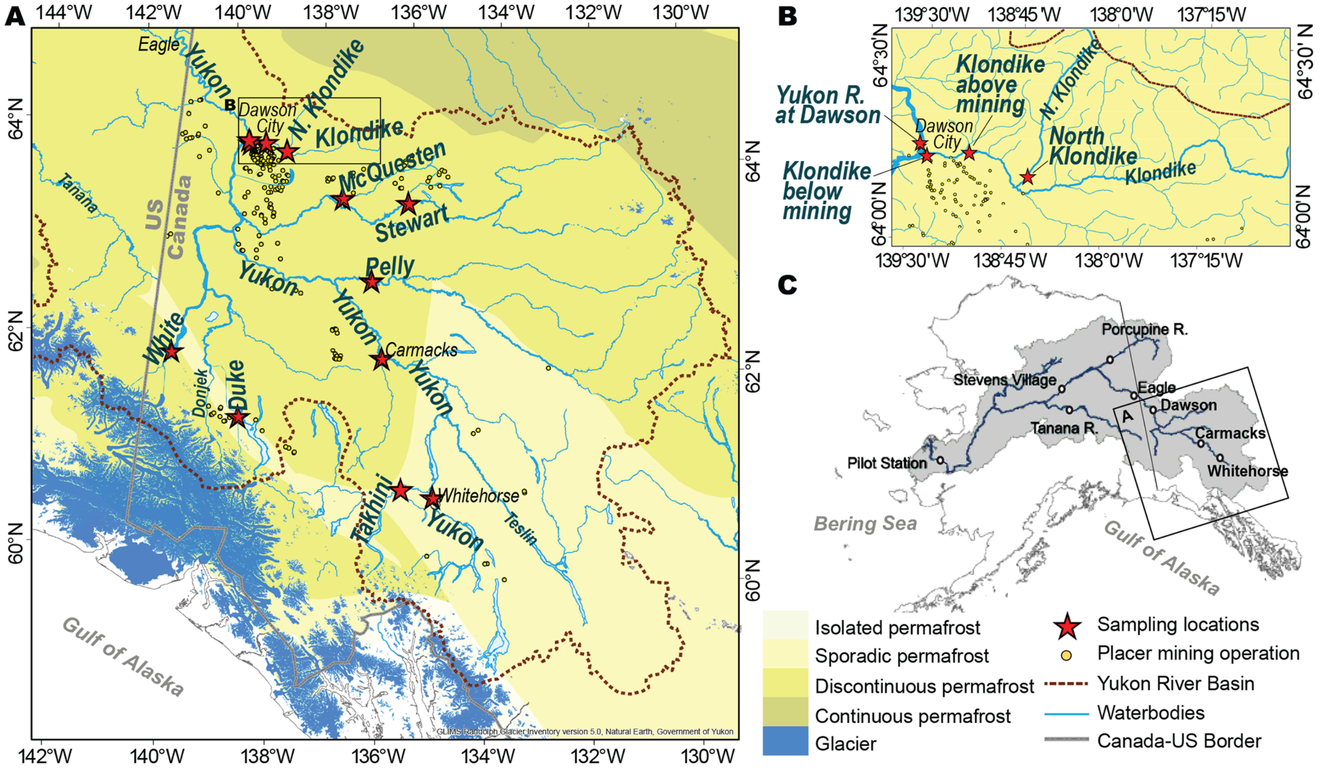


Figure 1. (A) Map of 2021 Yukon River basin sampling locations (red stars), 2015−2017 placer mining operations (yellow circles), the distribution of glaciers in southwest Yukon (dark blue polygon), and regional permafrost distribution (yellow polygons).[26] (B) Inset of the Klondike region, showing sampling site locations relative to extensive placer mining activity. (C) Yukon River basin within Yukon and Alaska, location of inset A (rectangle). Modified from ref 27.

■ STUDY REGION

The Yukon River basin (YRB; ∼833 000 km^2) drains regions of isolated through continuous permafrost, glaciated headwaters,[22] lake-source headwaters, and has an extensive footprint of active and historic surface mining (Figure 1).[23−25] The YRB has the largest area of glacier cover of all of the large panarctic rivers. We sampled 12 gauged and physiographically distinct sub-basins of the YRB within Canada with variable contributions from lakes, mining, glacier melt, and pristine and disturbed permafrost (Table S.1).

■ METHODS

Field collection and laboratory analyses for total and dissolved mercury, methylmercury, organic carbon, inorganic carbon, total suspended sediment, total dissolved solids, anions (SO_4 and Cl), turbidity, conductivity, pH, and alkalinity followed U.S. Environmental Protection Agency methodologies (see the Supporting Information).[28−32] River water grab samples were collected from a wadable distance from shore, >10 cm below the surface at 12 sites across the YRB from July 19−23, 2021 (Figures 1 and 2) following Method 1669.[28] Sampling sites were within 1 km of Water Survey of Canada gauge stations (Table S.1). Unfiltered phases are termed total, which includes particulate, colloidal, and ionic species; filtered phases are termed dissolved, which includes any colloid or ion with a diameter of <0.45 μm. Total samples were preserved during collection; dissolved samples were filtered and preserved within 48 h. Mercury was analyzed by oxidation, purge and trap, and cold vapor atomic fluorescence spectrometry (CVAFS) on a Tekran 2600 Mercury Analyzer using Method 1631E.[29] Methylmercury was analyzed by distillation, aqueous ethylation, purge and trap, isotope dilution, and CVAFS on a Tekran 2700 Methylmercury Analyzer coupled to an ICP-MS using Method 1630.[30] Field blanks were collected at three sites, and duplicates at two sites (Table S.2). All field blanks were at or near detection limits. Duplicates had low relative percent differences (<16%) with the exception of the total and dissolved organic carbon (48% and 40%, respectively) duplicate on White River. Concentrations (nanograms per liter) were multiplied by the mean daily discharge (cubic meters per second) to obtain the flux (mass per day) and divided by the drainage area to calculate the yield (grams per square kilometer per day).

■ RESULTS

Water Quality across the Yukon River Basin. July 2021 sampling occurred following a late-June to mid-July heat wave over western North America.[33] However, discharge was typical in most sites, except in the Yukon River at Whitehorse, where it reached a historical maximum (Figure S.1). Discharge in the glacierized White and Duke rivers and the lake-sourced Takhini River was near the upper quartile of historical values. Elsewhere, in the Klondike, Pelly, Stewart, McQuesten rivers, discharge was near the lower quartile of historical values.[33] Discharge at the Yukon River at Dawson City, the furthest downstream site, was near the historical late-July median.

The Yukon River and its tributaries were characterized by a wide range of water quality conditions (Table S.3). Total suspended sediment (TSS) concentrations ranged from 1.2 mg/L in the mining-impacted McQuesten River to 4206 mg/L (duplicate of 4012−4400 mg/L) in the glacial-fed White River

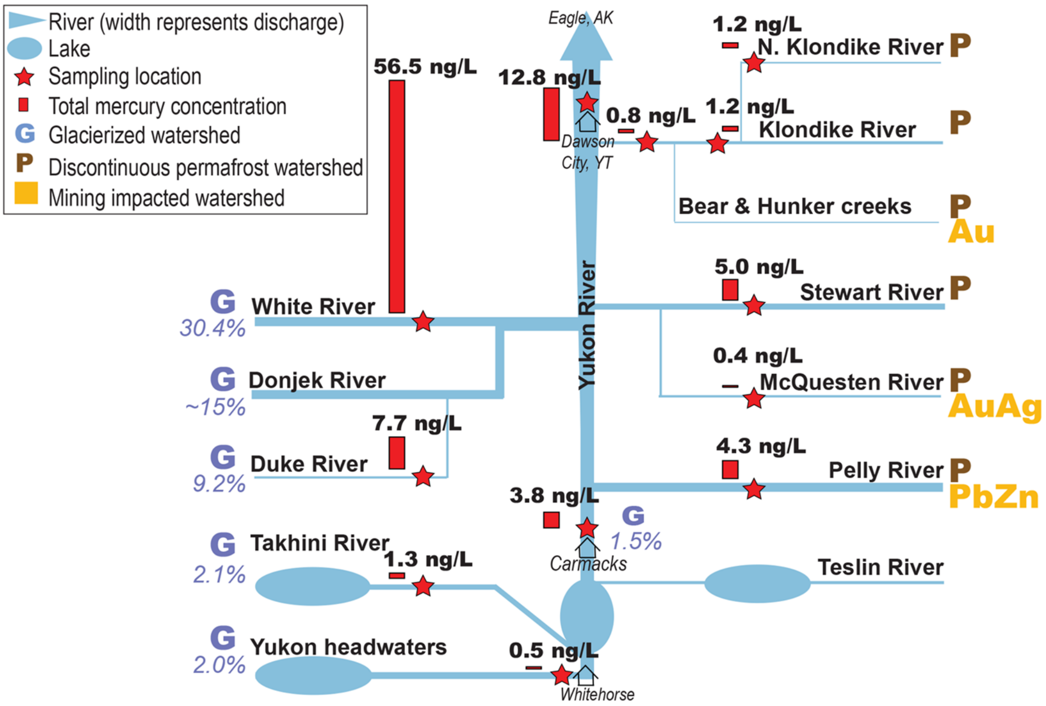


Figure 2. Schematic diagram of Yukon River water flow paths (line width representing July discharge; ovals represent lakes along the river flow path), with mercury concentrations (red bars, bold text) next to sampling locations (red stars). The proportion of glacier cover[22,24] in a basin at the sampling location is indicated by percentage values beneath G (glacierized watersheds). Discontinuous permafrost is indicated by "P",[26] and mining[23] influence is indicated by yellow text for the ore deposit.

(Figure 3). We note that grab sampling for TSS, compared to more complex field sampling techniques, likely results in underestimates of suspended sediment concentration.[34] Total organic carbon (TOC) concentrations were low and ranged from 1.5 mg/L in the Yukon River at Whitehorse to 7.1 mg/L (duplicate of 5.4−8.8 mg/L) in the White River. Organic carbon was predominantly particulate at the glacier-fed White (97%) and Duke (89%) rivers. Dissolved organic carbon comprised 45−70% of TOC at all other tributaries.

Total mercury (THg) concentrations (Figures 2 and 3) were highest in the glacier-fed White River (duplicate of 52.3−60.7 ng/L) and moderate in the Duke River (7.7 ng/L) and ∼300 km downstream of the White River at Yukon River at Dawson (duplicate of 12.7−12.9 ng/L). Total mercury concentrations were lower in rivers in pristine sporadic to discontinuous permafrost: North Klondike (1.2 ng/L), Klondike above mining (1.2 ng/L), Yukon at Carmacks (3.8 ng/L), Pelly (4.3 ng/L), and Stewart (5.0 ng/L) rivers. Total mercury concentrations were lowest in rivers flowing from large lakes in deep basins (Yukon River at Whitehorse and Takhini River; 0.5 and 1.3 ng/L, respectively) and downstream of active placer mining (McQuesten and Klondike rivers below mining; 0.4 and 0.8 ng/L, respectively).

Dissolved mercury (DHg) concentrations were broadly similar among tributaries (Figure 3), despite different catchment characteristics, ranging from 0.09 ng/L at Duke River to 0.80 ng/L at Klondike River above mining. The DHg/THg ratio was <10% at White, Duke, Yukon at Dawson, and Stewart rivers and 53−69% at North Klondike, Klondike above mining, and McQuesten and Klondike rivers below mining.

Total methylmercury [TMeHg (Figure 3)] concentrations were highest at the White River (duplicate of 0.090−0.094 ng/L), followed by the Yukon River at Dawson City (0.040−0.044 ng/L), the Yukon River at Carmacks (0.038 ng/L), and the Klondike River above mining (0.034 ng/L). TMeHg/THg ratios were <1% (e.g., 0.16% at the White River) at all tributaries except the Yukon River at Carmacks (1.0%), the Yukon River at Whitehorse (2.2%), and the Klondike River above (2.9%) and below mining (3.5%).

Dissolved methylmercury (DMeHg) concentrations (Table S.3) were only ∼2 times the detection limit, between 0.017 and 0.019 ng/L, at the Klondike River above and below mining, Pelly, and Stewart rivers and at or below the method detection limit (0.010 ng/L) at the Yukon River at Dawson City, the Yukon River at Whitehorse, and the White, Duke, Takhini, Stewart, McQuesten, and North Klondike rivers.

Suspended sediment and organic carbon have both been implicated as important drivers of mercury cycling in rivers.[13,15] THg concentrations correlated most strongly with TSS (Pearson $r > 0.90$); this correlation was biased by the White River high TSS and THg outlier, but the correlation was still strong ($r = 0.84$) if the White River outlier was excluded. The THg concentration also correlated strongly with TOC ($r = 0.87$), but the correlation was weaker if the White River was excluded ($r = 0.41$). DHg correlated most strongly with DOC ($r = 0.66$). Concentrations of total and dissolved MeHg correlated most strongly with TSS ($r = 0.93$ and -0.98, respectively). Discharge was a poor predictor for THg, DHg, and TMeHg concentrations ($r < 0.14$) yet was strongly negatively correlated with DMeHg ($r = -0.97$).

Mercury Species Daily Fluxes and Yields. Daily THg yields (milligrams per square kilometer per day) were highest in the White River (duplicate of 444−516 mg km^{-2} day^{-1}), in the Duke River (31.7 mg km^{-2} day^{-1}), and at the furthest downstream site, the Yukon River at Dawson (duplicate of 19.9−20.1 mg km^{-2} day^{-1}) (Figure 3). Other tributaries had low

THg yields of <5 mg km^{-2} day^{-1}. The White River contributed >50% of the THg flux to the Yukon River at Dawson (duplicate of 2.8−3.2 kg/day of 5.3−5.3 kg/day); the THg flux of other sites was <0.3 kg/day. Likewise, the White River had the highest DHg and TMeHg yields (1.4−1.4 mg km^{-2} day^{-1} and 782−799 μg km^{-2} day^{-1}, respectively); the remaining rivers had DHg and TMeHg yields of 0.2−0.7 mg km^{-2} day^{-1} and <100 μg km^{-2} day^{-1}, respectively. The highest fluxes of dissolved and methylated mercury were exported by catchments with large drainage basins (Yukon River at Carmacks, Pelly, Stewart, and White).

■ DISCUSSION

Sediment Erosion and Transport from Glacierized Catchments. Our results implicate the White River as the most important summer source of mercury to the Yukon River within Canada. The White River transports high concentrations of suspended sediment, supplied predominantly through glacial erosion and downstream remobilization of glaciofluvial sediment. The White River TSS concentration on the day of sampling (∼4200 mg/L) was on the low end of historical summer values [∼750−12000 mg/L (Figure S.2)].[35] This sediment is largely inorganic (TOC/TSS < 1%), and the proportion of MeHg was low (TMeHg/THg, 0.16%). Mercury methylation and biomagnification are strongly affected by complexation, redox conditions, pH, salinity, temperature,[36] dissolved organic carbon concentrations, and trophic structure.[37] Mercury in inorganic sediment has a lower potential to methylate[36] and subsequently enter food webs except by benthic organisms that ingest sediment.

The White River exported 2.9 kg of THg/day in late July, accounting for more than half of the THg export from the Yukon River at Dawson (5.3 kg/day) (Figure 3). This is despite the White River catchment accounting for <3% of the Yukon River basin (at Dawson) watershed. Moreover, our sampling did not incorporate the ungauged Donjek River, which is similar to the White River with respect to basin area, water yield, and glacial cover.[38] Future work in this region will benefit from a detailed characterization of mercury export and hydrology in other large glacierized tributaries.

Suspended sediment and particulate THg export from the heavily glacierized tributary rivers persisted 300 km downstream to the Yukon River at Dawson (12 ng/L). Below the Yukon River at Dawson, some portion of this particulate THg export is likely deposited in the ∼28 000 km^2 Yukon Flats in central Alaska (Figure 1),[39] where the sediment budget is likely balanced on centennial/millennial time scales,[40] and in the Yukon River delta and the shallow Bering Strait.[13,41] Methylation, and delivery of this pool of particulate mercury to the Arctic Ocean by Pacific currents and the transpolar drift, is likely limited.[16]

The Yukon River is the most extensively glacierized of the large panarctic rivers. Glaciers in Alaska experienced the largest mass loss of glaciers worldwide at the beginning of the 21st century,[42] and glacierized rivers draining to the Gulf of Alaska have high particulate mercury yields.[43,44] High mercury concentrations in other glacierized Arctic basins have been attributed to snow, ice, and geogenic sources.[5,7−9,45] Together, our results suggest that subglacial and proglacial sediment erosion-driven export of mercury from large glacierized tributaries has been an underappreciated component of riverine mercury export. However, mercury exports may decline before the end of the 21st century due to extensive deglaciation[42] and, later, depletion of easily eroded sediment stores out of deglaciated catchments.[46]

Legacy Contaminants in Snow and Ice. Snow and ice melt in the Takhini River headwaters, where snowpack mercury concentrations reach 5.9 ng/L,[5] are a key component of atmospheric mercury delivered to that tributary system. Mercury concentrations also reach 5 pg/g in nearby Mount Logan firn.[17] Mass balance considerations suggest that contributions from legacy atmospheric mercury in ice and snow (5−6 ng/L) do not drive the much higher mercury concentrations we observed in the nearby White River (57 ng/L). Rather, the White River's high concentration of TSS [∼4200 mg/L in July 2021, ∼5800 mg/L as the 1975−1992 July mean (Figure S.2)] with sediment mercury concentrations approximating that of the bulk continental crust (∼56 ng/g)[47] likely accounts for the majority of mercury exports in this heavily glaciated catchment.

Influence of Reservoirs on Mercury Exports. Mercury concentrations and yields were especially low in tributaries with reservoirs, lakes, wetlands, or settling ponds along their flow path, irrespective of the water yield. Such still-water environments act as sediment traps, reducing downstream particulate mercury export. Land-use management practices in the YRB[48,49] include the use of settling ponds downstream of placer mines, many of which disturb organic carbon-rich permafrost exposures.[50] Remarkably, despite being potential methylation hot spots,[51,52] methylmercury concentrations were extremely low (<0.03 ng/L) downstream of the lakes, wetlands, and mining districts examined during this study. Nonetheless, sediment exports are periodically elevated downstream of Yukon placer mines.[53] Dry antecedent conditions may have limited particulate mercury mobilization; follow-up work using continuous turbidity monitoring equipment may be an effective monitoring strategy.[54]

Land Disturbance and Thermokarst Influences on Mercury Exports. Organic-rich permafrost in the YRB contains stocks of legacy mercury;[10,55] thermokarst and land disturbance can mobilize this mercury downstream.[11] The positive DHg−DOC correlation reflects an organic source for this minor (∼3%) component of summer mercury exported from the Yukon River at Dawson. It has been suggested that total mercury exports from the Yukon River basin may increase by ≤200% by 2200 as permafrost degradation mobilizes organic matter-bound mercury in the basin.[14] However, organic-rich streams and rivers in permafrost regions of the Yukon River basin have modern ^{14}C-DOC values; this suggests that present-day permafrost thaw is mobilizing little old carbon (and mercury).[56] Additionally, mercury concentrations are very low—despite the presence of pervasive thermokarst—in the Old Crow River basin, which is underlain by continuous permafrost and drains into the Porcupine and Yukon rivers (Figure 1C).[57]

Understanding the present drivers of mercury export in the Yukon River is crucial to modeling the Yukon River basin's response to climate change. Decreasing contributions from glacierized tributaries may be balanced in the future by increasing contributions from thermokarst. The frequency of thaw slumps and active layer detachment slides is increasing across the panarctic, particularly in previously glaciated regions.[58−60] However, the downstream effect on water quality is modulated by factors such as watershed scale,[61] ground-ice content, slope, talik connectivity, and surficial materials.[62−64] Careful consideration of these physiographic factors, and

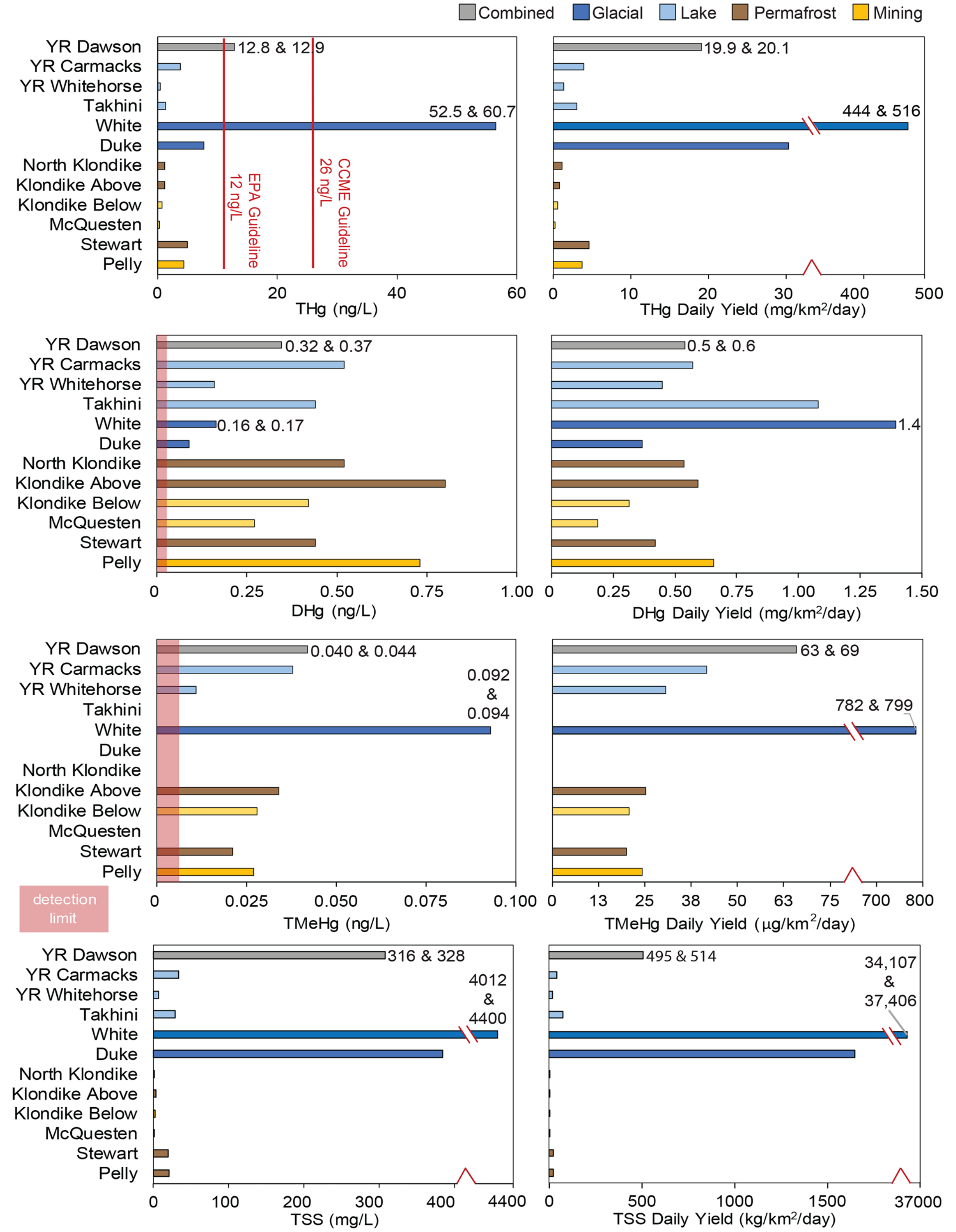


Figure 3. July 2021 concentrations and estimated daily yields of total mercury (THg), dissolved mercury (DHg), total methylmercury (TMeHg), and total suspended sediment (TSS) at 12 sites across the Yukon River basin (note the axis breaks in the THg, TMeHg daily yield plots, and TSS concentration and daily yield plots). Dashed lines indicate total mercury guidelines [26 ng/L, Canadian Council of Ministers of the Environment (CCME), and 12 ng/L, U.S. Environmental Protection Agency (EPA)]. Colors represent dominant landscape types: glacierized (dark blue), large lake (light blue), placer mining impacted (yellow), pristine permafrost (brown), and mixed (gray).

detailed sampling to assess seasonal patterns in mercury cycling across the basin, will be crucial to predicting the future hydrochemical response of the Yukon and other panarctic rivers to cryosphere degradation.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.3c00427.

> Detailed methods; tables of site sampling descriptions and results; the results of duplicate and field blank analyses; historical plus 2021 hydrographs from each site sampled; and historical suspended sediment concentration data for the White River (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Kasia J. Staniszewska − *Department of Earth and Atmospheric Sciences, University of Alberta, Edmonton, Alberta T6G 2E3, Canada;* orcid.org/0000-0002-8906-1744; Email: katarzyn@ualberta.ca

Authors

Alberto V. Reyes − *Department of Earth and Atmospheric Sciences, University of Alberta, Edmonton, Alberta T6G 2E3, Canada;* orcid.org/0000-0002-7838-4214

Colin A. Cooke − *Department of Earth and Atmospheric Sciences, University of Alberta, Edmonton, Alberta T6G 2E3, Canada; Environment and Protected Areas, Government of Alberta, Edmonton, Alberta T5K 2G6, Canada;* orcid.org/0000-0002-7417-5263

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.estlett.3c00427

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The authors thank NSERC (Discovery Grant), the University of Alberta (Northern Research Award), and the Northern Scientific Training Program for funding; Corey Benbow, Elizabeth Hall, and Dr. Marianne Douglas for field assistance; and the kind staff at the University of Alberta Biogeochemical Analytical Services Laboratory. The authors also thank the editor and four anonymous reviewers for thoughtful and constructive comments. The authors are are grateful for permission to conduct field research in the traditional territories of multiple First Nations in the Yukon River basin.

■ REFERENCES

(1) Donaldson, S. G.; Van Oostdam, J.; Tikhonov, C.; Feeley, M.; Armstrong, B.; Ayotte, P.; Boucher, O.; Bowers, W.; Chan, L.; Dallaire, F.; Dallaire, R.; Dewailly, É.; Edwards, J.; Egeland, G. M.; Fontaine, J.; Furgal, C.; Leech, T.; Loring, E.; Muckle, G.; Nancarrow, T.; Pereg, D.; Plusquellec, P.; Potyrala, M.; Receveur, O.; Shearer, R. G. Environmental Contaminants and Human Health in the Canadian Arctic. *Sci. Total Environ.* **2010**, *408*, 5165−5234.

(2) Kirk, J. L.; Lehnherr, I.; Andersson, M.; Braune, B. M.; Chan, L.; Dastoor, A. P.; Durnford, D.; Gleason, A. L.; Loseto, L. L.; Steffen, A.; St. Louis, V. Mercury in Arctic Marine Ecosystems: Sources, Pathways and Exposure. *Environ. Res.* **2012**, *119*, 64−87.

(3) Miner, K. R.; Blais, J.; Bogdal, C.; Villa, S.; Schwikowski, M.; Pavlova, P.; Steinlin, C.; Gerbi, C.; Kreutz, K. J. Legacy Organochlorine Pollutants in Glacial Watersheds: A Review. *Environ. Sci. Process. Impacts* **2017**, *19*, 1474−1483.

(4) Stern, G. A.; Macdonald, R. W.; Outridge, P. M.; Wilson, S.; Chetelat, J.; Cole, A.; Hintelmann, H.; Loseto, L. L.; Steffen, A.; Wang, F.; Zdanowicz, C. How Does Climate Change Influence Arctic Mercury? *Sci. Total Environ.* **2012**, *414*, 22−42.

(5) Zdanowicz, C.; Karlsson, P.; Beckholmen, I.; Roach, P.; Poulain, A.; Yumvihoze, E.; Martma, T.; Ryjkov, A.; Dastoor, A. Snowmelt, Glacial and Atmospheric Sources of Mercury to a Subarctic Mountain Lake Catchment, Yukon, Canada. *Geochim. Cosmochim. Acta* **2018**, *238*, 374−393.

(6) Carling, G.; Tingey, D.; Fernandez, D. Mercury and Other Trace Elements in Glacial Meltwater at Grand Teton National Park, Wyoming. *UW Natl. Park. Serv. Res. Stn. Annu. Reports* **2013**, *36*, 2−9.

(7) Chételat, J.; McKinney, M. A.; Amyot, M.; Dastoor, A.; Douglas, T. A.; Heimbürger-Boavida, L. E.; Kirk, J.; Kahilainen, K. K.; Outridge, P. M.; Pelletier, N.; Skov, H.; St. Pierre, K.; Vuorenmaa, J.; Wang, F. Climate Change and Mercury in the Arctic: Abiotic Interactions. *Sci. Total Environ.* **2022**, *824*, 153715.

(8) St. Pierre, K. A.; St. Louis, V. L.; Schiff, S. L.; Lehnherr, I.; Dainard, P. G.; Gardner, A. S.; Aukes, P. J. K.; Sharp, M. J. Proglacial Freshwaters Are Significant and Previously Unrecognized Sinks of Atmospheric CO2. *Proc. Natl. Acad. Sci. U. S. A.* **2019**, *116*, 17690−17695.

(9) Arctic Monitoring and Assessment Panel. *Assessment 2021: Mercury in the Arctic*; Tromsø, Norway, 2021.

(10) Schuster, P. F.; Schaefer, K. M.; Aiken, G. R.; Antweiler, R. C.; Dewild, J. F.; Gryziec, J. D.; Gusmeroli, A.; Hugelius, G.; Jafarov, E.; Krabbenhoft, D. P.; Liu, L.; Herman-Mercer, N.; Mu, C.; Roth, D. A.; Schaefer, T.; Striegl, R. G.; Wickland, K. P.; Zhang, T. Permafrost Stores a Globally Significant Amount of Mercury. *Geophys. Res. Lett.* **2018**, *45*, 1463−1471.

(11) St. Pierre, K. A.; Zolkos, S.; Shakil, S.; Tank, S. E.; St. Louis, V. L.; Kokelj, S. V. Unprecedented Increases in Total and Methyl Mercury Concentrations Downstream of Retrogressive Thaw Slumps in the Western Canadian Arctic. *Environ. Sci. Technol.* **2018**, *52*, 14099−14109.

(12) Colombo, N.; Salerno, F.; Gruber, S.; Freppaz, M.; Williams, M.; Fratianni, S.; Giardino, M. Review: Impacts of Permafrost Degradation on Inorganic Chemistry of Surface Fresh Water. *Glob. Planet. Change* **2018**, *162*, 69−83.

(13) Schuster, P. F.; Striegl, R. G.; Aiken, G. R.; Krabbenhoft, D. P.; Dewild, J. F.; Butler, K.; Kamark, B. E. N.; Dornblaser, M. Mercury Export from the Yukon River Basin and Potential Response to a Changing Climate. *Environ. Sci. Technol.* **2011**, *45*, 9262−9267.

(14) Schaefer, K.; Elshorbany, Y.; Jafarov, E.; Schuster, P. F.; Striegl, R. G.; Wickland, K. P.; Sunderland, E. M. Potential Impacts of Mercury Released from Thawing Permafrost. *Nat. Commun.* **2020**, *11*, 1−6.

(15) Zolkos, S.; Krabbenhoft, D. P.; Suslova, A.; Tank, S. E.; McClelland, J. W.; Spencer, R. G. M.; Shiklomanov, A.; Zhulidov, A. V.; Gurtovaya, T.; Zimov, N.; Zimov, S.; Mutter, E. A.; Kutny, L.; Amos, E.; Holmes, R. M. Mercury Export from Arctic Great Rivers. *Environ. Sci. Technol.* **2020**, *54*, 4140−4148.

(16) Dastoor, A. P.; Durnford, D. A. Arctic Ocean: Is It a Sink or a Source of Atmospheric Mercury? *Environ. Sci. Technol.* **2014**, *48*, 1707−1717.

(17) Beal, S. A.; Osterberg, E. C.; Zdanowicz, C. M.; Fisher, D. A. Ice Core Perspective on Mercury Pollution during the Past 600 Years. *Environ. Sci. Technol.* **2015**, *49*, 7641−7647.

(18) Wada, T.; Chikita, K.; Kim, Y.; Kudo, I. Glacial Effects on Discharge and Sediment Load in the Subarctic Tanana River Basin, Alaska. *Arctic, Antarct. Alp. Res.* **2011**, *43*, 632−648.

(19) Sonke, J. E.; Teisserenc, R.; Heimbürger-Boavida, L.-E. E.; Petrova, M. V.; Marusczak, N.; Le Dantec, T.; Chupakov, A. V.; Li, C.; Thackray, C. P.; Sunderland, E. M.; Tananaev, N.; Pokrovsky, O. S. Eurasian River Spring Flood Observations Support Net Arctic Ocean Mercury Export to the Atmosphere and Atlantic Ocean. *Proc. Natl. Acad. Sci. U. S. A.* **2018**, *115*, E11586−E11594.

(20) Braune, B.; Chételat, J.; Stow, J.; Tomlinson, S. Special Issue on Mercury in Canada's North. *Sci. Total Environ.* **2015**, *509*−*510*, 1−2.

(21) Brown, T. M.; Macdonald, R. W.; Muir, D. C. G.; Letcher, R. J. The Distribution and Trends of Persistent Organic Pollutants and Mercury in Marine Mammals from Canada's Eastern Arctic. *Sci. Total Environ.* **2018**, *618*, 500−517.

(22) Chesnokova, A.; Baraër, M.; Laperrière-Robillard, T.; Huh, K. Linking Mountain Glacier Retreat and Hydrological Changes in Southwestern Yukon. *Water Resour. Res.* **2020**, *56*, 1−26.

(23) Government of Yukon. Geology maps. https://yukon.ca/en/geology-maps (accessed 2023-03-06).

(24) Brabets, T. P.; Wang, B.; Meade, R. H. *Environmental and Hydrologic Overview of the Yukon River Basin, Alaska and Canada*; Anchorage, AK, 2000; Vol. *99-4204*.

(25) Governemnt of Canada. Faro Mine Remediation Project: Yukon. https://www.rcaanc-cirnac.gc.ca/eng/1480019546952/1537554989037 (accessed 2023-03-06).

(26) Brown, J.; Ferrians, O. J., Jr.; Heginbottom, J. A.; Melinkov, E. S. Circum-Arctic Map of Permafrost and Ground Ice Conditions, 1:10,000,000- Scale Map. 1997.

(27) Public domain image, Wikimedia Commons. http://commons.wikimedia.org/wiki/File:Bw_Yukon_River_drainage_basin.png (accessed 2023-03-06).

(28) U.S. Environmental Protection Agency. Method 1669: Sampling Ambient Water for Trace Metals at EPA Water Quality Criteria Levels. 1996; pp 1−35.

(29) U.S. Environmental Protection Agency. Method 1631, Revision E: Mercury in Water by Oxidation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry. 2002; pp 1−38.

(30) U.S. Environmental Protection Agency. Method 1630: Methyl Mercury in Water by Distillation, Aqueous Ethylation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry. 1998; pp 1−46.

(31) U.S. Environmental Protection Agency. Method 300.1, Revision 1.0: Determination of Inorganic Anions in Drinking Water by Ion Chromatography. 1993; pp 1−39.

(32) U.S. Environmental Protection Agency. Method 415.1 Total Oganic Carbon in Water (Combustion or Oxidation). 1999; pp 1−6.

(33) WMO. State of the Global Climate 2021 - No. 1290. 2022.

(34) Groten, J. T.; Johnson, G. D. Comparability of River Suspended-Sediment Sampling and Laboratory Analysis Methods. *Sci. Invest. Rep. (U. S. Geol. Surv.)* **2018**, 1−23.

(35) The Government of Canada. Historical Sediment Data. https://wateroffice.ec.gc.ca/search/sediment_e.html (accessed 2023-09-15).

(36) Ullrich, S. M.; Tanton, T. W.; Abdrashitova, S. A. Mercury in the Aquatic Environment : A Review of Factors Affecting Methylation. *Environ. Sci. Technol.* **2001**, *31*, 241.

(37) Lavoie, R. A.; Jardine, T. D.; Chumchal, M. M.; Kidd, K. A.; Campbell, L. M. Biomagnification of Mercury in Aquatic Food Webs: A Worldwide Meta-Analysis. *Environ. Sci. Technol.* **2013**, *47*, 13385−13394.

(38) Yukon Geological Survey. Yukon Bedrock Geology Map. http://data.geology.gov.yk.ca/Compilation/3#InfoTab (accessed 2022-08-05).

(39) Dornblaser, M. M.; Striegl, R. G. Suspended Sediment and Carbonate Transport in the Yukon River Basin, Alaska: Fluxes and Potential Future Responses to Climate Change. *Water Resour. Res.* **2009**, *45*, n/a DOI: 10.1029/2008WR007546.

(40) Froese, D. G.; Smith, D. G.; Clement, D. T. Characterizing Large River History with Shallow Geophysics: Middle Yukon River, Yukon Territory and Alaska. *Geomorphology* **2005**, *67*, 391−406.

(41) Danielson, S.; Dobbins, E.; Jakobsson, M.; Johnson, M.; Weingartner, T.; Williams, W.; Zarayskaya, Y. Sounding the Northern Seas. *Eos (Washington, DC).* **2015**, *96*, n/a DOI: 10.1029/2015EO040975.

(42) Zemp, M.; Huss, M.; Thibert, E.; Eckert, N.; McNabb, R.; Huber, J.; Barandun, M.; Machguth, H.; Nussbaumer, S. U.; Gärtner-Roer, I.; Thomson, L.; Paul, F.; Maussion, F.; Kutuzov, S.; Cogley, J. G. Global Glacier Mass Changes and Their Contributions to Sea-Level Rise from 1961 to 2016. *Nature* **2019**, *568*, 382−386.

(43) Vermilyea, A. W.; Nagorski, S. A.; Lamborg, C. H.; Hood, E. W.; Scott, D.; Swarr, G. J. Continuous Proxy Measurements Reveal Large Mercury Fluxes from Glacial and Forested Watersheds in Alaska. *Sci. Total Environ.* **2017**, *599*−*600*, 145−155.

(44) Nagorski, S. A.; Vermilyea, A. W.; Lamborg, C. H. Mercury Export from Glacierized Alaskan Watersheds as Influenced by Bedrock Geology, Watershed Processes, and Atmospheric Deposition. *Geochim. Cosmochim. Acta* **2021**, *304*, 32−49.

(45) Hawkings, J. R.; Linhoff, B. S.; Wadham, J. L.; Stibal, M.; Lamborg, C. H.; Carling, G. T.; Lamarche-Gagnon, G.; Kohler, T. J.; Ward, R.; Hendry, K. R.; Falteisek, L.; Kellerman, A. M.; Cameron, K. A.; Hatton, J. E.; Tingey, S.; Holt, A. D.; Vinšová, P.; Hofer, S.; Bulínová, M.; Větrovský, T.; Meire, L.; Spencer, R. G. M. Large Subglacial Source of Mercury from the Southwestern Margin of the Greenland Ice Sheet. *Nat. Geosci.* **2021**, *14*, 496−502.

(46) Church, M.; Ryder, J. M. Paraglacial Sedimentation: A Consideration of Fluvial Processes Conditioned by Glaciation. *Geol. Soc. Am. Bull.* **1972**, *83*, 3059−3072.

(47) Wedepohl, H. K. The Composition of the Continental Crust. *Geochim. Cosmochim. Acta* **1995**, *59*, 1217−1232.

(48) Yukon Placer Secretariat. *Guidebook of Mitigation Measures for Placer Mining in the Yukon*; 2010. https://yukon.ca/en/guidebook-mitigation-measures-placer-mining-yukon (accessed 2023-06-03).

(49) Chevreux, A.; Clarkson, R. Best Management Practices for Placer Mining in Yukon Wetlands - Recommnedations and Guidelines. 2015. https://www.kpma.ca/wp-content/uploads/2017/07/KPMA-Best-Management-Practices-for-Placer-Mining-in-Yukon-Wetlands.pdf (accessed 2023-09-26).

(50) Fraser, T. A.; Burn, C. R. On the Nature and Origin of "Muck" Deposits in the Klondike Area, Yukon Territory. *Can. J. Earth Sci.* **1997**, *34*, 1333−1344.

(51) Tarbier, B.; Hugelius, G.; Kristina Sannel, A. B.; Baptista-Salazar, C.; Jonsson, S. Permafrost Thaw Increases Methylmercury Formation in Subarctic Fennoscandia. *Environ. Sci. Technol.* **2021**, *55*, 6710−6717.

(52) Branfireun, B. A.; Cosio, C.; Poulain, A. J.; Riise, G.; Bravo, A. G. Mercury Cycling in Freshwater Systems - An Updated Conceptual Model. *Sci. Total Environ.* **2020**, *745*, 140906.

(53) Yukon Placer Secretariat. Water Quality Monitoring Annual Report Klondike Watershed 2016. 2016. https://emr-ftp.gov.yk.ca/emrweb/CMI/Water_Quality/Klondike_River/2016/Klondike_River_Summary_2016.pdf (accessed 2023-09-26).

(54) Bieroza, M.; Acharya, S.; Benisch, J.; ter Borg, R. N.; Hallberg, L.; Negri, C.; Pruitt, A.; Pucher, M.; Saavedra, F.; Staniszewska, K.; van't Veen, S. G. M.; Vincent, A.; Winter, C.; Basu, N. B.; Jarvie, H. P.; Kirchner, J. W. Advances in Catchment Science, Hydrochemistry, and Aquatic Ecology Enabled by High-Frequency Water Quality Measurements. *Environ. Sci. Technol.* **2023**, *57*, 4701−4719.

(55) Olson, C.; Jiskra, M.; Biester, H.; Chow, J.; Obrist, D. Mercury in Active-Layer Tundra Soils of Alaska: Concentrations, Pools, Origins, and Spatial Distribution. *Global Biogeochem. Cycles* **2018**, *32*, 1058−1073.

(56) Aiken, G. R.; Spencer, R. G. M.; Striegl, R. G.; Schuster, P. F.; Raymond, P. A. Global Biogeochemical Cycles in the Yukon River Basin. *Global Biogeochem. Cycles* **2014**, *28*, 525−537.

(57) Staniszewska, K. J.; Reyes, A. V.; Cooke, C. A.; Miller, B. S.; Woywitka, R. J. Permafrost, Geomorphic, and Hydroclimatic Controls on Mercury, Methylmercury, and Lead Concentrations and Exports in Old Crow River, Arctic Western Canada. *Chem. Geol.* **2022**, *596*, No. 120810.

(58) Kokelj, S. V.; Tunnicliffe, J.; Lacelle, D.; Lantz, T. C.; Chin, K. S.; Fraser, R. Increased Precipitation Drives Mega Slump Development and Destabilization of Ice-Rich Permafrost Terrain, Northwestern Canada. *Glob. Planet. Change* **2015**, *129*, 56−68.

(59) Lewkowicz, A. G.; Way, R. G. Extremes of Summer Climate Trigger Thousands of Thermokarst Landslides in a High Arctic Environment. *Nat. Commun.* **2019**, *10*, 1−11.

(60) Runge, A.; Nitze, I.; Grosse, G. Remote Sensing Annual Dynamics of Rapid Permafrost Thaw Disturbances with LandTrendr. *Remote Sens. Environ.* **2022**, *268*, No. 112752.

(61) Kokelj, S. V.; Kokoszka, J.; Van Der Sluijs, J.; Rudy, A. C. A.; Tunnicliffe, J.; Shakil, S.; Tank, S. E.; Zolkos, S. Thaw-Driven Mass Wasting Couples Slopes with Downstream Systems, and Effects Propagate through Arctic Drainage Networks. *Cryosphere* **2021**, *15*, 3059−3081.

(62) Tank, S. E.; Vonk, J. E.; Walvoord, M. A.; McClelland, J. W.; Laurion, I.; Abbott, B. W. Landscape Matters: Predicting the Biogeochemical Effects of Permafrost Thaw on Aquatic Networks with a State Factor Approach. *Permafr. Periglac. Process.* **2020**, *31*, 358−370.

(63) Vonk, J. E.; Tank, S. E.; Walvoord, M. A. Integrating Hydrology and Biogeochemistry across Frozen Landscapes. *Nat. Commun.* **2019**, *10*, 3−6.

(64) Walvoord, M. A.; Kurylyk, B. L. Hydrologic Impacts of Thawing Permafrost-A Review. *Vadose Zone J.* **2016**, *15*, 1.